

1 Effectiveness of landscape decontamination following the Fukushima nuclear accident: A review

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11 **Abstract**

12 The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in March 2011 resulted in the
13 contamination of Japanese landscapes with radioactive fallout. Accordingly, the Japanese authorities
14 decided to conduct extensive remediation activities in the impacted region to allow for the relatively
15 rapid return of the local population. The objective of this review is to provide an overview of the
16 decontamination strategies and their potential effectiveness in Japan, focussing on particle-bound
17 radiocesium. In the Fukushima Prefecture, the decision was taken to decontaminate the fallout-
18 impacted landscapes in November 2011 for the 11 municipalities evacuated after the accident (Special
19 Decontamination Zones – SDZ, 1117 km²) and for the 40 non-evacuated municipalities affected by
20 lower, although still significant, levels of radioactivity (Intensive Contamination Survey Areas, 7836
21 km²). Decontamination activities predominantly targeted agricultural landscapes and residential areas.
22 No decontamination activities are currently planned for the majority of forested areas, which cover
23 ~75% of the main fallout-impacted region. Research investigating the effectiveness of
24 decontamination activities underlined the need to undertake concerted actions at the catchment scale
25 to avoid the renewed supply of contamination from the catchment headwaters after the completion
26 of remediation activities. Although the impact of decontamination on the radioactive dose rates for
27 the local population remains a subject of debate in the literature and in the local communities, outdoor
28 workers in the SDZ represent a group of the local population that may exceed the long-term dosimetric
29 target of 1mSv yr⁻¹. Decontamination activities generated ~20 million m³ of soil waste by early 2019.
30 The volume of waste generated by decontamination may be decreased through incineration of
31 combustible material and recycling of the less contaminated soil for civil engineering structures.
32 However, most of this material will have to be stored for ~30 years at interim facilities opened in 2017
33 in the close vicinity of the FDNPP before being potentially transported to final disposal sites outside of
34 the Fukushima Prefecture. Further research is required to investigate the perennial contribution of
35 radiocesium from forest sources. In addition, the re-cultivation of farmland after decontamination
36 raises additional questions associated with the fertility of remediated soils and the potential transfer
37 of residual radiocesium to the plants. Overall, we believe it is important to synthesize the remediation
38 lessons learnt following the FDNPP nuclear accident, which could be fundamental if a similar
39 catastrophe occurs somewhere on Earth in the future.

40

41 **Keywords:** radiocesium; caesium-137; FDNPP; remediation; phytoextraction; Japan.

42

43 1. Introduction

44 Large quantities of radiocesium (12–62 PBq) were released into the environment by the Fukushima
45 Dai-ichi Nuclear Power Plant (FDNPP) accident in March 2011 (Stohl et al., 2012;Chino et al., 2011).
46 Accordingly, this accident hit the maximum level (i.e. 7) on the International Nuclear and Radiological
47 Event Scale (INES)(IAEA, 2013a). Airborne and ground contamination surveys demonstrated that the
48 contamination was the highest (i.e., initial ^{137}Cs levels $>100,000 \text{ Bq m}^{-2}$) in a plume extending to the
49 northwest of the FDNPP covering an area of $\sim 3000 \text{ km}^2$ (Kinoshita et al., 2011;Chartin et al.,
50 2013;Yasunari et al., 2011). Although many radioactive substances were released into the
51 environment by the FDNPP accident, radiocesium (i.e., ^{134}Cs and ^{137}Cs) presents the most serious risk
52 to the local population over the medium to long term as it was emitted in very large quantities and it
53 has a relatively long half-life (i.e., ^{134}Cs - 2 years; ^{137}Cs - 30 years) (Steinhauser et al., 2014). The highest
54 contamination radiocesium levels observed in Fukushima ($>185 \text{ kBq m}^{-2}$) are similar to those recorded
55 in the area impacted by Chernobyl fallout, although on a much smaller land surface area in Japan (1700
56 km^2) compared to eastern Europe (29,400 km^2)(Steinhauser et al., 2014).

57 Numerous investigations have been conducted by Japanese and international researchers to improve
58 our understanding of the fate of radiocesium in the Fukushima region (for a review, see: Evrard et al.,
59 2015). In general, radiocesium sorption mechanisms were characterised (Fan et al., 2014;Nakao et al.,
60 2015) and their fluxes measured in riverine systems draining the main radioactive plume (Nagao et al.,
61 2013). Land use (Koarashi et al., 2012) and soil properties (Nakao et al., 2014) were shown to control
62 the migration of radiocesium in soils. Accordingly, the fate of this contaminant was intensively
63 investigated in forest ecosystems (Gonze and Calmon, 2017) and cultivated landscapes (Yoshimura et
64 al., 2016), which are the two main land uses in the fallout-impacted region. Typhoons and other major
65 rainfall events were also demonstrated to drive soil erosion and sediment migration processes thus
66 directly influencing post-fallout radiocesium dynamics (Chartin et al., 2017).

