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1 **Title: Reconstructing the deposition environment and long-term fate of Chernobyl ^{137}Cs**
2 **at the floodplain scale through mobile gamma spectrometry.**

3
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13
14 **Keywords: Caesium redistribution; Sedimentation rates; Floodplain functioning;**
15 **Gamma-ray spectrometry; Chernobyl**

16
17 **Abstract:**

18
19 Cs-137 is considered to be the most significant anthropogenic contributor to human dose and
20 presents a particularly difficult remediation challenge after a dispersal following nuclear
21 incident. The Chernobyl Nuclear Power Plant meltdown in April 1986 represents the largest
22 nuclear accident in history and released over 80 PBq of ^{137}Cs into the environment. As a result,
23 much of the land in close proximity to Chernobyl, which includes the Polessie State
24 Radioecology Reserve in Belarus, remains highly contaminated with ^{137}Cs to such an extent
25 they remain uninhabitable. Whilst there is a broad scale understanding of the depositional
26 patterns within and beyond the exclusion zone, detailed mapping of the distribution is often
27 limited. New developments in mobile gamma spectrometry provide the opportunity to map
28 the fallout of ^{137}Cs and begin to reconstruct the depositional environment and the long-term
29 behaviour of ^{137}Cs in the environment. Here, full gamma spectrum analysis using algorithms
30 based on the peak-valley ratio derived from Monte Carlo simulations are used to estimate the
31 total ^{137}Cs deposition and its depth distribution in the soil. The results revealed a pattern of
32 ^{137}Cs distribution consistent with the deposition occurring at a time of flooding, which is
33 validated by review of satellite imagery acquired at similar times of the year. The results were
34 also consistent with systematic burial of the fallout ^{137}Cs by annual flooding events. These

35 results were validated by sediment cores collected along a transect across the flood plain. The
36 true merit of the approach was confirmed by exposing new insights into the spatial distribution
37 and long term fate of ^{137}Cs across the floodplain. Such systematic patterns of behaviour are
38 likely to be fundamental to the understanding of the radioecological behaviour of ^{137}Cs whilst
39 also providing a tracer for quantifying the ecological controls on sediment movement and
40 deposition at a landscape scale.

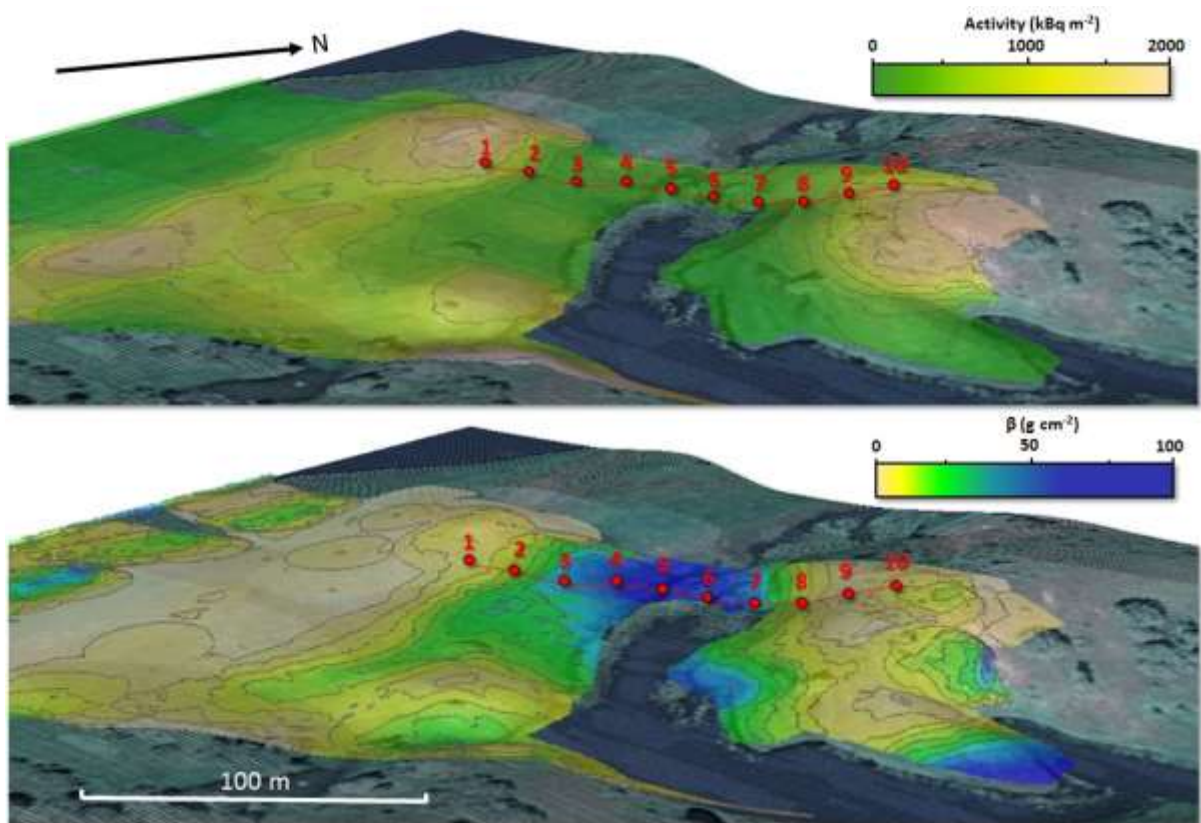
41

42 **Research capsule:**

43

44 Complex deposition patterns of ^{137}Cs have been observed on floodplains providing insight into
45 the role environmental processes play in the spatio-temporal redistribution of contamination

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48

49 Graphical abstract. Interpolated handheld gamma-ray spectrometry estimates for activity
50 (kBq m^{-2}) and β (g cm^{-2})

