

Accepted refereed manuscript of:

Varley A, Tyler A, Dowdall M, Bondar Y & Zabrotski V (2017) An in situ method for the high resolution mapping of  $^{137}\text{Cs}$  and estimation of vertical depth penetration in a highly contaminated environment, *Science of the Total Environment*, 605-606, pp. 957-966.

DOI: [10.1016/j.scitotenv.2017.06.067](https://doi.org/10.1016/j.scitotenv.2017.06.067)

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1 **An *In situ* Method for the High Resolution Mapping of <sup>137</sup>Cs and Estimation of Vertical**  
2 **Depth Penetration in a Highly Contaminated Environment.**

3

4 Adam Varley<sup>β</sup>, Andrew Tyler<sup>β</sup>, Mark Dowdall<sup>α</sup>, Yuri Bondar,

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6 <sup>β</sup> Department of. Biological and Environmental Sciences, University of Stirling, Stirling, FK9

7 4LA, United Kingdom

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9 <sup>α</sup> Norwegian Radiation Protection Authority, Grini næringspark 13, 1332 Østerås, Norway

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35 **Abstract**

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37 The Chernobyl nuclear power plant meltdown has to date been the single largest release of  
38 radioactivity into the environment. As a result, radioactive contamination that poses a  
39 significant threat to human health still persists across much of Europe with the highest  
40 concentrations associated with Belarus, Ukraine and western Russia. Of the radionuclides  
41 still prevalent with these territories  $^{137}\text{Cs}$  presents one of the most problematic remediation  
42 challenges. Principally, this is due to the localised spatial and vertical heterogeneity of  
43 contamination within the soil (~10's of meters), thus making it difficult to accurately  
44 characterise through conventional measurement techniques such as static *in situ* gamma-ray  
45 spectrometry or soil cores. Here, a practical solution has been explored, which utilises a large  
46 number of short-count time spectral measurements made using relatively inexpensive,  
47 lightweight, scintillators (sodium iodide and lanthanum bromide). This approach offers the  
48 added advantage of being able to estimate activity and burial depth of  $^{137}\text{Cs}$  contamination in  
49 much higher spatial resolution compared to traditional approaches. During the course of this  
50 work, detectors were calibrated using the Monte Carlo simulations and depth distribution was  
51 estimated using the peak-to-valley ratio. Activity and depth estimates were then compared to  
52 five reference sites characterised using soil cores. Estimates were in good agreement with the  
53 reference sites, differences of ~25 % and ~50 % in total inventory were found for the three  
54 higher and two lower activity sites, respectively. It was concluded that slightly longer count  
55 times would be required for the lower activity (<1 MBq m<sup>-2</sup>) sites. Modelling and reference  
56 site results suggest little advantage would be gained through the use of the substantially more  
57 expensive lanthanum bromide detector over the sodium iodide detector. Finally, the potential  
58 of the approach was demonstrated by mapping one of the sites and its surrounding area in  
59 high spatial resolution.

60

61 **Keywords**

62 **Cs-137, *in situ*, field gamma-ray spectrometry, peak-to-valley, PVT, Chernobyl**

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69 **Highlights**

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71 • Chernobyl-derived  $^{137}\text{Cs}$  poses a risk to human health.

72 • Highly heterogeneous contamination within tens of kilometres of Chernobyl particularly  
73 difficult to characterise.

74 • Numerous short count time gamma-ray measurements can address spatial resolution issues.

75 • Peak-to-valley method provides improved depth and activity estimates.

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77 **1. Introduction**

78  
79 The accident that occurred at the Chernobyl Nuclear Power Plant (ChNPP) in April 1986  
80 resulted in the radioactive contamination of wide areas of Europe and, to a greater extent, the  
81 territories of Belarus, Ukraine and the western Russian Federation. In areas of south Belarus  
82 the deposition exceeded 1500 kBq/m<sup>2</sup> <sup>137</sup>Cs, 3 orders of magnitude greater than that which  
83 resulted from atmospheric nuclear weapons as of 1986 (UNSCEAR, 2000). As a result of the  
84 accident, a total area exceeding 60,000 km<sup>2</sup> of forested land was contaminated, including over  
85 20,000 km<sup>2</sup> of the Gomel and Mogilev regions of Belarus in addition to areas near Kiev and in  
86 the Russian Federations' Bryansk region. The primary contaminants as a result of the accident  
87 were <sup>137</sup>Cs, <sup>90</sup>Sr and <sup>239</sup>Pu. Highest levels of contamination, in excess of 1.4 MBq/m<sup>2</sup> of <sup>137</sup>Cs  
88 occur with the southernmost parts of the reserves territory over lands nearest the Chernobyl  
89 reactor while localised areas of high contamination can be found distributed throughout the  
90 reserve. Sr-90 is present at densities up to 3 MBq/m<sup>2</sup> while <sup>241</sup>Am levels have approximately  
91 doubled over the past twenty years and now approach 0.1 MBq/m<sup>2</sup> in places as a result of  
92 ingrowth from <sup>241</sup>Pu (Smith and Beresford, 2002). Actinides such as <sup>238</sup>Pu and <sup>239,240</sup>Pu are  
93 present at levels up to 0.037 MBq/m<sup>2</sup> and 0.074 MBq/m<sup>2</sup> respectively. The Polessie State  
94 Radioecology Reserve (PSRER) in southern Belarus is one of the most contaminated regions  
95 with associated total inventory estimates of 2.5 x 10<sup>9</sup> MBq of <sup>137</sup>Cs and 0.18 x 10<sup>9</sup> MBq of  
96 <sup>90</sup>Sr (Izrael and Bogdevich, 2009). Contamination is spatially highly heterogeneous over  
97 distances of 10's of meters or less while significant contamination is also found in the tree  
98 canopy and vegetation and in aquatic systems of the area. A general overview of <sup>137</sup>Cs levels  
99 in the PSRER is presented in Figure 1.

100

101 .....Insertion point for Figure 1.....

102

103 ***Figure 1. Spatial distribution of the estimated <sup>137</sup>Cs inventory (kBq m<sup>2</sup>) across the Polessie***  
104 ***State Radioecology Reserve***

105 Cs-137 is usually considered to be the most important long-term anthropogenic contributor to  
106 the dose-levels in the environment (Miller, 2007). Its radiological prominence is due to its  
107 relatively long half-life of 32 years, its potential to be remobilised through sediment  
108 redistribution as it binds strongly to clays and its affinity to be taken in by living tissue given  
109 that it readily displaces biological potassium within the body (Povinec et al., 2003). These

110 factors, amongst others, impart a certain emphasis on the monitoring of levels of this isotope  
111 within the environment and such monitoring is especially important in highly contaminated  
112 areas such as those of the PSRER where considerable efforts have been expended in  
113 establishing the spatial distribution and environmental behaviour of this and other nuclides  
114 (see for example Kudelsky, et al., 1996; Knatko et al., 1996; Guermentchuk et al., 1997;  
115 Sokolik et al., 2001; Yoschenko et al., 2006).

116  
117 Monitoring of  $^{137}\text{Cs}$  in soils is conventionally achieved by either of two approaches. The first  
118 consists of extracting discrete samples or cores from a site and measuring the activity of either  
119 the whole sample or of individually sliced samples representative of different depths using  
120 laboratory based gamma-ray spectrometers (Tyler, 2008). Although precise information as to  
121 the  $^{137}\text{Cs}$  depth penetration can be obtained using this method, the resources it takes to obtain,  
122 prepare and count individual samples results in spatial coverage being limited and spatial  
123 heterogeneities may not always be captured (Tyler et al., 1996a). Estimates of  $^{137}\text{Cs}$   
124 penetration down the soil column are particularly important for two reasons. Firstly,  
125 knowledge of depth distribution can contribute greatly to remediation strategies and decisions  
126 on subsequent site use. Secondly, depth distribution can influence the environmental  
127 behaviour of  $^{137}\text{Cs}$  particularly with regards to surface remobilisation.

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129 The environment of the PSRER constitutes an interesting problem with respect to how high  
130 resolution mapping of contaminant  $^{137}\text{Cs}$  can be conducted. The territories not covered by  
131 forest (basically abandoned agricultural lands) constitute 38.0 % and non-agricultural  
132 unforested lands occupy 9.3 %. Road coverage is sparse and the terrain is generally rugged.  
133 While topographically flat, the area has local depressions and elevations as well as a variety  
134 of watercourses and bodies (both natural and man-made) in addition to extensive seasonally  
135 flooded areas. In general, much of the area is inaccessible by vehicle and can be considered as  
136 difficult terrain. In light of the above considerations, the development of a man portable  
137 gamma-ray spectrometry system capable of accurate high spatial resolution  $^{137}\text{Cs}$  mapping of  
138 contamination patterns is paramount (Plamboeck et al., 2006). Within the PSRER, due to  
139 spatial heterogeneity  $^{137}\text{Cs}$  activity the capability to map changes over metres to tens of metres  
140 would provide significant improvement in spatial resolution compared to current approaches.