67 Between 2011–2018, there were 578 published studies with the keywords ‘radiocesium’ and
68 ‘Fukushima’ in the Scopus database (Figure 1). Approximately 90% of these articles were published by
69 Japanese scientists, demonstrating the extensive research effort conducted by the national scientific
70 community in Japan on the processes occurring in this post-accidental context. Since the second half
71 of 2013, remediation activities started to be implemented under the supervision of the Japanese
72 authorities to decontaminate soils. These activities have significantly affected the spatial and temporal
73 redistribution of radionuclides in the Fukushima-impacted area. As decontamination is now completed
74 in many regions and more than 50 scientific studies have been conducted on different aspects of these
75 operations (Figure 1), synthesizing the results obtained by this applied research is important for the
76 scientific community. Of note, this review will not synthesize non-peer reviewed reports published by
77 the Japanese authorities, although numerous resources are available on the official websites of
78 multiple Japanese ministries (Table 1).

79 Although radiocesium is mainly transported in particle-bound form (i.e. through sorption and fixation
80 to micaceous clay minerals) in the Fukushima fallout-impact area (Konoplev et al., 2016), dissolved
81 radiocesium was found in numerous environmental compartments, primarily during the immediate
82 post-accidental phase (Yoshimura et al., 2014). As most of the dissolved radiocesium migrated through
83 these landscapes immediately after the FDNPP accident, this literature review will focus on particulate
84 radiocesium. Furthermore, as ^{134}Cs and ^{137}Cs were emitted in equivalent proportions into the
85 environment in March 2011, with an initial $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio of ~ 1 (Kobayashi et al., 2017), this
86 review will focus primarily on ^{137}Cs owing to its longer half-life and thus greater risk to the local
87 population over the medium to long-term. Although knowledge has been gained on radiocesium
88 transfers (Ivanov et al., 1997) and ecosystem remediation (Santschi et al., 1990) after the Chernobyl

89 accident, the circumstances in which Fukushima and Chernobyl accidents occurred are very different
90 (Steinhauser et al., 2014). Moreover, the contrasting environmental conditions prevailing in the
91 fallout-affected areas of Japan and eastern Europe (Konoplev et al., 2016) complicate the direct
92 comparison of the fate of radionuclides and the effectiveness of potential remediation measures in
93 both regions. Accordingly, the goal of this review is to examine the remediation strategies and their
94 effectiveness for particulate bound ^{137}Cs in Japan.

95 This literature review will be divided into five main sections. First, the spatial extent of the
96 decontaminated zone and the schedule of these remediation activities will be outlined. Second, the
97 remediation strategies in different environments (i.e., farmland, river, forests) will be presented along
98 with a summary of their cost effectiveness. Third, the impacts of remediation activities on dosimetry
99 will be summarized. Fourth, the initiatives to manage the large volume of waste generated by
100 remediation will be discussed. Fifth, major research questions and requirements to guide the future
101 management of Fukushima fallout-impacted areas will be identified and presented. The objective of
102 this review is to provide a synthesis of the remediation lessons learnt in Japan following the FDNPP
103 nuclear accident, which are fundamental in light of the potential for a similar situation to occur
104 somewhere on Earth in the future (Christoudias et al., 2014).

105

106 **2. Areas targeted by decontamination**

107 The FDNPP suffered major damage following an earthquake and a tsunami that occurred on March 11,
108 2011. At this time, Units 1, 2 and 3 of the power plant were operational, and they suffered a series of
109 major failures (Burns et al., 2012). The main resulting radionuclide emissions that affected Japanese
110 landscapes occurred on March 15, 2011. Rainfall and snowfall that occurred on March 15 and 16
111 resulted in the formation of a radionuclide plume on soils located to the northwest of the power plant,
112 up to ~70 km from the site (Yasunari et al., 2011). The inhabitants living in areas coinciding with this
113 plume were progressively evacuated in spring and summer in 2011, and it took time for finding long-
114 term housing solutions for these populations (Asanuma-Brice, 2012).