51

52

53 1. Introduction

54

55 Cs-137 is considered the one of the most radiologically significant anthropogenic
56 radionuclide within the environment (Miller, 2007). The majority of present day global
57 inventory of ^{137}Cs is derived from atmospheric weapons testing (1945-1963). More recently,
58 further more-localised contamination has been introduced into the environment through
59 reactor meltdowns occurring at Chernobyl Nuclear Power Plant (ChNPP) in 1986 and
60 Fukushima Daiichi Nuclear Power Plant (FDNPP) in 2011. The widespread nature of ^{137}Cs
61 and the risk it poses to human health are as a result of a combination of physical and chemical
62 properties. Foremost, it is one of the more abundant fission products generated during a
63 nuclear chain reaction and it has a relatively low melting point (670.8 °C) allowing it to
64 readily disperse into the atmosphere after a meltdown or nuclear bomb detonation (Isaksson
65 and Raaf, 2017). Once in the environment, it can exhibit complex biogeochemical behaviour
66 given it is highly soluble, has a tendency to react with environmental media in particular clay
67 minerals and can be readily mobilised in the presence of organic matter. It can also easily
68 enter the food chain as it is readily taken up into plant material substituting for naturally
69 occurring potassium (Penrose et al., 2016). In terms of human exposure, the combination of
70 30.1 year half-life, high energy beta decay and the 662 keV gamma-ray from the short-lived
71 daughter product $^{137\text{m}}\text{Ba}$ make ^{137}Cs both an internal and external radiation hazard. As a
72 result, following any nuclear incident one of the fundamental tasks is to establish, using high
73 quality data, the extent and intensity of ^{137}Cs deposition to inform decision making. This need
74 has been exemplified by, for example, the largescale data collection efforts initiated by the
75 Ministry of Education, Culture, Sports, Science and Technology (MEXT) within a month of
76 the FDNPP accident (Povinec et al., 2013) and the multinational efforts over a number of
77 decades to confidently characterise the spatial distribution of fallout across Europe after the
78 ChNPP accident (Izrael et al., 1996).

79

80 In the longer term, continued measurement of ^{137}Cs is also important, not only to update
81 population dose assessments (Beresford et al., 2016; Konoplev et al., 2017), but to monitor
82 redistribution of the radiocontaminant within the environment occurring as a function soil
83 movement, weathering and erosion (Rawlins et al., 2011). These are the same process that
84 have enabled soil scientists to regularly utilise ^{137}Cs as a medium-term tracer for soil
85 redistribution since the 1960's (Mabit and Dercon, 2014; Ritchie and McHenry, 1990).

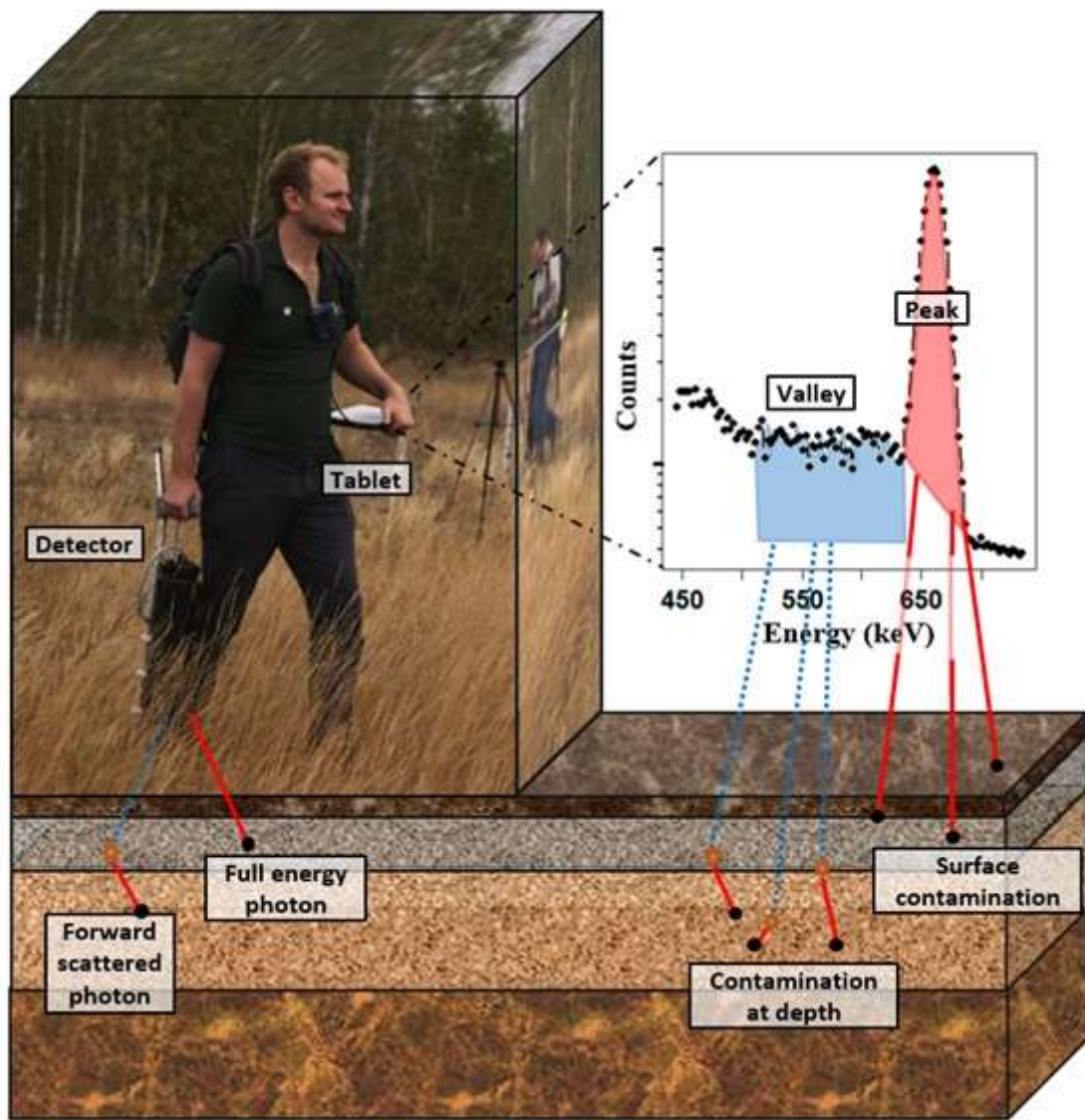
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87 Cs-137 is relatively straightforward to measure through gamma-ray spectrometry of the 662
88 keV photon released by its daughter product $^{137\text{m}}\text{Ba}$ within which it is always in secular
89 equilibrium (Povinec et al., 2003). However, difficulties often arise when trying to account
90 for depth variation of ^{137}Cs contamination given that significant penetration into the soil
91 column can occur over time making precise estimates of the total inventory (Bq m^2)
92 problematic (Tyler, 2004). The oldest and most direct way to measure the depth distribution
93 is by extracting cores from a site and measuring small increments of the core (typically less
94 than 5 cm slices) within the laboratory using low background High Purity Germanium
95 (HPGe) detectors; thus building a profile. Collection, preparation and analysis of
96 environmental samples can nonetheless make this procedure time consuming and expensive
97 often yielding limited spatial representivity, which can become a major issue when
98 significant spatial heterogeneity of contamination is encountered (Golosov et al., 2000).
99 Possibly a more appropriate method that can address spatial resolution issues more
100 effectively is the deployment of field-based mobile gamma-ray spectrometry (Tyler, 2008).
101 Utilising this technique, measurements can be made directly in the field using a variety of
102 gamma-ray detectors mounted to a specific platform such as static surveyor stands (*in situ*)
103 (Gering et al., 1998), vehicles (carborne) (Aage et al., 2006), airborne (helicopters and fixed
104 wing) (Rawlins et al., 2011) and unmanned aerial vehicle (UAVs) (Okuyama et al., 2008).
105 The height of a detector primarily determines the spatial quality of data given that the field of
106 view (FOV) of the detector will be increased with increasing height (Tyler et al., 1996a).
107 Alongside time and cost restraints this factor is crucial in determining the choice of platform
108 to characterise the underlying spatial distribution of ^{137}Cs contamination.