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144 1.1. *In Situ Gamma-ray Spectrometry of <sup>137</sup>Cs and Depth Estimation.*

145 The fundamentals of gamma-ray spectrometry can be found elsewhere, for example Knoll  
146 (2010). The principle of *in situ* gamma-ray spectrometry is the placing of a suitable  
147 instrument above the ground and making a measurement over a specified time period. The  
148 resultant measurement offers information as to the localised radiation field through the  
149 differential measurement of gamma-rays that interact with the active volume of the detector.  
150 Critically, the energies of individual gamma-rays are recorded and accumulated into a  
151 differential energy spectrum. Estimating the <sup>137</sup>Cs inventory (Bq m<sup>-2</sup>) directly from the count  
152 rate under the characteristic <sup>137</sup>Cs 662 keV photopeak is not always a straightforward task  
153 given that a number of factors can significantly change the count rate. Ground curvature  
154 (Laedermann et al., 1998) and the influence of trees and other vegetation (Gering et al., 2002)  
155 can introduce a considerable amount of uncertainty. However, the primary source of  
156 uncertainty often arises when significant changes in depth distribution are encountered, as  
157 considerable variations can alter the count rate by orders of magnitude (Sowa et al., 1989).  
158 Besides altering the count rate, changes in depth distribution can also change the detector's  
159 Field of View (FoV) and can have an influence on the spatial response of single  
160 measurements. The FoV of a detector can be defined as the effective volume of soil being  
161 sampled and is often characterised by the lateral distance or radius from the detector over  
162 which the sampling area extends to (Tyler et al., 1996a). Problems can arise when  
163 contamination is highly associated with the soil surface, following fresh deposition, reducing  
164 the detectors ability to resolve small scale localised heterogeneity given that the FoV for a  
165 detector at 1 m above the ground is greater than 70 m radius. Moreover, at this scale  
166 topographic anomalies become important in defining the FoV. Nevertheless, Chernobyl  
167 deposition has aged over 30 years and much of the contamination is understood to have  
168 penetrated sufficiently into the soil column (Smith and Beresford, 2002). This process should  
169 have led to reduction in the FoV for a detector at 1m above the ground to approximately less  
170 than 10 m (Tyler et al., 1996a). A further way to reduce the FoV is to operate detectors at a  
171 nominal height of 0.1 m, which should reduce the FoV down to between 2 to 5 m.

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173 Importantly, prior to any inventory estimations the count rate must be corrected for the  
174 appropriate depth distribution and the influence of the FoV must also be considered prior to  
175 interpolation mapping. The peak-to-valley ratio (PTV) provides a means of performing this  
176 task through spectral information alone (Zombori et al., 1992). PTV has been widely used to

177 estimate the burial depth distribution of  $^{137}\text{Cs}$  and improve the overall accuracy of inventory  
178 estimations (Feng et al., 2012; Gering et al., 1998; Hillmann et al., 1996; Tyler, 2004). Tyler  
179 et al (1996b) puts forward a particularly visual description of the physical processes that the  
180 PTV method is developed upon. In summary, the PTV is the ratio between the number of full  
181 energy and forward scattered photons (Figure 2). Forward scattered photons are essentially  
182 photons that have lost a small amount of energy whilst exiting the soil and can be located in  
183 the “valley” region of the spectrum between the full energy peak at 662 keV and the Compton  
184 edge at 480 keV (Figure 2b). Importantly, the greater the depth at which a source is present  
185 (Figure 2a), the higher is the probability of photons undergoing forward scattering and  
186 occupying the valley region, leading to an increase in valley height and a decrease in the PTV  
187 (Figure 2c).

188

189 The PTV method can be applied to any detector of adequate energy resolution (Kastlander  
190 and Bargholtz, 2005; Tyler, 2004). At present, two of the most commercially widely viable  
191 available scintillators that are eminently suitable for portable field operations and that offer  
192 high enough energy resolution to measure  $^{137}\text{Cs}$  are sodium iodide (NaI:Tl) and lanthanum  
193 bromide (LaBr:Ce) (Guss et al., 2010). Of these two, NaI:Tl has been the workhorse of *in*  
194 *situ* and mobile gamma-ray spectrometry for a number a decades as it is relatively cheap,  
195 robust and provides acceptable energy resolution (~7% at 662 keV). LaBr:Ce on the other  
196 hand is a relatively novel scintillator providing superior energy resolution (2.5% at 662 keV)  
197 and higher energy efficiency due to its higher density ( $5.1 \text{ g cm}^{-3}$ ) compared to NaI:Tl ( $3.7 \text{ g}$   
198  $\text{cm}^{-3}$ ). LaBr:Ce does contain an intrinsic background component that can deteriorate the  
199 signal to noise ratio especially at low count rates (Nilsson et al., 2014). Another key  
200 limitation of LaBr:Ce is that it costs almost an order of magnitude significantly more than  
201 NaI:Tl. There have been a relatively large number of studies comparing the performance of  
202 NaI:Tl and LaBr:Ce through theoretical or experimental means (Guss et al., 2010; Iltis et al.,  
203 2006; Milbrath et al., 2007). Nonetheless, limited comparisons have been undertaken in the  
204 field (Duval and Arnold, 2013; Nilsson, 2010) and furthermore only a subset of these have  
205 pertained to the measurement of  $^{137}\text{Cs}$  (Nilsson et al., 2014).

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207 .....Insertion Point for Figure 2.....

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209 **Figure 2. The relationship between changes in depth distribution ( $\beta$ ) and the peak-to-valley**  
210 **ratio observed in a  $71 \times 71 \text{ mm}$  sodium iodide detector. A) Normalised depth distributions**



211 *for the same inventory ; B) Peak normalised spectra for different  $\beta$  values demonstrating*  
212 *the relative increase in valley counts with increasing burial; C) A straight-forward*  
213 *regression between calibrated peak-to-valley ratio and  $\beta$  value.*

214 The overarching aim of this investigation is to compare NaI:Tl and LaBr:Ce detector systems  
215 for the purposes of characterising spatial  $^{137}\text{Cs}$  contamination including its vertical depth  
216 distribution at high spatial resolution using one second acquisition times within the PSRER.

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## 220 2. Methodology

### 221 2.1. Field Sites.

222 The selected sites within the PSRER were topographically flat over areas of 50 m in diameter,  
223 the centre of each site being marked with a square area of the size 100 m<sup>2</sup>. Each site was  
224 specifically chosen to be as homogeneous as possible. As a general indicator of the  
225 conditions of the site, the gamma dose rate was recorded with an AT6130 Radiation Monitor  
226 (Scientific and Production Enterprise ATOMTEX, Belarus) at each corner and in the centre at  
227 a height of 1 m and soil samples were taken at the same points. The location and ambient dose  
228 rates over the five sites were as follows: Site 1, a soddy podzolic soil at N 51°33'07,9" E  
229 029°55'26,1" (2.21 to 2.50 µSv/h); Site 2, a sandy soil at N 51°33'17,1" E 029°55'12,1", (0.29  
230 to 0.35 µSv/h); Site 3, an organic peat soil at N 51°32'54,7" E 029°55'52,2" (1.55 to 1.73  
231 µSv/h); Site 4, a regularly flooded, mineral floodplain soil at N 51°31'45,2" E 029°56'07,5"  
232 (0.52 to 0.89 µSv/h); Site 5, a mineral, ploughed soil at N 51°47'11,8" E 030°01'16,8" (0.25  
233 to 0.39 µSv/h). A standard cylindrical corer of 4 cm in diameter and 20 cm extent was used to  
234 take cores, each divided into the following increments: 0-3 cm, 3-6 cm, 6-10 cm, 10-15 cm,  
235 15-20 cm. All layers of the same depth from the five cores were bulked and homogenized for  
236 gamma-spectrometry. The preparation of soil samples for the radioanalytical measurements  
237 included air drying and homogenization by mixing and sieving (2 mm aperture). After  
238 weighing, a subsample was transferred to the analytical geometry for counting. A HPGe  
239 (Canberra, Be5030, carbon fibre window, 50% rel. eff.) detector was used for <sup>137</sup>Cs  
240 determination. Count times varied from 6000 to 80000 sec. Statistical uncertainties for the  
241 photo peak of <sup>137</sup>Cs were less than 5 %. Details as to <sup>137</sup>Cs contamination levels at the five test  
242 sites are provided in Table 1. For further site information and detailed photographs of the sites  
243 refer to Dowdall et al., (2015).