115 In November 2011, the Japanese government adopted the *Act on Special Measures Concerning the*
116 *Handling of Pollution by Radioactive Materials* (Japanese Ministry of the Environment, 2011b) in order
117 to reduce the impact of radioactive substances from the FDNPP accident on human health and the
118 environment (Yasutaka and Naito, 2016). In support of this Act, decontamination guidelines were
119 released by the Japanese Ministry of Environment in December 2011 and updated in 2013. These
120 guidelines outlined the methods for surveying and quantifying the levels of contamination and the way
121 to prepare these areas targeted for remediation (Japanese Ministry of the Environment, 2013). A
122 decontamination roadmap (*Policy for Decontamination in the Special Decontamination Area*) was
123 implemented in January 2012 under the direct supervision of the Japanese government.

124 According to the decontamination roadmap, the remediation programme had to be implemented in
125 'special areas' where targets were set for the exposure of the public to external dose rates in order for
126 residents to return to their day-to-day lives (Yasutaka and Naito, 2016). Achieving pre-accident
127 radiation levels is not the objective, rather the effectiveness of decontamination will ultimately depend
128 upon the land use and the air dose of each particular area. Two zones were delineated with different
129 strategies for remediation (Figure 2), based on different values of dose equivalent (i.e. the biological
130 effect of ionizing radiation) expressed in Sievert (Sv) and its sub-units. First, Special Decontamination
131 Zones (SDZ) are areas located within a 20-km radius of the FDNPP or areas where the cumulative dose
132 one year after the accident was expected to exceed 20 mSv yr^{-1} . SDZs occur in 11 municipalities (1117
133 km^2) where residents were evacuated after the FDNPP accident in 2011. The central government of

134 Japan is responsible for remediation works in SDZs. Second, Intensive Contamination Survey Areas
135 (ICAs) refer to 102 municipalities from eight Prefectures with ambient dose rates exceeding $0.23 \mu\text{Sv}$
136 h^{-1} (equivalent to 2 mSv yr^{-1}), designated as ICAs by the Ministry of Environment on December 28, 2011
137 (Mori et al., 2017). The area of the ICAs is eight times greater than the SDZ (Yasutaka and Naito, 2016).
138 In particular, the decontamination methods and target areas for remediation in the ICAs differ from
139 those of the SDZ with decontamination activities for the ICAs conducted by local governments with
140 support from the central government. In total, the SDZs and ICAs cover a surface area of 8953 km^2 with
141 a population that was not evacuated after the accident of ~ 1.7 million (Yasutaka and Naito, 2016).

142 In the literature, there is debate regarding the need to initiate decontamination so quickly after the
143 FDNPP accident (e.g., Yasutaka et al., 2013a). Delaying decontamination could allow for the natural
144 decay of radioisotopes and thus significantly lower the costs of achieving radiation exposure targets.
145 For example, Munro (2013) estimated that the optimal delay for implementing remediation activities
146 was in the range of 3–10 years after the accident, with an optimal delay of 8.8 years.

147

148 **3. Decontamination Strategies and their Cost Effectiveness**

149 The effectiveness of decontamination was assumed to strongly vary depending on the remediation
150 method and the initial radiation dose rates prior to decontamination. Different remediation
151 techniques were proposed depending on the land use and the zone (i.e., SDZ vs. ICA). Yasutaka et al.
152 (2013a) and Yasutaka et al. (2013b) compared the impact of four scenarios of decontamination,
153 including two very unlikely options (i.e., minimal and maximal scenarios), in terms of effectiveness and
154 cost according to the results of demonstration tests conducted by JAEA. These results were updated
155 in a more recent publication (Yasutaka and Naito, 2016). Only the two scenarios following the
156 guidelines provided by the Japanese government are assessed in this review. As such, the results of
157 both the minimal and maximal remediation options are therefore not discussed. In the first scenario,
158 5 cm of topsoil was removed from 50% of agricultural land in the SDZ where ^{137}Cs concentrations
159 exceed 5000 Bq kg^{-1} , and replaced with a 5-cm layer of 'clean' soil (measure A1; Table 2). In the
160 remaining 50% of agricultural land with ^{137}Cs concentrations below 5000 Bq kg^{-1} , topsoil was replaced
161 with subsoil (measure A3). Ploughing with zeolite and potassium (measure A4) was adopted for
162 agricultural land in ICAs where the annual additional effective dose exceeded 1 mSv yr^{-1} . In the second
163 scenario, measure A1 was applied to all cultivated land. Similar measures (e.g. Table 2) were included
164 in both scenarios to decontaminate forested areas, roads and houses. The total decontamination cost
165 for implementing these remediation measures varied between 2–5.1 trillion JPY (~ 16 –41 billion EUR),
166 with 1.3–2 trillion JPY (~ 10 –16 billion EUR) for the SDZ and 0.7–3.1 (~ 6 –25 billion EUR) for the ICAs.
167 Although the area where decontamination has been implemented in the ICAs (922 – 3330 km^2) covers
168 a surface 3 to 11 times larger than that of SDZ (295 km^2), the decontamination costs for the SDZ and
169 ICAs are in the same order of magnitude.