109

110 *In situ* technologies, utilising highly sensitive HPGe detectors, represent the standard for
111 characterisation of gamma-ray emitting radionuclides (ICRU, 1994). Nonetheless, HPGe
112 detectors generally have low efficiency, require heavy cooling systems and are as a result
113 bulky and not suited for mapping large areas in high spatial resolution. Therefore portable
114 systems, known as mobile gamma-ray spectrometry, tend to use more robust sodium iodide
115 (NaI:Tl) detectors with larger detection volumes (and hence efficiency) in order to improve
116 statistical accuracy (Tyler, 2008). Airborne survey systems as an example can carry
117 hundreds of kilos of NaI:Tl providing very high detection efficiencies that are ideal to rapidly
118 characterise ^{137}Cs contamination on a national scale. However, such surveys come at
119 considerable cost and for safety reasons tend to be conducted at altitudes above 100 m
120 resulting in a large FOV, making resolution of localised contamination difficult. Carborne

121 systems on the other hand can provide a higher spatial quality of data for significantly less
122 cost whilst still being able to carry significant detection volumes, such surveys tending to be
123 restricted to roads or open fields. UAVs offer enormous potential in the field of gamma-ray
124 spectrometry, however current affordable technologies cannot provide the payloads to carry
125 detectors with sufficient detection efficiency, for long enough periods, to characterise large
126 contaminated areas in high spatial resolution (Martin et al., 2016).
127



128
129 **Figure 1.** Photograph of a handheld detection system alongside a schematic representation of
130 the peak to valley ratio methodology used to account for burial depth of contamination
131

132 Lightweight detection systems originally designed for nuclear security missions are now
133 commonly being implemented in the field as backpack or handheld devices (Figure 1) to
134 characterise ^{137}Cs in relatively heavily contaminated environments (Cresswell et al., 2013;
135 Kock and Samuelsson, 2011; Nilsson et al., 2014; Plamboeck et al., 2006; Sanderson et al.,
136 2013). Typically, a large number of georeferenced spectra are taken during a survey and,
137 conventionally, the count rate in the full energy peak is used to determine the amount of ^{137}Cs
138 contamination in the soil. However, large uncertainties in inventory estimates (Bq m^{-2}) can
139 be encountered when significant spatial heterogeneities in burial depth are encountered.
140 Recently, Varley et al., (2017) demonstrated that through the use of the peak-to-valley ratio
141 (PVR) improved inventory estimates could be made for aged Chernobyl deposits in Belarus.
142 The benefit of utilising the PVR to account for burial depth was first realised by Zombori et
143 al. (1992) and has since been used widely to estimate the burial depth of ^{137}Cs (Feng et al.,
144 2012; Gering et al., 2002, 1998; Kastlander and Bargholtz, 2005; Tyler, 2004). In essence,
145 the PVR is the ratio of the full energy photopeak and the forward scattered photons (Figure
146 1). Forward scattered photons are those photons that have lost a small amount of energy on
147 transition out of the soil and are represented by the “valley” region of the gamma spectrum
148 that may be observed between the full energy photopeak at 662 keV and the Compton edge
149 occurring at 480 keV. The greater the depth within the soil at which the photon originates,
150 the more probable it is that the photon will undergo forward scattering and generate a count
151 within the valley region of the spectrum. This leads to an increase of the valley “height” and a
152 concomitant decrease in the PVR.

153

154 Following the calibration and validation work of assessing the PVR method for mobile
155 handheld gamma spectrometry (Varley et al., 2017) it was apparent that one calibration site
156 exhibited a systematic change in the deposited ^{137}Cs activity and depth distribution towards a
157 tributary of the Pripyat River. The primary aim was to follow up on this observation and to
158 complete a high resolution (<1 m) mobile gamma spectrometry survey across the flood plain
159 to characterise and explain the systematic distribution in a landscape that had been highly
160 impacted by the 1986 Chernobyl accident and that had hitherto been considered to be highly
161 heterogenous with regards to the ^{137}Cs distribution.

162

163 Large areas of the watershed of the Pripyat River, the main watercourse of the impacted
164 region and including feeder streams, lakes, marshes and drainages, are within the 30-km
165 exclusion zone that was ultimately imposed around the ChNPP. Since the accident, high

166 concentrations of radionuclides have been observed within stagnant and slow moving bodies
167 of water associated with the Pripjat River. Such radionuclides accumulate primarily in
168 relation to sediments although accumulations of contaminants can also be observed within the
169 water itself and associated biota (Gudkov et al., 2010). Floodplains have been shown to
170 represent relatively complex post-depositional environments (Golosoov et al., 2013; Iwasaki et
171 al., 2015) due to the influence of flood events on a number of processes that govern the
172 environmental behaviour of ^{137}Cs and the contaminated areas of Belarus and Ukraine also
173 bear witness to this complexity (Burrough et al., 1999a; Kagan and Kadatsky, 1996; Zhukova
174 et al., 1997). In this context, it is likely that the systematic mapping of the distribution of
175 ^{137}Cs using handheld mobile gamma spectrometry within the watershed of the Pripjat River
176 in the Polessie State Radioecology Reserve (PSRER), will reveal a complex redistribution of
177 deposited ^{137}Cs at a scale that is likely to be overlooked by conventional sampling, in situ,
178 mobile or airborne gamma spectrometry due to the limitations of sampling or measurement
179 resolution.