244

### 245 2.2. Gamma-ray Spectrometry System

246 A 71 (Ø) × 71 mm NaI:Tl and 71 (Ø) × 71 mm LaBr:Ce detectors (both made by Saint  
247 Gobain) were connected to individual Ortec digiBases recording a 1024 channel spectral  
248 every second using Ortec's Maestro software (ORTEC, 2005). Accompanying each spectrum,  
249 GPS coordinates were recorded using an SX Blue II differential GPS, with a resolution of 0.6  
250 m. The software used to combine spectral measurements and spatial coordinates was built

251 within the JAVA framework and was able to provide real-time feedback on count rates in  
252 specified windows. To avoid significant spectral drift occurring, Maestro's gain stabilisation  
253 software was operated on the 662 keV peak. In order to capture local inhomogeneities, the  
254 detectors were to the operators best abilities held at a constant height of 0.1 m from the  
255 ground at all times to narrow the field of view. This was expected to deviate by 0.1 m either  
256 way. The shape of the spectrum was assumed not to change significantly therefore, only the  
257 count rate was altered according to the changing field of view and calculated through a  
258 deterministic model; this was built in as an uncertainty following Monte Carlo simulations. A  
259 walking speed of approximately 0.5 m s<sup>-1</sup> and transect spacing of 0.5 to 1 m was maintained  
260 throughout the surveys. Approximately 10 m outside of the test site area were measured in  
261 addition to the areas inside the test sites.

262

### 263 *2.3. Monte Carlo Simulations*

264 The individual instruments were calibrated using Monte Carlo Simulations (MCS). MCS  
265 were preferred over analytical calibration given that the relationship between PTV and  
266 changes in <sup>137</sup>Cs depth distribution could be modelled with greater precision compared to a  
267 limited number of soil core data (Likar et al., 2000). MCS are widely used to estimate  
268 detector responses as radiation transport is probabilistic by nature and very difficult to resolve  
269 use deterministic approaches (Stromswold, 1995). Hence, by tracking individual particles,  
270 through repeated random sampling of well-defined probability distributions a complex  
271 problem can be addressed (Maučec et al., 2009).

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Soil characteristics	Layer	<sup>137</sup> Cs Bq/kg	% of Total <sup>137</sup> Cs Inventory
Soddy podzolic	Site 1		
	0-3 cm	41450+/-8290	66.1+/-13.2
	3-6 cm	15350+/-3070	28.8+/-5.8
	6-10 cm	1360+/-270	3.6+/-0.7
	0-15 cm	314+/-72	1.1+/-0.3
	15-20 cm	92+/-27	0.3+/-0.1
Bulk density / <sup>137</sup> Cs Inventory			1.62 g/cm <sup>3</sup> / 2414 kBq/m <sup>2</sup>
Sandy	Site 2		
	0-3 cm	876+/-175	24.0+/-4.8
	3-6 cm	814+/-163	19.4+/-3.9
	6-10 cm	693+/-139	22.6+/-4.5
	0-15 cm	813+/-163	33.4+/-6.7
	15-20 cm	16+/-8	0.6+/-0.3
Bulk density / <sup>137</sup> Cs Inventory			1.77 g/cm <sup>3</sup> / 215 kBq/m <sup>2</sup>
Organic peat	Site 3		
	0-3 cm	33380+/-6675	38.7+/-7.7
	3-6 cm	16000+/-3200	30.1+/-6.0
	6-10 cm	5910+/-1180	17.3+/-3.5
	0-15 cm	2740+/-550	10.0+/-2.0
	15-20 cm	947+/-189	3.9+/-0.8
Bulk density / <sup>137</sup> Cs Inventory			1.15 g/cm <sup>3</sup> / 1672 kBq/m <sup>2</sup>
Mineral floodplain	Site 4		
	0-3 cm	41810+/-8360	56.6+/-11.3
	3-6 cm	8665+/-1733	22.2+/-4.4
	6-10 cm	3076+/-615	13.4+/-2.7
	0-15 cm	940+/-189	5.9+/-1.2
	15-20 cm	292+/-65	1.8+/-0.4
Bulk density / <sup>137</sup> Cs Inventory			1.23 g/cm <sup>3</sup> / 1178 kBq/m <sup>2</sup>
Ploughed mineral	Site 5		
	0-3 cm	2114+/-423	17.8+/-3.6
	3-6 cm	2147+/-435	15.3+/-3.1
	6-10 cm	2164+/-433	20.6+/-4.1
	0-15 cm	2235+/-447	24.9+/-5.0
	15-20 cm	1814+/-362	21.5+/-4.3
Bulk density / <sup>137</sup> Cs Inventory			1.55 g/cm <sup>3</sup> / 644 kBq/m <sup>2</sup>

288 The software package Monte Carlo N-Particle 5 (MCNP5) was used to obtain spectral  
289 responses to recover PTV (Briesmeister, 1993). In individual MCS, a basic geometry  
290 encompassing the detector, soil column and air filled with constituent materials were  
291 described through input cards (Figure 3).

292

293 .....*Insertion Point for Figure 3*.....

294

295 **Figure 3. MCNP setup and method used to derive PTV from an exponential distributed**  
296 **source.**

297

298 Descriptions of the starting source were also given to MCNP5, alongside F8 tally instructions  
299 that captured the differential energy spectrum within the simulated detector's active volume.  
300 Given that the PTV is generated using energies considerably higher than those significantly  
301 affected by the photoelectric effect and backscatter (>350 keV), Beck et al. (1972) a standard  
302 soil composition was used. All nuclide decay data was acquired from the National Nuclear  
303 Data Centre (2013).

304

305 Validated detector geometries utilised in previous studies (Varley et al., 2015a) were used to  
306 derive spectral responses for 71 (Ø) × 71 mm NaI:Tl and LaBr:Ce detectors. Varley et al.,  
307 (2015a) reported good agreement between MCS and experimental benchmark results  
308 collected from doped-concrete calibration pads (1 m<sup>2</sup>). The code making up the detector  
309 geometries was modelled on manufactures specification provided by Saint Gobain along with  
310 receipt of detectors. Detector models encompassed the active volume, its outer casing, a  
311 simplified photomultiplier tube and the PVC plastic piping used to house the detection unit  
312 (Figure 3). The simulated contamination was an unbiased source of 0.662 MeV photons and  
313 the geometry was truncated just above the detector where beyond this point all particles were  
314 killed to maximise computer time. No specific reduction variance techniques were used such  
315 as Russian roulette.

316

317 To optimise the geometry of the simulation, adjoint calculations were utilised to establish the  
318 necessary limits or effective FoV of the detector to the source (Varley et al., 2015b). A  
319 conservative extent for the surface layer (0-0.01 m) was adopted (25 m) and successively  
320 deeper layers were established in the same manner. A Gaussian function was fitted to the

321 adjoint results that included 99.9 % of the total photons. Importantly, MNCP5 does not take  
322 into consideration pile-up effects in the electronics of the scintillator for instance when chance  
323 coincidence is encountered (Lindstrom and Fleming, 1995). This phenomenon within  
324 environmental spectra can significantly alter the shape of the spectrum at high count rates  
325 leading to a smearing of the spectrum and importantly removing counts from the full energy  
326 peak to a higher energy channel when combined with lower energy photons (Mowlavi and  
327 Hadizadeh Yazdi, 2011). Pile-up can be modelled using a Monte Carlo regime if the spectrum  
328 is accurate enough across all energies. However, it was accepted that the lower energy part of  
329 the spectrum could not be modelled accurately due to uncertainties associated with the  
330 photoelectric effect which is driven by the chemical composition of the soil. Therefore no  
331 attempt was made to model this occurrence through Monte Carlo methods. Yet, full energy  
332 photon losses in the field were not expected to exceed 0.5 % even at the highest count rates  
333 observed. This was established by monitoring the smearing of photons to higher energies.  
334

335 To achieve spectral responses to a variety of depth distributions and inventories, MCS were  
336 simulated at discrete 10 mm depth intervals down to a depth of 70 g cm<sup>-2</sup> (Figure 3). Beyond  
337 70 g cm<sup>-2</sup> it was found that contributions to PTV were infinitesimal even for large  $\beta$  values.  
338 To form the spectral response for a given value of  $\beta$  and inventory, spectral contributions  
339 from each 10 mm slice were weighted in accordance with the appropriate exponential  
340 function value (see Eqn [1]). This approach not only permitted the PTV to be defined at a  
341 higher depth resolution but also improved counting statistics for later regression models. The  
342 reason for this is that the maximum number of particles in MCNP5 ( $2 \times 10^9$ ) could be run in  
343 each simulation effectively increasing source density without repeated random number  
344 sampling (Briesmeister, 1993).  
345

346 Environmental spectra obtained from the test sites contained contributions from the natural  
347 radioelements: <sup>40</sup>K and the <sup>238</sup>U and <sup>232</sup>Th series. Although spectral inputs from the natural  
348 nuclides were small in comparison to <sup>137</sup>Cs, contributions to the valley region were found to  
349 influence the PTV particularly at low <sup>137</sup>Cs photon fluxes. Therefore, straightforward uniform  
350 distributions for the natural radioelements were included as separate MCS (Thummerer and  
351 Jacob, 1998).  
352  
353  
354