170 The decontamination program includes a variety of other activities on top of the actual on-site
171 remediation works, including the transport of waste, the volume reduction of waste along with the
172 temporary, the interim, and the final storage of decontamination waste and removed soil in
173 containers. Depending on the set of measures implemented in the field, the cost of the remediation
174 works will therefore be highly variable. A synthesis of the unit costs for waste management and storage
175 is provided in Table 3 (Yasutaka and Naito, 2016). As shown by Yasutaka et al. (2013b), the quantity of
176 waste generated when decontaminating agricultural land varies considerably depending on the
177 decontamination method used. Consequently, differences in the quantity of waste generated resulted

178 in large differences between agricultural land decontamination methods and the costs associated with
179 storage containers, temporary storage sites and interim storage facilities.

180 According to the latest available figures from the Japanese Ministry of the Environment (2019b) at the
181 end of 2018, the volume of soil waste generated in the SDZ was 9,100,000 m³ with a remediation cost
182 of approximately 1.5 trillion JPY (~12 billion EUR). In the ICAs, the latest figures available for March
183 2018 showed that 7,900,000 m³ of waste soil were produced with a remediation cost of approximately
184 1.4 trillion JPY, equivalent to ~11 billion EUR (Japanese Ministry of the Environment, 2019b). This
185 amount corresponds to ~8% of the total annual expenses of the European Union (~134 billion EUR in
186 2017) (European Union, 2018).

187

188 **4. Strategies for decontamination in various environments, and their effectiveness**

189 *Radiocesium distribution with depth in the soil*

190 In general, owing to the strong and nearly irreversible bond of radiocesium to fine soil particles, the
191 majority of FDNPP derived ¹³⁷Cs is stored within the topsoil (i.e. the top 5 cm) in undisturbed soils
192 (Lepage et al., 2014;Matsunaga et al., 2013;Takahashi et al., 2015;Mishra et al., 2015;Matsuda et al.,
193 2015). Mishra et al. (2015) reported that for these undisturbed soils, the vertical migration of
194 radiocesium down the soil profile was slower in forest soils compared to grassland soils. In disturbed
195 soils, anthropogenic activities may increase the depth migration of radiocesium down the soil profile
196 (Lepage et al., 2015;Matsunaga et al., 2013). For example, Lepage et al. (2015) illustrated that 90% of
197 the FDNPP derived radiocesium was homogeneous throughout the tilled soil layer in cultivated soils.
198 Endo et al. (2013) reported that radiocesium concentrations were not depth dependent in cultivated
199 soils (i.e. paddy fields) whereas they declined exponentially in uncultivated soil. Both Sakai et al.
200 (2014b) and Tanaka et al. (2013) also demonstrated that radiocesium from the FDNPP accident was
201 measureable at 15cm depth in rice paddy fields. As illustrated by Koarashi et al. (2012), the penetration
202 of radiocesium in the soil differed depending on both the land use and the physicochemical properties
203 of the soil (e.g., bulk density, clay content and organic matter content). However, remediation
204 strategies consisting in removing the top 5 cm layer of the soil should have been effective as cultivation
205 or other human activities that may have led to the redistribution of radiocesium with depth after the
206 accident were prohibited in the main fallout impacted region.

207 *Soil/farmland decontamination, and soil to plant transfers*

208 Different strategies were carried out in Japan to decontaminate soil in farmland, either removing the
209 contaminated layer of soil or through the sowing of plants having the capacity of extracting and
210 concentrating radiocesium from the soil. There are few publications in international journals regarding
211 the potential or effectiveness of the latter strategy (e.g. Pareniuk et al., 2015). Among the few available
212 studies, Kobayashi et al. (2014) grew thirteen plant species from three families (*Asteraceae*, *Fabaceae*,
213 and *Poaceae*) in shallow and deeply cultivated fields where the 0–8 cm and 0–15 cm soil layers were
214 respectively ploughed. The variation in plough depth was expected to reflect the impact of different
215 contact zones between the root systems and radiocesium in the soil. Overall, 29 to 225 Bq kg⁻¹ dry
216 weight of ¹³⁷Cs were found in the plants, corresponding to transfer factors ranging from 0.019 to 0.13
217 (geometric mean (GM), 0.057) for plants growing in shallow soils, and from 0.022 to 0.13 (GM 0.063)
218 for plants growing in deeper soils (Kobayashi et al., 2014). The authors found that none of their tested
219 plant species resulted in a significant decrease in radiocesium in soil likely because of the strong
220 fixation of ¹³⁷Cs to clay particles. This result was confirmed by Yamashita et al. (2014) who showed that
221 99 wild plants grown in paddy and upland fields had a very low phytoextraction efficiency. Tamaoki et

222 al. (2016) reached the same conclusions, although they suggested *Kochia (Bassia scoparia)* as a
223 potential candidate for phytoremediation although its efficiency in removing ^{137}Cs would require
224 numerous cultivation rounds.