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198 2. Methodology

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200 2.1. The Study Site

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202 The selected site (centred at approximately N 51°31'45.2", E 029°56'07.5") was known to be
203 periodically inundated given that it lay in a low-lying floodplain within the drainage system
204 of the Pripyat River. Lying some 19 km from the Chernobyl Plant the site had moderate dose
205 rates ranging from 0.1 to 0.5 $\mu\text{Sv/h}$ recorded at 1 m in height and contamination densities of
206 ^{137}Cs , ^{90}Sr and ^{241}Am of the order of 1.2 MBq/m², 0.2 MBq/m² and 0.032 MBq/m²,
207 respectively. The soils at the site consist primarily of soddy gleys and peat soils of varying
208 density and being better drained at the elevated locations of the site. Vegetation at the site
209 consisted primarily of grasses and mosses, with emergent and submergent macrophytes at
210 lower elevations and near water with some sparse stands of low trees and bushes at the higher
211 elevations.

212 2.2. Soil sampling and measurement

213

214 Soil samples along a transect of approximately 120 m were taken using a steel cylindrical
215 corer (4 cm diameter, 20 cm depth). The transect was selected such that the main features of
216 the site, with respect to potential redistribution of ^{137}Cs , were accounted for. Cores were
217 divided into 2 cm increments. Overlying vegetation and humus was removed prior to
218 sectioning. Soil samples were prepared for radioanalytical measurements by air drying and
219 homogenization followed by mixing and sieving (2 mm aperture). After weighing, a
220 subsample was transferred to an analytical geometry for counting. A HPGe (Canberra,
221 Be5030, carbon window, 50% rel. eff.) detector was used for ^{137}Cs determination. Count
222 times varied from 6000 to 80000 sec. Statistical uncertainties for the photo peak of ^{137}Cs were
223 less than 5 %.

224 2.3. In situ Gamma Spectrometry

225

226 The survey was conducted over the course of a day by two NaI:Tl detectors, a 76 × 76 mm
227 NaI:Tl and a 51 × 51 mm, by two operators. Each detector was connected to an Ortec
228 digiBase recording a 1024 channel spectral per second using Ortec's Maestro software
229 (ORTEC, 2005). GPS coordinates were recorded individually for each spectrum by an SX
230 Blue II differential GPS with an uncertainty of 0.6 m. Spectral measurements and spatial
231 coordinates were combined using software built within the JAVA framework and able to

232 provide real-time feedback on count rates in specified windows. Maestro's gain stabilisation
233 software was used on the 662 keV peak to avoid significant spectral drift. The detectors were
234 held at a height of about 0.1 m from the ground to reduce the field of view with a traverse
235 speed of approximately 0.5 m s^{-1} being maintained throughout the surveys. To compare the
236 performance of the $51 \times 51 \text{ mm NaI:Tl}$ against $76 \times 76 \text{ mm NaI(Tl)}$ concomitant transects for
237 undertaken and tie lines were run perpendicularly across the survey area and compared
238 during the later spatial analysis. Certain sections of the survey area could not be accessed on
239 foot due to the thickness of vegetation and water inundation.

240 *2.4. Detector calibration*

241

242 Monte Carlo Simulations (MCS) were chosen to derive the relationship between the
243 contamination burial depth and PVR over alternative expensive analytical experiments or
244 complex deterministic equations (Maučec et al., 2009; Stromswold, 1995). Crucially, MCS
245 can depict the variation of PVR with ^{137}Cs depth distribution much more precisely than using
246 a small sample size of soil core data collected in the field (Likar et al., 2004). Furthermore,
247 uncertainties associated with field measurements can be better controlled (Boson et al., 2009)
248 making MCS an important calibration tool in field gamma-ray spectrometry (Cinelli et al.,
249 2016; Clouvas et al., 1998; Maučec et al., 2004; Thummerer and Jacob, 1998; Yip et al.,
250 2015).

251

252 In this study, the software package Monte Carlo N-Particle 5 code (MCNP5) was used to
253 obtain spectral responses in order to derive PVR values for appropriate depth distribution
254 (Briesmeister, 1993). To ensure that each depth distribution could be adequately modelled,
255 disk-shaped source descriptions measuring 10 mm thick were simulated down to a depth of
256 70 g cm^{-3} (Varley et al., 2017). In this manner the PVR could be defined with higher depth
257 resolution and facilitated improved counting statistics as the maximum particle number in
258 MCNP5 (2×10^9) could be run for each simulation, thereby effectively increasing the source
259 density without repeated random number sampling (Hendriks et al., 2002). Although
260 contributing a relatively small number of counts to the detector, the natural radioelements
261 were also modelled in MCNP5 as they were deemed to contribute to the valley region
262 (Varley et al., 2017). Resultantly, source descriptions for ^{40}K and the ^{238}U and ^{232}Th series
263 were also provided to MCNP5 in separate simulations and were assumed to be uniform
264 (Thummerer and Jacob, 1998). Only the main components of each detector were used in

265 models encompassing the active volume of the detector (NaI:Tl; density 3.51 g cm^{-3}), the
266 aluminium outer casing, the outer casing and the glass window of the photomultiplier tube
267 and the PVC protective outer casing. Detector geometries that were validated in early studies
268 were utilised: $51 \times 51 \text{ mm}$ (Varley et al., 2015) and $76 \times 76 \text{ mm}$ (Varley et al., 2017). The
269 codes used to derive these models have been validated using concrete calibration pads
270 (Grasty et al., 1991). The F8 tally implemented in MCNP5 was used to capture the
271 differential energy spectrum within the active volume of each simulated detector. A
272 Gaussian broadening parameter was applied to simulate the statistical broadening of peaks
273 brought about by the imperfect collection of photons within each detector (Kangas et al.,
274 2008). Additionally, as PVR data was generated using energies that can be considered as
275 being immune to photoelectric effect and backscatter by virtue of their energy ($>350 \text{ keV}$), a
276 standard soil composition from Beck et al. (1972) was employed and photons below 350 keV
277 were terminated in each run to save on computer time. Photons were recorded at lower
278 energy than the Compton edge ($350\text{-}478 \text{ keV}$) as it was first thought that this statistically
279 defined region could be used alongside the valley region ($496\text{-}584 \text{ keV}$) to improve depth
280 estimates. All nuclide decay data used throughout was obtained from the National Nuclear
281 Data Centre (2013).