355 2.4. Vertical Distribution of <sup>137</sup>Cs

356 In the first decade or so following Chernobyl, many early studies that used field gamma-ray  
357 spectrometry to measure <sup>137</sup>Cs at undisturbed sites tended to adopt an exponential function,  
358 whereby the activity of contamination was assumed to decrease exponentially with increasing  
359 depth as the heavily concentrated <sup>137</sup>Cs source on the surface gradually penetrated into the soil  
360 column (Hillmann et al., 1996). A number of studies have confirmed this in Belarus and  
361 PSRER (Askbrant et al., 1996; Ivanov et al., 1997) However, more recent observations from  
362 other areas of Europe have shown the exponential function to break down as the source  
363 further penetrates into the soil column and fresh relatively uncontaminated soil buries  
364 contamination to form a subsurface maximum (Almgren and Isaksson, 2006; Bernhardsson et  
365 al., 2015). However, the most recent study conducted in the PSRER suggests that the  
366 exponential function is perhaps still the most appropriate (Dowdall et al., 2017). There reason  
367 are three reasons that might explain this outcome. Firstly, the PSRER in comparison to other  
368 parts of Europe, where many of the aforementioned studies were conducted, receives  
369 relatively low annual rainfall possibly limiting penetration. Secondly, the primary mode of  
370 contamination deposition at the time of the accident was dry deposition rather than wet  
371 deposition reducing initial infiltration (Drozdovitch et al., 2013). Finally, within a 30 km  
372 radius of the ChNPP, radiocaesium was thought to be highly associated with fuel fragments  
373 significantly affecting its physiochemical form and ultimately its mobility (Smith and  
374 Beresford, 2002).

375

376 These facts combined with the knowledge that the PSRER has not been disturbed to any great  
377 extent in over 30 years, therefore justify an exponential decay function to be used in the  
378 modelling of the <sup>137</sup>Cs depth distribution in this environment (ICRU, 1994). Wherein the  
379 activity ( $A_m$ ) (Bq kg<sup>-1</sup>) of the soil changes with mass per unit area ( $\zeta$ ) (g cm<sup>-2</sup>) (Eqn [1]).

380

$$381 \quad A_m(\zeta) = A_{m,0} \cdot e^{(-\zeta/\beta)} \quad [1]$$

382

383 Notice, mass per unit area is preferred over physical depth (cm) as it takes into account  
384 density ( $\rho$ ), which generates a better approximation of shielding and thus permits inventory  
385 (Bq m<sup>-2</sup>) to be calculated more accurately (Eqn [1]). The relaxation mass per unit area ( $\beta$ )  
386 relates the rate at which the surface activity ( $A_0$ ) decreases with mass depth. Resultantly,  $\beta$

387 values approaching 0 will define surface contamination whilst uniform contamination will  
388 approach  $\infty$ .

389

### 390 *2.5. Peak and Valley Calculation*

391 The first step in calculation of the PTV was identification of a peak within the correct area of  
392 a spectrum obtained in the field. The gain stabilisation software, in the vast majority of cases,  
393 centred the 662 keV peak in the correct channel, however small amounts of spectral drift  
394 occurred during the survey. To ensure the peak (NaI:Tl = 586.8-737.2 keV; LaBr:Ce = 618.3-  
395 705.7 keV), valley (NaI:Tl = 496.5-583.8 keV; LaBr:Ce = 498.7-615.3 keV) and background  
396 (NaI:Tl = 740.2-755.3 ; LaBr:Ce = 708.7-726.8 keV) region of interests remained the same  
397 between spectra, a Gaussian function was fitted to the peak, the maximum of which could  
398 then act as a more precise energy calibration. Thereafter, the background under the peak was  
399 estimating using a straightforward linear model fitted to five channels either side of the base  
400 of the peak. The background was then subtracted and a second Gaussian function was fitted  
401 and integrated to calculate the area. The valley height was calculated by subtracting the area  
402 to the high energy side of the peak from the valley region (Tyler, 2004). This routine was  
403 written using the base features of the R statistical package (R Core Development Team,  
404 2016).

405

### 406 *2.6. Data Generation*

407 For each detector a calibration dataset was generated, comprised of 1000 spectral responses,  
408 derived through randomly sampling input  $\beta$  and inventory parameters and applying them to  
409 the MCS dataset. The calibration dataset provided spectra with relatively low uncertainty to  
410 facilitate fitting of statistical models. An independent dataset (of 2000 spectral responses) was  
411 also formed. This dataset, known as the cross-validation dataset, provided a means of  
412 assessing performance through Root Mean Square Error (RMSE) for  $\beta$  and inventory  
413 estimates. The same modelling constraints applied to the calibration dataset were used to form  
414 the cross-validation dataset, but with the addition of the natural radionuclides ( $^{40}\text{K}$ ,  $^{238}\text{U}$  and  
415  $^{232}\text{Th}$  series) and Poisson noise generated from the one second counts. A similar  
416 bootstrapping subroutine of MCS has proven valuable in previous studies to sample both  
417 background and source populations when *a priori* knowledge is not available (Varley et al.,  
418 2015b).



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Model parameters (Table 2) were chosen to be representative of the variation that could take place within the PSRER environment. Simulated inventories were markedly higher than those at the five test sites, but it was deemed necessary to develop an approach that could be used for higher deposition densities. Additionally, a maximum  $\beta$  value of 100 was chosen as it was discovered through preliminary investigations that larger values contributed negligible counts into the valley regions and would be almost impossible to differentiate. In any case,  $\beta$  values above 100 make very little difference to final inventory estimations and will have negligible contribution if dose rate calculations are performed (ICRU, 1994).

Table 2. Parameters used to construct a synthetic dataset.

Parameter	Minimum	Mean	Median	Maximum
$^{137}\text{Cs}$ inventory ( $\text{kBq m}^{-2}$ )	7.2	2479.3	282	86962.2
$\beta$ ( $\text{g cm}^{-2}$ )	0.13	18.3	7.6	100.0
$^{40}\text{K}$ inventory ( $\text{kBq m}^{-2}$ )	28.2	226.7	222.6	565.2
$^{238}\text{U}$ inventory ( $\text{kBq m}^{-2}$ )	7.1	17.5	16.7	42.39
$^{232}\text{Th}$ inventory ( $\text{kBq m}^{-2}$ )	7.1	19.3	18.4	42.4

431  
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2.7. Calibration Curves

433 To establish appropriate calibration curves to estimate  $\beta$  and inventory for each detector, a  
434 linear model was fitted to the respective calibration datasets through a conventional least-  
435 squares fitting regime (Crawley, 2012). The relationship between explanatory and response  
436 variables was found to be log-linear whereby both response ( $\beta$ ) and explanatory (PTV)  
437 variables were logarithmic. Formulation of the depth model was performed by regressing  
438 PTV against  $\beta$ , where  $\beta_0$  is the point at which PTV is equal to zero and  $\beta_s$  is the slope factor  
439 applied to PTV (Eqn [1])

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$$\log(\beta) = \beta_0 + \beta_s \cdot \log(\text{PTV}) \quad [2]$$

443 The inventory model was estimated by calculating the calibration coefficient  $C$ , by dividing  
444 the inventory ( $A$ ) by the number of counts in the full energy peak ( $N_p$ ) (Eqn [3]).  $C$  could then  
445 be regressed against the PTV to generate the intercept ( $C_0$ ) and the slope factor ( $C_s$ ) (Eqn [4]).

446

$$447 \quad C = A/N_p \quad [3]$$

448

$$449 \quad \log(C) = C_0 + C_s \cdot (PTV) \quad [4]$$

450

451 Applied to field estimates the PTV can be used to derive  $C$ , which need only be multiplied by  
452  $N_p$  to estimate inventory (Tyler et al., 1996b).

453

#### 454 *2.8. Detector-core Comparisons*

455 To develop a firm basis for statistical comparison with the soil core results, all spectra  
456 obtained inside the sites were isolated using a georeferenced polygon, run through the  
457 calibrated models and then aggregated to form a median and interquartile ranges. On average  
458 approximately 600 spectra were collected at each site. Median and interquartile ranges were  
459 preferred over the mean and standard deviation as estimates were found to be highly skewed.  
460 Core inventory estimates were the sum of the total activity of the core multiplied by the  
461 reciprocal of the total core diameter ( $m^2$ ). Uncertainties were principally based on counting  
462 uncertainty. Derivation of  $\beta$  for core data was performed by using a least-squares fitting  
463 regime on the function described in Eqn [1]. Uncertainties were derived by Monte Carlo  
464 simulation.