225

226 Accordingly, given the low efficiency of phytoextraction, the main remediation strategy consists of
227 removing the surface layer of soils with the majority of radiocesium. The effectiveness of this strategy
228 was examined by Sakai et al. (2014a) in Kawamata Town. Approximately 5-10 cm of the surface soil
229 was removed from one rice paddy by heavy machinery, whereas a nearby paddy field was not
230 decontaminated and used as control plot. Both of these paddies then were ploughed and planted with
231 rice. Five surface soil samples (0-5 cm) were collected after decontamination and prior to ploughing
232 on June 12, 2011. Thereafter five soil cores (20 cm depth) were collected on July 13, 2012 at 3 m
233 intervals across both rice paddy fields. In 2011, the accumulation of radiocesium in the 0-5 cm surface
234 layer of the soil in the decontaminated paddy field ($170\pm 64 \text{ Bq kg}^{-1}$) was lower than the control rice
235 paddy field ($2231 \pm 64 \text{ Bq kg}^{-1}$). However, the ^{137}Cs concentration of the surface soil layer in the
236 decontaminated rice paddy field ($753\pm 62 \text{ Bq kg}^{-1}$) was significantly higher in 2012 than in 2011 (i.e.
237 after decontamination but prior to ploughing). This result suggests that radiocesium is likely
238 redistributed through the rice paddy field irrigation and drainage networks. The authors concluded
239 that the redistribution of soil within the paddy fields may decrease the effectiveness of
240 decontamination. A lack of replicates was outlined by the authors and prevented them to finally
241 conclude on the effectiveness of surface removal for decontamination (Sakai et al., 2014a). In contrast,
242 Kurokawa et al. (2019) observed a 80% decrease in ^{137}Cs activities in the ploughed layer after
243 decontamination in cultivated land of Tomioka Town, showing the efficiency of this remediation
244 strategy.

245

246 Another study was conducted in experimental paddy fields located ~40 km from the FDNPP (Wakahara
247 et al., 2014). Two plots were established: a paddy field where the top 5–10 cm of soil was removed
248 before cultivation and a control paddy. The ^{137}Cs soil inventory measured 3 months after the FDNPP
249 accident was approximately $200,000 \text{ Bq m}^{-2}$. However, after decontamination, this inventory
250 decreased to 5000 Bq m^{-2} . Suspended sediment and ^{137}Cs fluxes were measured in the outflow of the
251 paddy fields after puddling (i.e., the mixing of soil and water before planting rice) and they were 11.0
252 kg and $630,000 \text{ Bq}$ (1240 Bq m^{-2}) respectively in the control paddy, versus 3.1 kg and $24,800 \text{ Bq}$ (47.8
253 Bq m^{-2}) in the decontaminated paddy. After irrigation, 5.5 kg of particles and $51,900 \text{ Bq}$ (102 Bq m^{-2}) of
254 ^{137}Cs were discharged from the control plot, whereas 70 kg of suspended sediment and $165,000 \text{ Bq}$
255 (317 Bq m^{-2}) of ^{137}Cs were discharged from the remediated field. This 3-fold higher export of ^{137}Cs from
256 the decontaminated paddy was likely explained by the supply of contamination from upper paddy
257 fields, which remained connected to the remediated field through the irrigation network. This result
258 highlights the importance of remediation strategies focusing on the entire catchment scale.

259

260 Although this practice has not been specifically investigated in the literature, decontamination of
261 farmland in the Fukushima fallout-impacted region was not limited to the removal of the 5-cm topsoil
262 layer concentrating the radiocesium. After this first step, a layer of crushed granite, directly available
263 in the region as it was extracted from the bedrock in local quarries dedicated to decontamination, is
264 used to replace the removed soil layer (Evrard et al., 2019). The entire soil profile consisting of the
265 residual initial soil (in depth) and this crushed granite layer (on top) is then thoroughly mixed to
266 prepare the soil for recultivation with the objective being to further dilute the residual radiocesium
267 activities in the soil (Figure 3).