282 *2.5. Depth distribution model*

283

284 For the purpose of the study, the relaxation mass per unit area (β) was utilised (eq 1). This
285 relates the rate by which the surface ^{137}Cs activity (A_0) decreases with mass depth in the form
286 of an exponential decay function (ICRU, 1994). Where (A_m) is the ^{137}Cs activity of the soil in
287 Bq kg^{-1} and ζ is the mass of soil per unit area (g cm^{-2}). The value of β will approach 0 for
288 surface contamination and approach ∞ as the contamination is evenly distributed over the
289 entire column. Mass per unit area was preferred over physical depth (cm) in this discussion as
290 it incorporates density (ρ), facilitating a better approximation of shielding effects and
291 permitting a more accurate estimate of ^{137}Cs inventory (Bq m^{-2}). Using this method, each 10
292 mm slice of ^{137}Cs source model was weighted according to a specific β and inventory value.
293 The total inventory is calculated by integrating eq 1 between the limits of 0 and 70 g cm^{-2} .
294 Given there was a significant layer of vegetation above the soil surface and surrounding the
295 detector, a thickness of 1 g cm^{-2} of lower level contamination was also included in
296 simulations. Contamination densities encountered at the site in overlaying vegetation,
297 although highly heterogeneous, was found to be approximately 5 % the contamination

298 density of the upper soil layer. Spectral responses derived using this adopted model
299 generated spectral responses that were far more representative of the scattering conditions
300 found in the field.

301

$$302 \quad A_m(\zeta) = A_{m,0} \cdot e^{(-\zeta/\beta)} \quad [1]$$

303

304 By adopting this approach, one stipulates that the maximum activity will lie within the first
305 few centimetres of soil near the surface; an observation that has, despite the age of
306 contamination, been noted relatively recently at undisturbed sites within the PSRER
307 (Dowdall et al., 2017). Nonetheless, it must be noted that there is still a high probability of a
308 subsurface maximum, particularly in areas of significant accumulation or in the case where
309 the soil type lends itself to high vertical migration rates as observed elsewhere in Europe for
310 example (Bernhardsson et al., 2015). However, in this case the difference in contamination
311 between surface and subsurface maximum is likely to be so small it will be beyond the
312 detection limits for the PVR method. Moreover, the overall difference between final
313 estimates of inventory will be negligible compared to an estimated high β value as would be
314 anticipated using the exponential model (eq 1). Interestingly, environmental processes in the
315 PSRER differ to the process of accumulation of historic Sellafield-derived ^{137}Cs discharges in
316 saltmarsh sediments off the coast of the United Kingdom, in which considerable subsurface
317 maxima can be encountered (Tyler, 1999; Tyler et al., 1996b).

318 *2.6. Estimating inventory using peak-to-valley ratio*

319

320 The primary purpose of utilising the PVR is as a correction factor to better estimate
321 inventory, therefore the area under the peak (587-737 keV) and the valley height must be
322 calculated for individual field spectra. As there was going to be a small amount of spectral
323 drift throughout the survey, efforts were made to find the peak centroid by fitting a Gaussian
324 function the maximum of which acted as an updated energy calibration. The background
325 under the peak was estimated using a linear function fitted to the adjacent five channels on
326 either side of the base. The estimated background was then subtracted before fitting of a
327 second Gaussian function and integrating to determine the area. The valley height was
328 determined via subtraction of the area on the high energy side of the peak (740-755 keV)
329 from the valley region (496-584 keV) in a manner described by (Tyler et al., 1996b). The
330 routine described above was performed using the base features of the R statistical package (R

331 Core Development Team, 2016). Once these values had been derived, a linear model was
332 fitted to MCS values of PVR and calculated calibration coefficients (C) (eq 2). Here, C is
333 defined by taking the ratio of the activity (A) measured in Bq m^{-2} and peak count rate (N_p) (eq
334 3). To obtain the final activity (A) estimate for a field spectrum, the predicted value of C
335 needs to be multiplied by the number of counts in the full energy peak (N_p).

336

$$337 \log(C) = C_0 + C_s \cdot (PVR) \quad [2]$$

338

$$339 C = A/N_p \quad [3]$$

340

341 *2.7. Quality assessment and mapping of detector estimates*

342

343 To assess and compare the quality of individual detector performance, approximately 300 one
344 second count time spectra were taken directly over each of the ten extracted cores sites. The
345 PVR method was then applied to each set of spectra to give some approximation of the
346 accuracy and precision of the method, assessed through the mean and standard deviation,
347 respectively. Final maps were produced by combining predicted β and activity values for all
348 data by using the inverse distance weighting algorithm to produce an interpolated surface
349 (Shepard, 1968).

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362 3. Results and Discussion