465

466 **3. Results and Discussion**

467 *3.1. Statistical Modelling of Monte Carlo Simulations*

468 Statistically significant relationships ( $R^2$  values greater than 0.93;  $p < 0.05$ ) were found for all  
 469 models derived from the calibration datasets (Table 3). Model parameter values are similar  
 470 between detectors, suggesting that the detectors respond in a similar manner to an extended  
 471 environmental  $^{137}\text{Cs}$  source.

472

473 Table 3. Parameter and  $R^2$  values for depth and inventory models for lanthanum bromide and  
 474 sodium iodide detectors.

Detector	$B_0$	Depth model			Inventory model			
		$B_s$	$R^2$	$RMSE$	$C_0$	$C_s$	$R^2$	$RMSE$
LaBr:Ce	1579.71	0.97	0.93	19.32	10.23	0.98	0.98	407.21
NaI:Tl	1423.46	0.97	0.94	22.04	11.92	0.98	0.99	483.85

475

476

477 Based on RMSE values on validation datasets in which Poisson noise and natural background  
 478 were introduced, LaBr:Ce (407.21 kBq m<sup>-2</sup> and 19.32 g cm<sup>-2</sup>) marginally outperformed NaI:Tl  
 479 (483.85 kBq m<sup>-2</sup> and 22.04 g cm<sup>-2</sup>) (Table 3). LaBr:Ce would at this stage appear to offer  
 480 increased performance. Nonetheless, to investigate the response of each detector further the  
 481 average uncertainty made on inventory and  $\beta$  estimates, as a function inventory, has been  
 482 presented (Figure 4).

483

484 .....Insertion Point for Figure 4.....

485

486 **Figure 4. A) Percentage error associated with inventory estimates as a function of**  
 487 **inventory for: lanthanum bromide (purely based on peak counts), lanthanum bromide**  
 488 **(peak-to-valley corrected) and sodium iodide (peak-to-valley corrected) B) Percentage error**  
 489 **associated with  $\beta$  estimates as a function of inventory for lanthanum bromide (peak-to-**  
 490 **valley corrected) and sodium iodide (peak-to-valley corrected).**

491  
492 Firstly, the advantage of using PTV corrected data can be realised when compared to  
493 uncorrected data derived from peak counts (Figure 4A). At inventories greater than 100 kBq  
494 m<sup>-2</sup>, there is clear divergence in the percentage error on inventory estimates between corrected  
495 and uncorrected LaBr:Ce results, which becomes significantly larger as inventory increases.  
496 This occurrence can be explained through counting statistics. For example, at low peak count  
497 rates (typically of the order of a few hundred) the uncertainty associated with the calculation  
498 of PTV is large enough that it does not aid in the estimation of inventory; the highest source  
499 of uncertainty in this instance will be associated with the valley height calculation. However,  
500 as count rates rise with increasing contamination, and the valley calculation becomes more  
501 reliable, the PTV correction significantly decreases the percentage error associated with  
502 inventory estimation.

503  
504 Reinforcing the RMSE values (Table 3), corrected NaI:Tl and LaBr:Ce would appear to  
505 produce similar percentage error across the inventory range (Figure 4A). Yet, at inventories  
506 less than ~1500 kBq m<sup>-2</sup>, NaI:Tl exhibits an advantage of ca. 5% lower percentage error than  
507 LaBr:Ce. Above ~1500 kBq m<sup>-2</sup>, LaBr:Ce would appear to improve on NaI:Tl by  
508 approximately 1%. This fact is associated with the intrinsic background contained with the  
509 LaBr:Ce active volume. Therefore, at lower count rates, the signal to noise ratio is lower than  
510 that of NaI:Tl, nonetheless, when count rates exceed a roughly thousand or more the  
511 improved energy resolution deems it superior. A similar aspect is observed when analysing  
512 percentage error associated with  $\beta$  estimates as a function of inventory (Figure 4B). Note,  
513 there is a similar crossover at approximately ~1000 kBq m<sup>-2</sup>, but in this case the advantage  
514 above this value is clearer for the LaBr:Ce detector (~10%). Interestingly, percentage error  
515 on  $\beta$  values are seen to plateau for both detectors at densities around 4000 kBq m<sup>-2</sup>. This  
516 possibly represents the absolute limit of detection where very little improvement on depth  
517 estimate can be made using the devices, this is due to the sensitivity of the ratio at greater  
518 values of  $\beta$ .

### 519 520 *3.2. Application to the Reference Sites*

521 Core results confirmed that there was a large variation in both the depth distribution and total  
522 inventory across the five test sites (Figure 5). General patterns included a clear surface  
523 association of contamination at the high activity organic-rich sites (1, 3 and 4) and sites with

524 significant vertical penetration: sites 2 (sandy) and 5 (ploughed). Applying the models  
525 developed from MCS to data collected over the test sites yielded encouraging results.  
526 Generally, there was good agreement between core and detector inventory estimates  
527 particularly for sites 1, 2 and 4 with less than 25 % difference between core and detector  
528 estimates (Figure 6A).

529

530 .....*Insertion Point for Figure 5*.....

531

532 ***Figure 5. Inventory depth distributions derived from soil cores extracted from the five***  
533 ***GAMFAC sites. Uncertainties are one standard deviation and based on counting statistics.***

534

535 Results are very similar for both detectors for all of the sites confirming earlier percentage  
536 error investigations (see Figure 4A). The detectors struggled equally to characterise the  
537 inventory at sites 2 and 5 where differences of ca. 53 % and 45 % were encountered (see  
538 Figure 6A). It became clear that this method may be limited at sites where the level of  
539 contamination is relatively low and at greater depths when operating with one second count  
540 times. To lower the uncertainty, it might be appropriate to lengthen the count time or,  
541 alternatively, conduct slower traverses and increasing the spatial density of spectra, which  
542 could, retrospectively, be summed to improve statistics. It must also be noted that remote  
543 measurements tend to only sample the top 20 cm or so of soil, much of the contamination at  
544 these sites is below this range and most possibly beyond the detection capabilities of these  
545 instruments (see Figure 4A). In addition, it may be argued that these sites could be described  
546 as having subsurface maxima ( $\sim 20 \text{ g cm}^{-2}$ ), which the exponential model would have  
547 struggled to accurately characterise (see Figure 5). Alternate more flexible models, for  
548 example the one described by Hillman et al. (1996) or (Almgren and Isaksson, 2006) might be  
549 more appropriate in this case. This could have been more representative of the PTV values  
550 found in the field and reduced the uncertainty associated with final inventory estimates.

551

552 .....*Insertion Point for Figure 6*.....

553

554 ***Figure 6. Median and interquartile range values for inventory (A) and  $\beta$  (B) estimates for***  
555 ***the five GAMFAC sites for lanthanum bromide and sodium iodide compared to core***  
556 ***results.***

557 An example of how complex the spatial distribution of  $^{137}\text{Cs}$  in the PSRER can be is  
558 provided, whereby inventory and  $\beta$  values obtained using NaI:Tl from site 2 and the area  
559 surrounding have been smoothed using universal kriging (Graphical abstract). First of all,  
560 notice the reference site chosen on the sand dune (red broken lines) was relatively stable with  
561 little variation in inventory and depth confirming it a good choice of reference site to apply  
562 static *in situ* systems. However, outside of the isolated site large fluctuations in inventory and  
563 depth were witnessed. For instance two relatively large hotspots (5-15 meters in diameter) to  
564 the north of the site were estimated to possess more than six times the inventory ( $\sim 1500 \text{ kBq}$   
565  $\text{m}^{-2}$ ) of the reference site. Due to the geometry and distance away from the reference site  
566 these sources would nonetheless have little impact on final calibration measurements taken at  
567 the site. Together with greater inventory,  $\beta$  values associated with these hotspot were  
568 estimated to be greater than the surrounding area suggesting contamination was deeper in the  
569 soil column. This finding could suggest accumulation and penetration of contamination  
570 particularly in areas of the topographic depression in the foreground, which was lower in  
571 elevation than the sand dune and could potentially accumulate  $^{137}\text{Cs}$  through erosion  
572 processes. Further investigative studies involving coring and chemical analysis would have to  
573 be performed to confirm this hypothesis.