268

269 Other studies investigated the impact of remediation works on the radiocesium levels measured in
270 sediment transiting the river networks draining the main radioactive pollution plume. Evrard et al.
271 (2016) modelled the progressive dilution of radiocesium concentrations measured in sediment
272 following decontamination works. They demonstrated a 90% decrease of the contribution of upstream
273 contaminated soils to sediment transiting the coastal plains of the Mano and Niida Rivers between
274 2012 and 2015. Furthermore, Osawa et al. (2018) monitored the radiocesium concentrations in
275 suspended sediment collected in two tributaries of the Mano and Niida Rivers from 2013 and 2016.
276 They also attributed a decrease in the ^{137}Cs concentrations observed in 2016 to the decontamination
277 efforts completed in 2015 in the local catchments.

278

279 *River channel decontamination*

280 Riverside parks and playgrounds are popular across Japan. Sediment containing high quantities of
281 radionuclides may also accumulate near these parks and playgrounds in river channels and floodplains
282 following flooding events (Saegusa et al., 2016). However, sediment deposition is highly
283 heterogeneous both horizontally and vertically across floodplains. Furthermore, sediment deposited
284 in the river channel may be resuspended during subsequent flood events. In these conditions, the
285 common decontamination guidelines (i.e. removing the uppermost layer; Table 2) are difficult to
286 implement effectively.

287 Nishikiori and Suzuki (2017) investigated this challenge in the 13-km² Kami-Oguni River catchment, a
288 tributary of the Abukuma River, in the Fukushima Prefecture. Decontamination of a 170-m long and 8
289 to 13-m wide river section isolated from the floodplain with 2-m high concrete dikes was studied, as
290 the roads located on top of the banks were used by children to go to school. First, all the vegetation
291 was removed from the channel. Then, the top 5-cm layer of sediment was excavated from the dike
292 slopes and planted with grass. Afterwards, sediment was removed from the channel, and the removal
293 depth (between 15-35 cm) was locally adjusted depending on the vertical distribution of radiocesium
294 measured using a NaI scintillation detector before, immediately after, and then three months after the
295 remediation campaign. In addition, sediment samples were collected along transects at various depths
296 in the floodplain and analysed with coaxial germanium detectors. Radiocesium contamination strongly
297 varied with depth, depending on changes in the mud versus sand fractions, the former being enriched
298 in radiocesium. Radiocesium concentration also varied across the channel, depending on the local flow
299 velocity, which varied depending on the flood magnitude, the plant density and the microtopography.
300 Before decontamination, air dose rates 1 cm above the ground varied between 0.2–1.99 $\mu\text{Sv h}^{-1}$,
301 demonstrating the heterogeneity of contamination. After remediation, the air dose rates decreased
302 by a factor of approximately 2, from a mean of 0.78 (± 0.41) $\mu\text{Sv h}^{-1}$ before decontamination to 0.34
303 (± 0.15) $\mu\text{Sv h}^{-1}$ after decontamination at 1 cm above the ground. However, Nishikiori and Suzuki (2017)
304 underlined the risk associated with the potential deposition of contaminated material originating from
305 upstream landscapes during subsequent flood events.

306 *Forest decontamination*

307 The guidelines for the decontamination of forested areas in the Fukushima Prefecture (Table 2)
308 indicate that only those areas lying within 20 m of houses should be targeted for remediation (Yasutaka
309 and Naito, 2016) (Figure 4). Although the remediation in forests has not been a priority for the
310 Japanese authorities during the early post-accidental phase, pilot studies were conducted to quantify
311 the potential effectiveness of wider remediation programmes. Ayabe et al. (2017) investigated the
312 impact of local-scale decontamination including the removal of the litter layer, the superficial soil layer,
313 and the understorey in a secondary mixed forest with a cover of bamboo grass, *Sasa nipponica*, as
314 understorey located in Kawamata Town. Although the total ^{137}Cs contamination in soil and litter was

315 reduced by ~20% after decontamination compared to an adjacent untreated area, the radioactive
316 contamination levels returned to their initial level four months after the completion of remediation
317 works. This was likely due to the occurrence of a torrential rainfall event and the supply of
318 contaminated foliage to the ground by litterfall. These results suggest that the removal of the litter
319 and superficial soil layers in a contaminated forest may have limited effectiveness if these operations
320 are conducted too early after the initial radionuclide deposition. Decontamination should take place
321 after the peak of humus contamination occurring typically ~5 years after the initial fallout although
322 temporal variations were observed depending on the tree and humus types (Thiry et al., 2018).