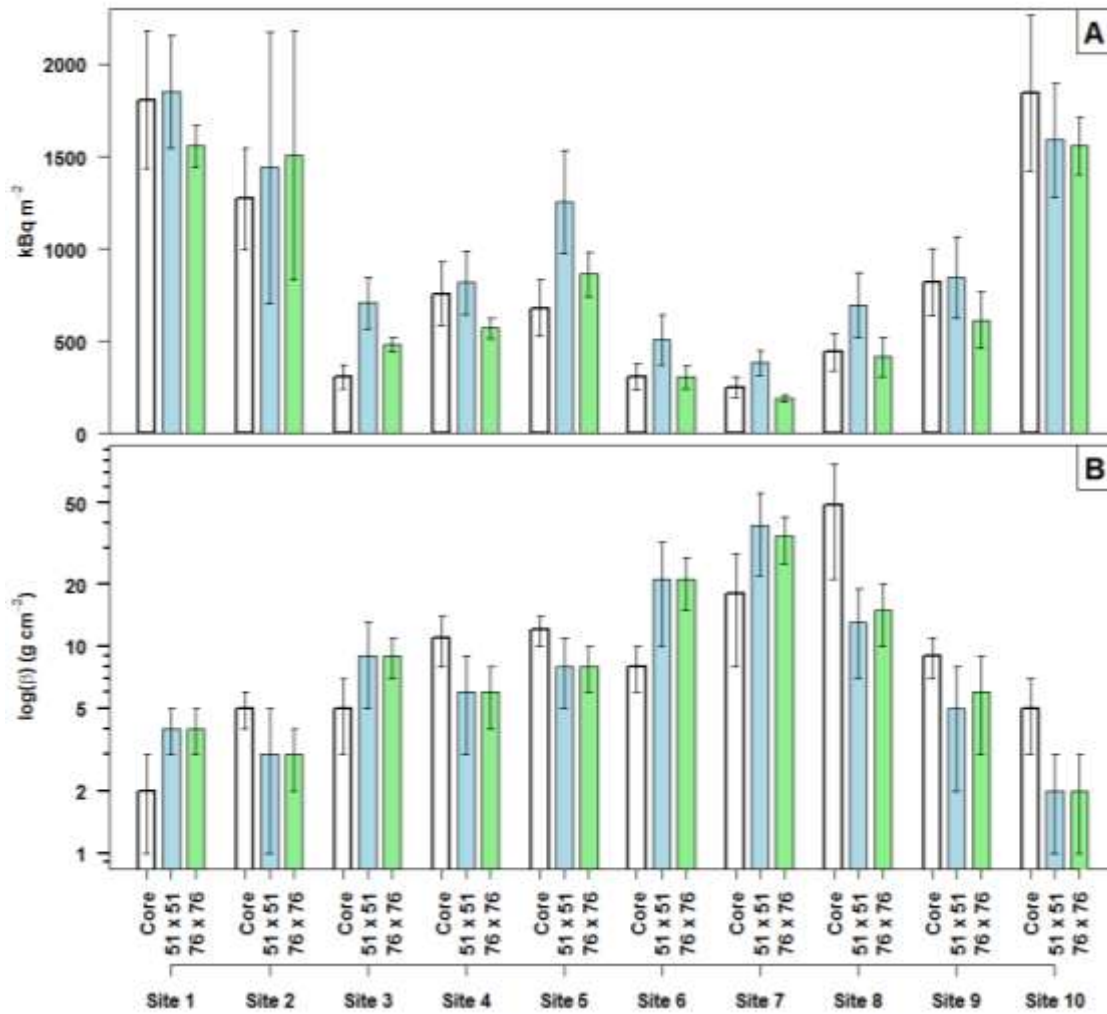
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364 3.1. Detector predictions compared to core results

365

366 Mean activity (kBq m^{-2}) estimations obtained using 51×51 mm and 76×76 mm sodium
367 iodide detectors over the top of the ten core positions show good agreement with activity and
368 depth results derived from cores (Figure 2). It should be emphasised that single point
369 comparisons may be influenced by the high scale localised heterogeneity emphasising the
370 differences in the spatial representativeness of a single core against a field gamma
371 spectrometry measurement that have >1000 the sample volume. This occurrence is most
372 apparent at site 2, whereby large standard deviations are seen on results for both detectors.
373 Based on this evidence it could be argued that the similarity between detector estimates,
374 particularly with regards to depth, could signal that this approach is in fact more reliable than
375 single coring estimates. In general, it becomes clear that the higher energy efficiency,
376 demonstrated by the 76×76 mm, presented an advantage over the 51×51 mm especially at
377 lower count rates, which can be realised by a reduction in the spread (standard deviation) of
378 activity and depth estimates at each site. However, this might not always be the case where, for
379 example, at much higher activity sites the 51×51 mm detector may be more appropriate for
380 deployment due to the amount of dead-time experienced using the 76×76 mm.

381



382
 383 Figure 2. Mean and standard deviations for estimates derived from cores, 51 × 51 mm and 76
 384 × 76 mm for the 10 sites along the transect A) activity (kBq m⁻²) and B) β (g cm⁻²).

385
 386 Overall, results derived using gamma-ray spectrometry provide sufficient evidence that this
 387 method is a highly practical option to map the spatial and depth distribution of ¹³⁷Cs
 388 contamination at this site and other sites with similar activities and vertical penetration.
 389 Equally, other scintillation instruments, such as lanthanum bromide or cerium bromide, of
 390 similar size could also be used effectively (Guss et al., 2010). In identifying ways to reduce
 391 uncertainty and thus improve the quality of field data it was concluded that the assumption of
 392 an exponential depth distribution must at some stage be reconsidered as this is thought to be
 393 the principle cause of error within this study. This fact was highlighted in the fitting of the
 394 exponential model to observed core data where large residuals were encountered. For
 395 example, R² values for cores 7 and 8 were 0.68 and 0.51, respectively. Although for the
 396 remaining cores R² values were all greater than 0.88. Additionally, vegetation cover and
 397 water inundation were revealed to significantly influence instrument response, which would

398 be hard to characterise by conventional in field calibration, but Monte Carlo simulations
399 present a practical alternative to account for these parameters. It will be the focus of future
400 work to test other distributions in more detail, but within the context of the present study it
401 was accepted that the uncertainty associated with using an exponential distribution would
402 have little influence on final conclusions of contamination distribution.