574

575 ***Graphical abstract. Estimated inventory and  $\beta$  obtained for  $71 \times 71 \text{ mm}$  sodium iodide***  
576 ***detector for site 2 (red box) and surrounding area. Notice the reference site (red box) is***  
577 ***significantly more stable than surrounding areas in terms of depth and activity.***

578 **4. Conclusions**

579 The Chernobyl Nuclear power plant disaster has been the single largest nuclear reactor  
580 meltdown in our history. Subsequently, highly radioactive material can be found in large  
581 areas of Ukraine, Belarus and Russia presenting a considerable remediation challenge that  
582 could take decades or even centuries to overcome. An important step in this process is  
583 develop methods to characterise the activity and burial depth of  $^{137}\text{Cs}$  in high spatial  
584 resolution. To this end, a highly practical method has been described and validated against  
585 reference sites within the Polessie State Radioecological Reserve in southern Belarus. It was  
586 shown that a conventional  $71 \times 71$  mm sodium iodide scintillation detector could exhibit  
587 equal performance to a significantly more expensive lanthanum bromide detector of the same  
588 size and power requirements. This outcome can be attributed to the intrinsic contamination  
589 associated with lanthanum bromide. Depth and activity maps of  $^{137}\text{Cs}$  contamination have  
590 demonstrated the advantage of the technique applied to the sodium iodide detector. Further  
591 application of this approach could significantly support future remediation activities  
592 ultimately lowering the risk to human health.

593

594 **5. Acknowledgements**

595 This work was performed as part of the GAMFAC intercomparison exercise funded by the  
596 Nordic Nuclear Safety Research. The authors would like to acknowledge the staff of the  
597 Polessie State Radiation Ecology Reserve of Belarus for the organisation of the GAMFAC  
598 project and processing of cores. Additonal gratitude must be served to Ms. Elena Almås for  
599 her invaluable translation.

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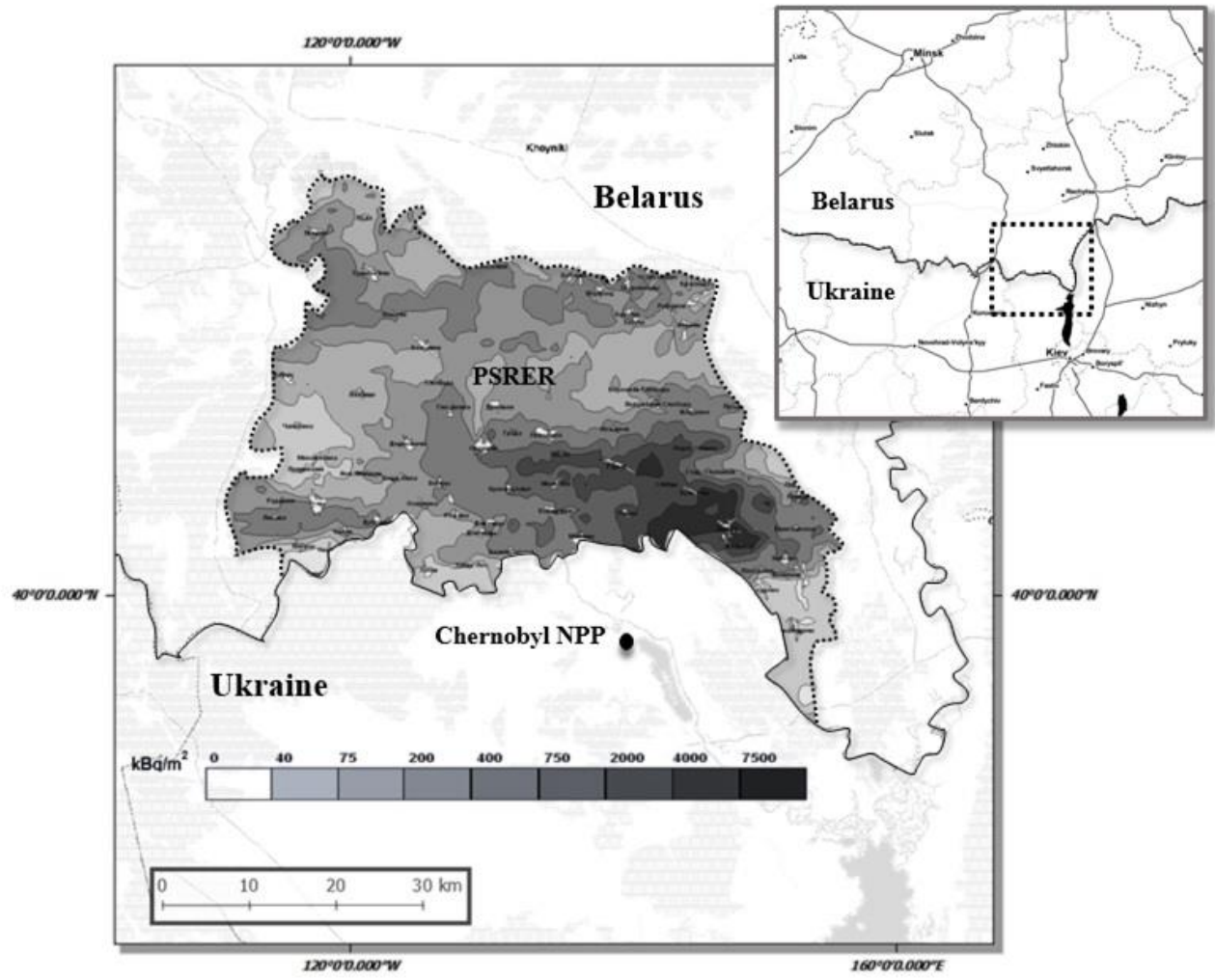
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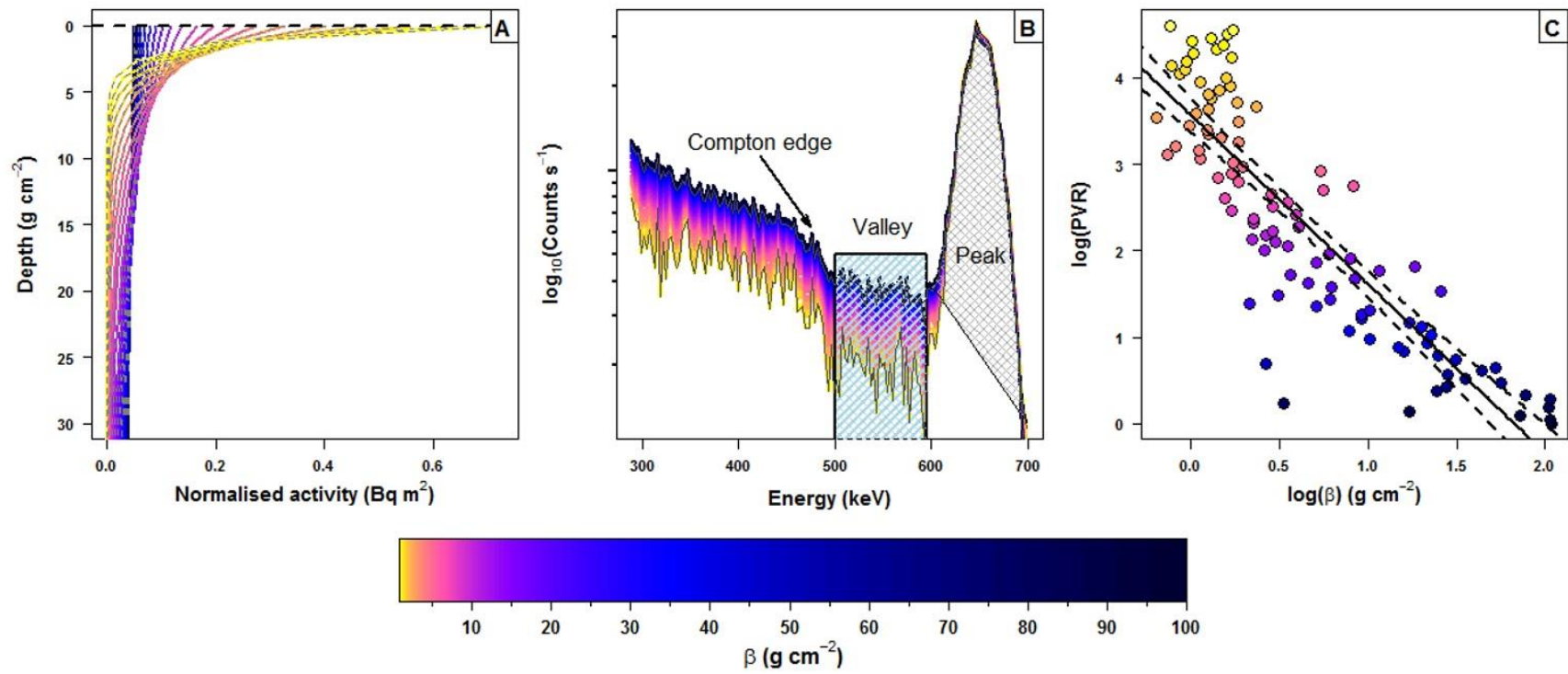