323 In another study by Lopez-Vicente et al. (2018), several different forest decontamination practices
324 were compared through the monitoring of ^{137}Cs contamination in soil and leaf samples in 10 plots
325 installed in the evacuation zone, 16 km to the southwest of FDNPP, between May 2013 and July 2015
326 (i.e. 27 months monitoring). Four potential forest remediation strategies were assessed. First, the
327 combination of tree thinning and litter removal provided the best results to reduce ^{137}Cs export from
328 the plots through soil and leaf flow rates ($350\text{--}380 \text{ Bq m}^{-2} \text{ day}^{-1}$), followed by the application of tree
329 thinning only ($163\text{--}174 \text{ Bq m}^{-2} \text{ day}^{-1}$). Clearcutting and litter removal provided limited results ($92\text{--}104$
330 $\text{Bq m}^{-2} \text{ day}^{-1}$), with higher ^{137}Cs export rates than those observed from the control plots ($52 \text{ Bq m}^{-2} \text{ day}^{-1}$
331 $^{-1}$). Finally, plots where tree matting was conducted had lower ^{137}Cs export rates ($19\text{--}25 \text{ Bq m}^{-2} \text{ day}^{-1}$)
332 than control plots. Overall, the decreasing trend in radiocesium concentrations measured in the plot
333 outflow was high in 2013, moderate in 2014 and low in 2015 owing to the vegetation recovery after
334 the countermeasures.

335

336 **5. Impact of decontamination on dosimetry**

337 Two parameters are assessed before authorizing evacuees to return home: the prevalent dose rate
338 and the cumulative dose. Importantly, background radiation levels need to be incorporated into this
339 assessment. In the Fukushima Prefecture, background dose rates before the FDNPP accident were
340 estimated to be $\sim 0.04\text{--}0.05 \text{ mSv h}^{-1}$ (National Institute of Advanced Industrial Science and Technology,
341 2011).

342 Individual external radiation doses (mSv day^{-1}) may not be directly related to outdoor air doses (mSv
343 day^{-1}) as people do not spend 24 hours a day outside. When people are inside, the distances from the
344 sources of radiation are greater and walls generate a shielding effect (IAEA, 2000). In Japan, when the
345 Ministry of the Environment estimated daily individual external effective dose rates, it was assumed
346 that people spent 8 hours outdoors and 16 hours indoors, with the indoor air dose rate being 40% of
347 the outdoor air dose rate (Japanese Ministry of the Environment, 2013). Based on these assumptions,
348 the external radiation dose rate is 60% of the air dose rate. Several researches have estimated external
349 conversion coefficients based on data provided by the Ministry of the Environment (Yasutaka and
350 Naito, 2016).

351 Figure 5 compares the annual and individual dose rates that the global population may be exposed to
352 in order to help facilitate a comparison with levels in the FDNPP fallout-impacted region. In Japan, a
353 long-term dosimetric target of 1 mSv yr^{-1} was adopted by the Nuclear Emergency Response
354 Headquarters. Accordingly, a guidance value of $0.23 \mu\text{Sv h}^{-1}$ was proposed to achieve the target by
355 implementing decontamination measures. In particular, areas with ambient dose rates exceeding this
356 value were defined as ICAs. This guidance value is based on a simplified deterministic model assuming
357 that inhabitants again spend 8 hours outdoors and 16 hours indoors (i.e. a shielding factor of 0.4) per
358 day and that the contribution of natural radiation is 0.04 mSv h^{-1} (IAEA, 2013b). According to Mori et
359 al. (2017), this model has three main challenges. First, the same behavioural pattern is assumed for

360 the entire population. Second, the radiation exposure is assumed to be uniform. Third, conservative
361 assumptions are adopted when converting the ambient dose into an effective dose. For instance, the
362 time spent outside is assumed to be 8 hours, which is more than anticipated for the majority of the
363 population and likely results in an overestimation of the actual measured doses (Nomura et al., 2015).

364 Although this approach is effective for the immediate post-accidental context, more sophisticated
365 approaches are required to estimate doses over the longer term. Therefore, a probabilistic method
366 that accounts for spatial variations (i.e., houses, workplaces, and other environments) of the
367 contamination and for inter-population variations (i.e., indoor workers, outdoor workers,
368 pensioners) in behavioural patterns was developed by Mori et al. (2017). For this approach, the 95th
369 percentile doses for outdoor workers were above 1 mSv yr⁻¹ in 25 of the 59 municipalities in Fukushima
370 Prefecture (1–35 mSv yr⁻¹). In particular, the doses to more than 90% of the outdoor workers in Okuma
371 town, Futaba town, Tomioka town, Namie town, and Iitate village were over 1 mSv yr⁻¹. Furthermore,
372 the 95th percentile doses for indoor workers were above 1 mSv yr⁻¹ in Okuma town, Futaba town,
373 Tomioka town, Namie town, and Iitate village. If people return home in these municipalities, it is
374 possible that they would be exposed to doses exceeding 1 mSv yr⁻¹ for all population groups. However,
375 the results indicate that the same behavioural patterns and contamination levels should not be
376 assumed for all inhabitants nor all municipalities. Based on the different behaviour of the local
377 population, the 95th percentile doses of indoor workers and pensioners in 53 of the 59 municipalities
378 were below the dosimetric target of 1 mSv yr⁻¹ (0.026–0.73 mSv yr⁻¹) (Mori et al., 2017). Radiation dose
379 rates were also measured among different types of workers having professional activities in the village
380 of Kawauchi where the annual doses of foresters (range: 0.7–1.9 mSv yr⁻¹) were not significantly higher
381 than those of farmers (0.7–1.5), builders (0.6–1.5), office workers (0.5–1.5) and unemployed
382 individuals (0.5–1.7). In contrast, decontamination workers (0.5–7.1) were found to have significantly
383 higher dose rates (Orita et al., 2017).