403 *3.2. Site interpretation*

404

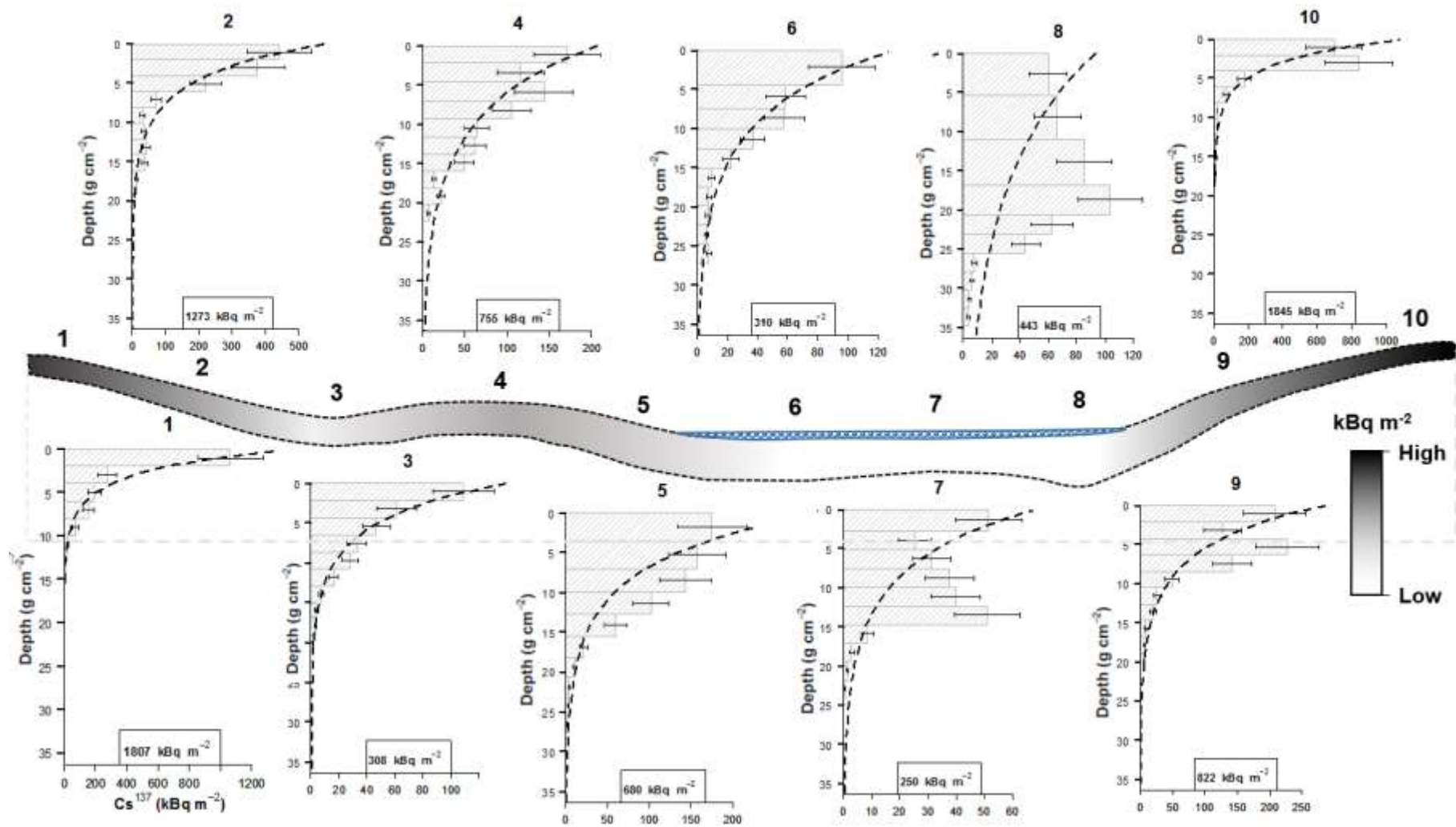
405 Based on the results of cores taken along the transect, ^{137}Cs is distributed heterogeneously
406 over the extent of the site and is characterised by higher ^{137}Cs inventories, between 1.5 and 2
407 MBq m^{-2} , exhibited on the elevated areas of the site on both sides of the waterbody (Figure
408 3). Higher inventories tend to match surface distributions (corresponding to low β values).
409 Conversely, lower ^{137}Cs inventories are primarily confined to the depressions along the
410 transect and correspond reasonably well with areas affected by the rise and fall of the water
411 level in the two bodies of water present; this can be seen in cores 3, 6, 7 and 8. Furthermore,
412 the soils in the associated deeper sections appear to exhibit markedly more pronounced
413 penetration of the contaminant down the soil column. Noticeable subsurface maxima can be
414 observed in cores 7, 8 and 9 (Figure 2). This general observation is, at first consideration,
415 perhaps contrary to what may be expected as there is extensive evidence in the literature that
416 periodic flooding often results in enhancement of ^{137}Cs soils levels due to the addition of
417 contaminated sediment following flood deposition (Burrough et al., 1999b), localised
418 redistribution of eroded soils (Golosov et al., 1999) and the fixation of water borne ^{137}Cs in
419 soils (Standing et al., 2002). Here, however, the lower ^{137}Cs inventories associated with
420 topographical depressions at this site may be explained by three factors possibly acting in
421 combination.

422

423 Firstly, contamination was known to be heterogeneously deposited especially in areas close
424 to the reactor such as the PSRER (Boulyga et al., 2003). The possibility therefore exists that
425 the depression at the site simply received less ^{137}Cs when deposition occurred. However, this
426 seems relatively unlikely given that the deposition densities on the elevations on both sides of
427 the water exhibit quite similar values (Figure 3). The systematic distribution of activity
428 estimated from handheld devices provides further support to oppose this notion given that
429 other areas within the study area also possess the highest activities on elevated areas at
430 greater distance from the water body (Figure 3).