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835 *Figure 1*

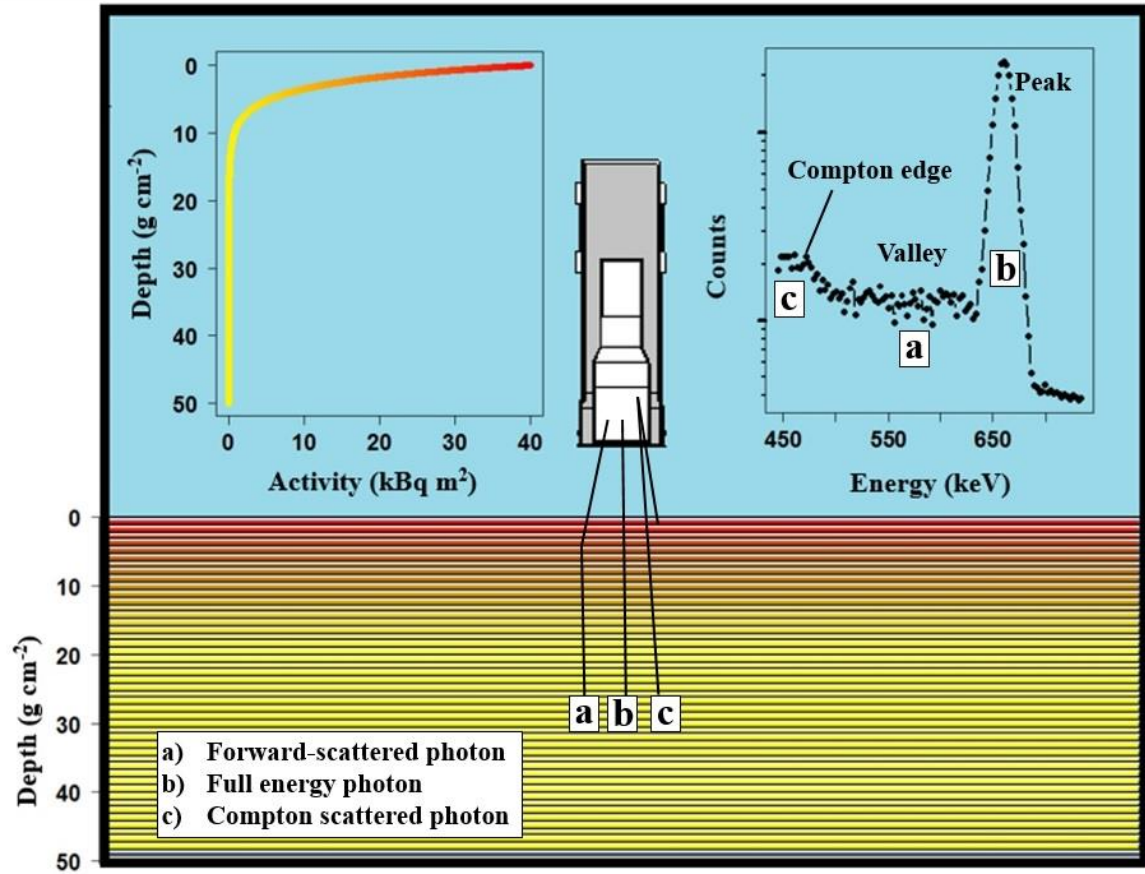
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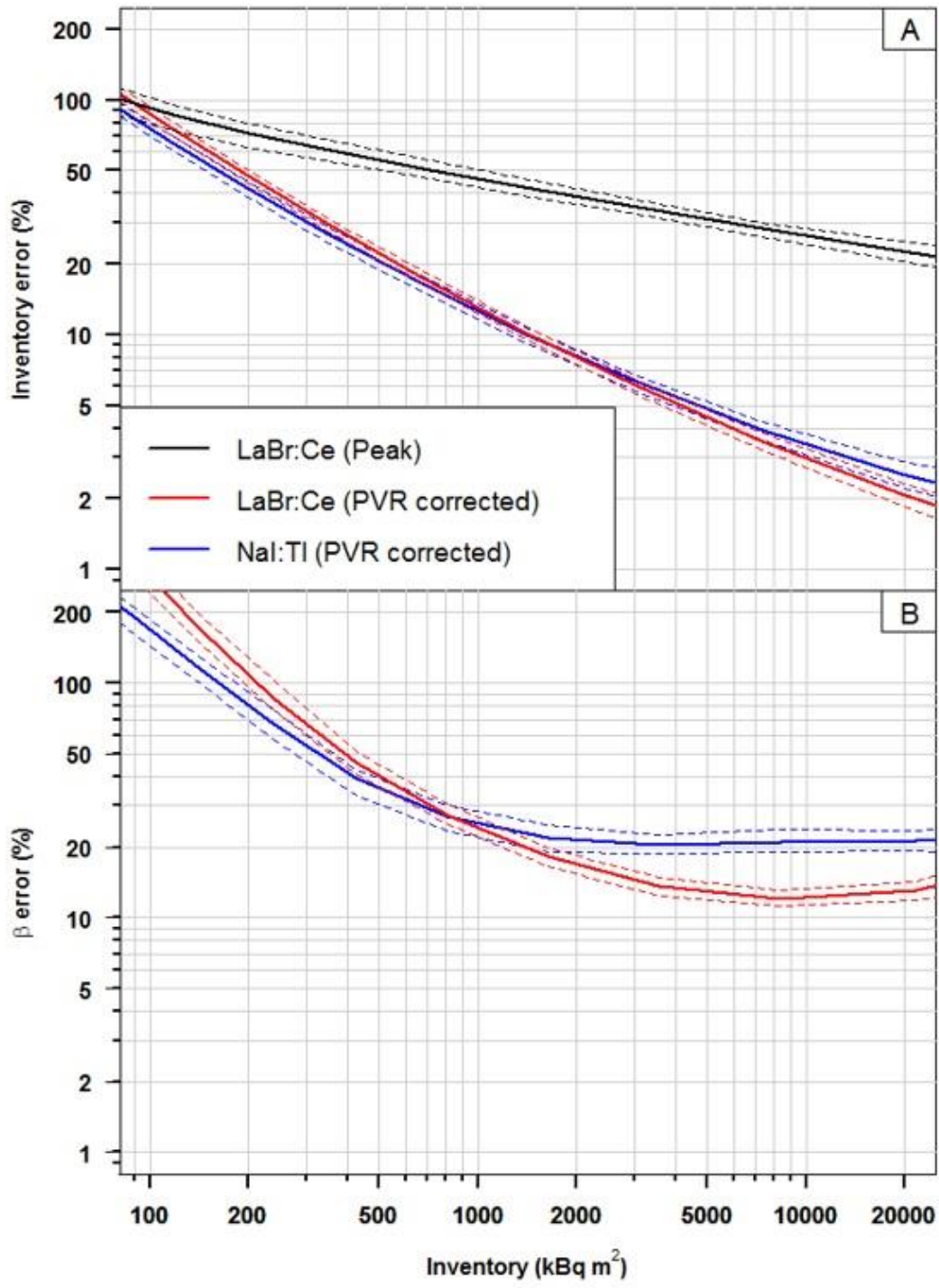
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838 *Figure 2*



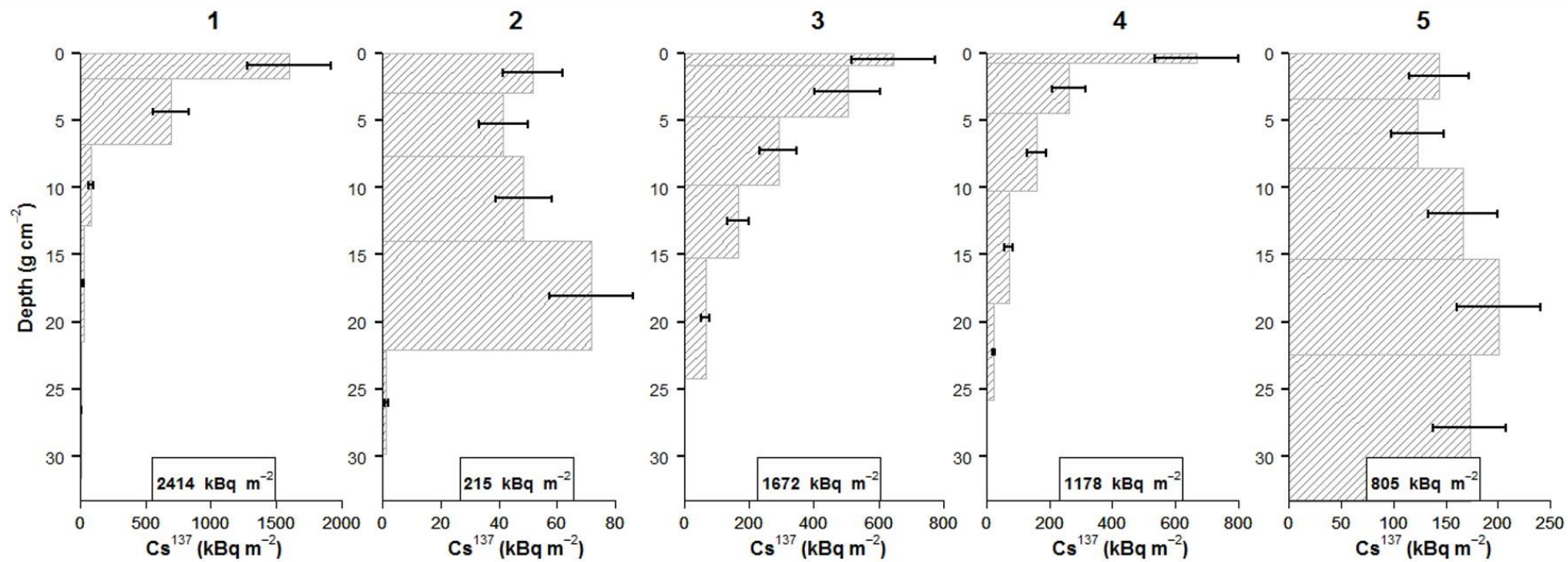
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840 *Figure 3*



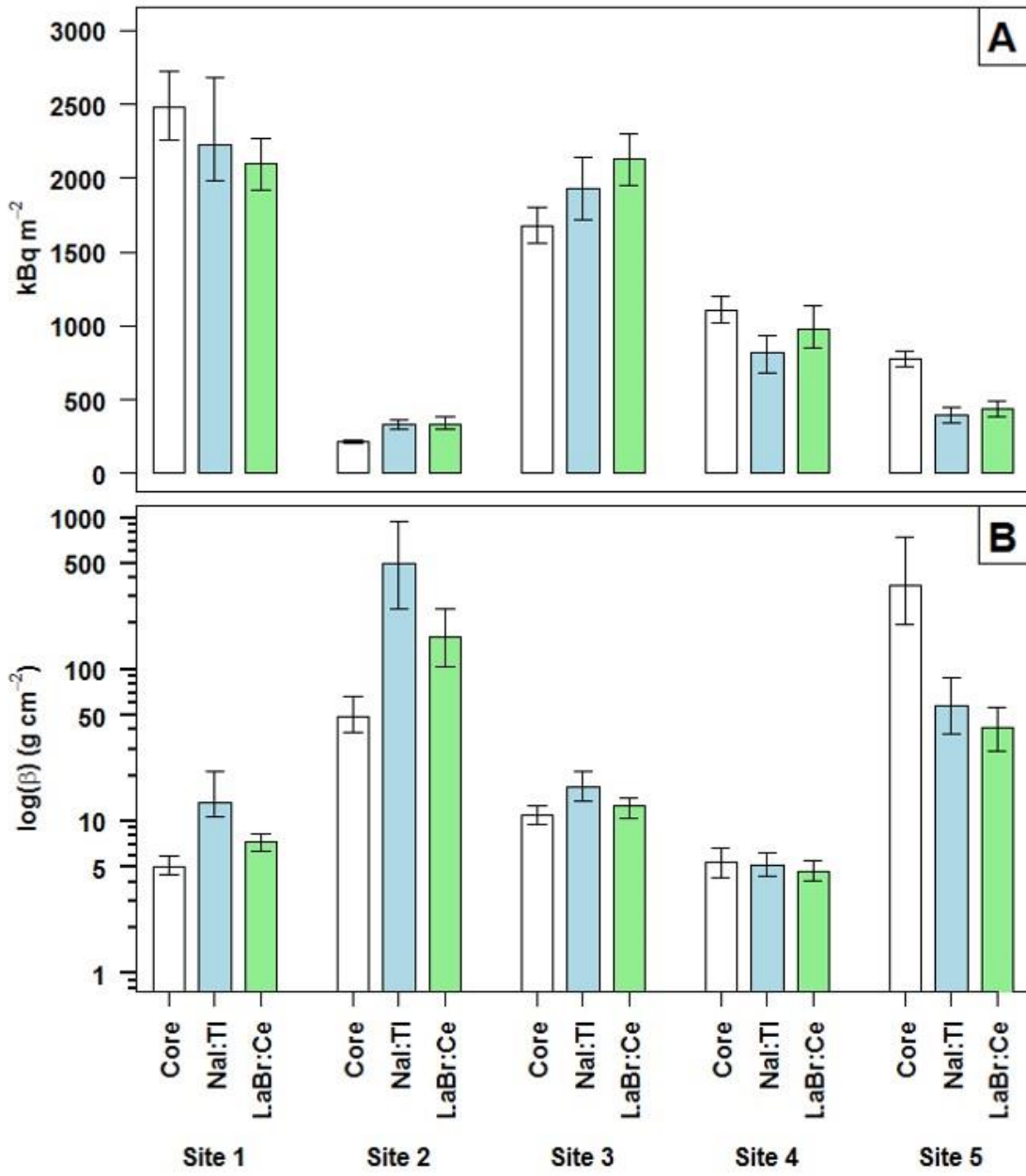
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842 *Figure 4*



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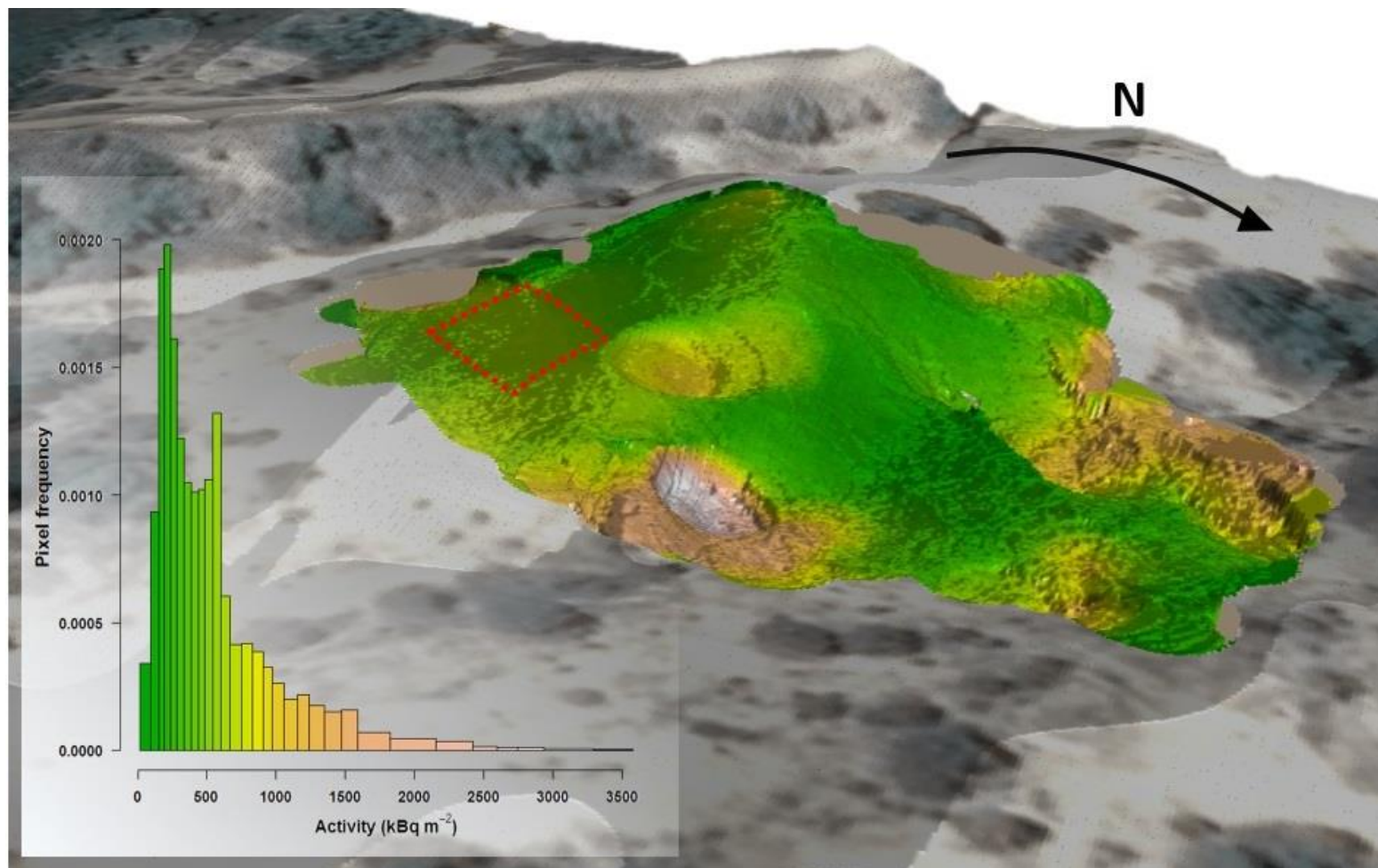
844 *Figure 5*



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*Graphical abstract*