431

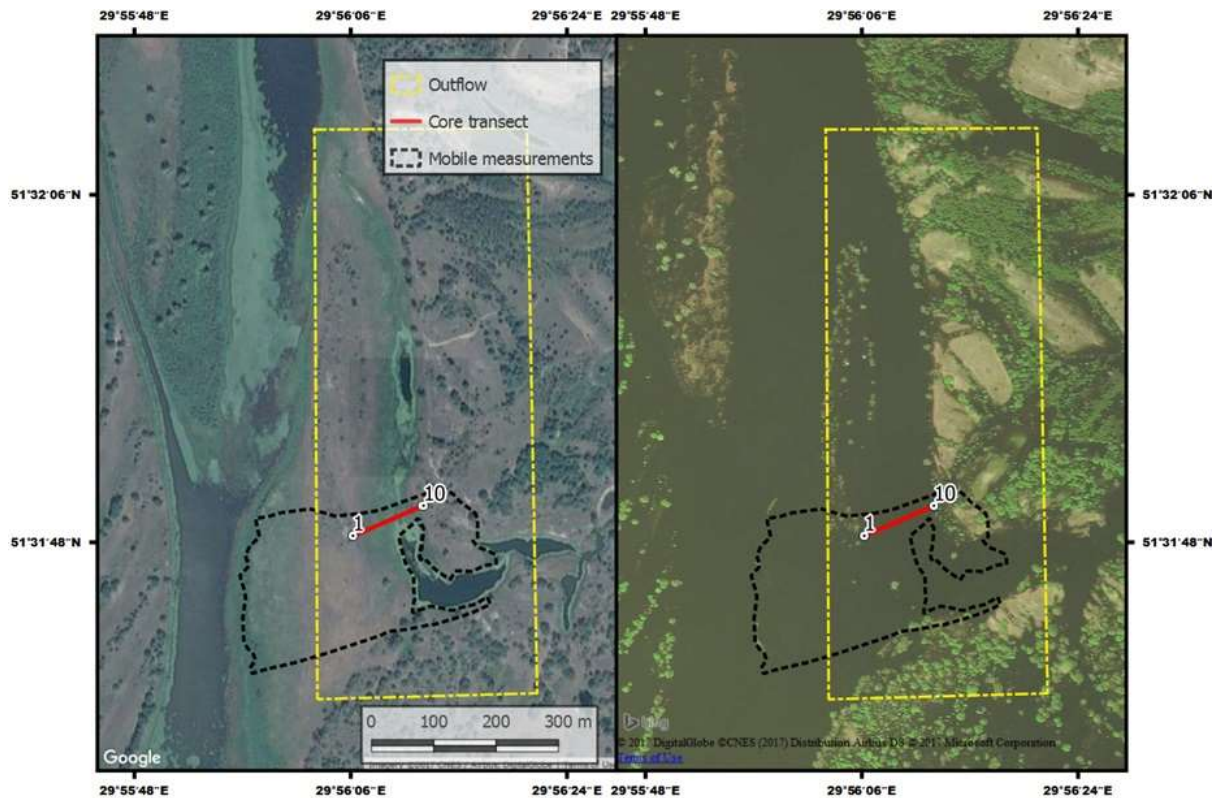
432 A second cause may be associated with soil biogeochemistry owing to the significant
433 presence of organic matter contained within the soil. It is well established that ^{137}Cs remains
434 highly mobile in organic soils (Kudelsky et al., 1996; Rigol et al., 2002; Ritchie and
435 McHenry, 1990; Tegen and Dörr, 1996) primarily due to lower clay mineral contents. As a
436 result, saturated organic soils may exhibit a tendency to lose ^{137}Cs due to its higher mobility,
437 which may ultimately result in lower inventories as ^{137}Cs is lost laterally. Crude examination
438 of the cores revealed the presence of more organic material contained within the lower cores,
439 particularly those that were submerged at the time of extraction (cores 6, 7 and 8). The fact
440 that these same cores exhibit greater depth penetration also supports the theory that
441 mobilisation (as well accumulation) has occurred. Similar profiles of contamination on flood
442 plain soils have been demonstrated by Golosov et al., (Golosov et al., 1999). Yet, without
443 comprehensive chemical analysis data available this theory is hard to prove. Nevertheless,
444 accepting that this is the primary driving force behind the spatial distribution at the site is
445 difficult when considering that significant activity and the gradient of depth variation
446 encountered between elevations and depressions (Graphical abstract).



447

448 Figure 3. Schematic diagram of estimated elevation alongside depth-activity profiles for cores taken from along the transect

449 However, the evidence points to a third and perhaps more unusual set of circumstances that
450 may explain the spatial distribution during primary deposition. Regional flooding was known
451 to have taken place during the time of the Chernobyl meltdown brought on by annual snow
452 melt and this can clearly be witnessed over the entire PSRER in 30 m resolution Landsat 5
453 images taken on the 29th of April 1986 courtesy of the U.S. Geological survey. The pixel
454 resolution of these images is not sufficient enough to directly assess the extent of flooding at
455 the site and it has been difficult to find specific evidence in the literature indicating that this
456 site was flooded in April of 1986. Resultantly, at this stage it cannot be confirmed the extent
457 of flooding at the site, although it was known that other water bodies of the region were in
458 flood at that time (Korobova et al., 2007). Nevertheless, the influence of flood water across
459 the study site can be demonstrated using more recent, considerably higher resolution, satellite
460 imagery taken in autumn 2003 by Googlemaps (Figure 4A) and spring 2008 by Bing imagery
461 (Figure 4B). For example in the Bing images, high flood water can be seen to be inundating
462 much of the flood plain within the main outflow (yellow box), resultantly only the highest
463 elevated areas of the study site (cores 1 and 10) can be seen to be above the water line, with
464 all other core sites between these end members being submerged under flood water (Figure
465 5B). This evidence, provides the explanation that the areas of lower ¹³⁷Cs deposition are as a
466 result of flooding at the time of contaminant deposition. Under these conditions, overlying
467 water, potentially containing large amounts of suspended sediment, to which the ¹³⁷Cs could
468 absorb, delayed it reaching the underlying soil and sediment column and preferentially
469 transporting the ¹³⁷Cs downstream from the study site. Only elevated areas or those that
470 became exposed during the period of the Chernobyl fire would have received conventional
471 atmospheric deposition (Graphical abstract). Post-accident deposition was then characterised
472 by sediment with similar or lower activity concentrations (cores 7 and 8) that was washed
473 into the area from upstream, gradually burying the ¹³⁷Cs peak deposition from the time of the
474 accident at a rate that is proportional to the frequency of flood inundation. The rate of
475 accumulation could relate to surface roughness a factor that may heavily be controlled by
476 aquatic and terrestrial vegetation in addition to topography (Figure 3).



477

478 Figure 4. Satellite imagery of the study site in A) Autumn 2003 (Googlemaps) and B) Spring
 479 2008 (Bing imagery). The location of the core transect, the outflow and convex hull of
 480 handheld measurements are marked out on the map. The ten samples were collected on the
 481 transect marked by the red line.

482 **4. Conclusions**

483

484 The peak-to-valley ratio method has been successfully applied to data, collected using an
 485 inexpensive and robust handheld scintillator, to derive activity and depth estimates of ^{137}Cs
 486 contamination within the Polessie State Radioecology Reserve. The study has verified that
 487 large areas can be mapped with relatively high accuracy and much faster compared to
 488 conventional resource intensive soil sampling approaches. Spatial information gained from
 489 the deployment of the handheld gamma spectrometry systems have provided a unique insight
 490 into what proved to be a highly complex post-depositional environment, the consequences of
 491 which have not been previously explored in the literature. The results have demonstrated that
 492 the flooding at the time of the Chernobyl accident can explain systematic variations in
 493 deposition across the floodplain and subsequent sediment accumulation is burying the peak
 494 ^{137}Cs fallout at a rate that is proportional to the frequency of flooding.

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496

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