384 The workers involved in decontamination activities were often directly exposed to internal irradiation
385 through inhalation, which is much more difficult to measure than the external irradiation. Accordingly,
386 eighty-three people who worked in highly contaminated areas where surface ¹³⁷Cs deposition density
387 was over 100 kBq m⁻² were enrolled in a study (Tsubokura et al., 2013). Using a database on internal
388 exposure from the Hirata Central Hospital in Fukushima Prefecture, data were compiled on age,
389 gender, body weight, equipment used in decontamination activity, total working period, duration
390 between the final working day and the day of an examination, and ¹³⁴Cs and ¹³⁷Cs body burden. Hirata
391 Central Hospital was also equipped with a permanent whole body counter with detection limits of 300
392 Bq per individual for both ¹³⁴Cs and ¹³⁷Cs measurements following a 2-min scan. The levels of internal
393 radiocesium exposure among all the decontamination workers were below the detection limits. No
394 other radionuclides besides natural ⁴⁰K were detected. No acute health problems had been reported.
395 However, levels of external exposure were not assessed, as individual data on dose rates were not
396 available. This study suggests that the resuspension of radioactive materials may cause a minimal
397 internal contamination during decontamination works (Yamaguchi et al., 2012). Other studies
398 calculated that radiation doses from internal exposure were marginal (Hayano et al., 2013; Tsubokura
399 et al., 2015). As such, remediation efforts should be concentrated on reducing the external exposure
400 of the local population.

401 According to the decontamination scenarios described in section 3, the reduction in annual individual
402 additional Effective Dose (ED) for all decontamination scenarios was 1666 person-Sv for the SDZ and
403 876-1245 person-Sv for the ICAs (Yasutaka and Naito, 2016). Despite the higher reduction rate
404 achieved in the SDZ compared to the ICAs, they remained in the same order of magnitude although
405 the decontamination efficiencies were very different in both areas. This result may be directly

406 attributed to the differences in population density in SDZ and ICAs, with 90,000 inhabitants living in
407 SDZs in 2010 versus approximately 1.5 million inhabitants living in ICAs areas exposed to over 1 mSv
408 yr⁻¹. This strong dependence of ED on population densities may lead the authorities to concentrate
409 their remediation efforts in the most densely populated areas. The results obtained also depend on
410 the effectiveness of these decontamination programmes. For instance in ICAs, where approximately 1
411 million inhabitants reside in areas exposed to 1–5 mSv yr⁻¹, the reduction in annual individual additional
412 ED was much larger in those areas where the full decontamination scenario would be implemented.

413 From the aforementioned research on river channel decontamination, the external radiation dose was
414 calculated for paths along the river used by children to go to school and the nearby playgrounds used
415 for outdoor activities incorporating an adapted time of exposition (35 h yr⁻¹ for commuting and 24 h
416 yr⁻¹ for outdoor activities) (Nishikiori and Suzuki, 2017). After decontamination of the river channel,
417 radiation dose rates decreased by a factor of approximately 2. These authors stated that the optimal
418 strategy should be to reduce the annual individual additional ED as much as possible for the whole
419 population, while also decreasing high dose individuals (Yasutaka and Naito, 2016). Indeed, the
420 authorities should not only assess the cost-benefit effectiveness of remediation programmes, they
421 must also consider ethical and social costs (Oughton et al., 2004).

422

423 **6. Treatment of decontamination waste (soil, vegetation)**

424 *Waste management*

425 The management of waste generated by the succession of catastrophes that affected the Fukushima
426 Prefecture in March 2011 has proved to be very complex, as debris derived from the earthquake, the
427 tsunami and the radioactive materials were mixed, resulting in a very atypical mixture of ‘disaster
428 waste’ (Shibata et al., 2012). Earthquake and tsunami-associated waste had elevated levels of metals
429 and metalloids (e.g., mercury, arsenic and lead), with the tsunami waste being particularly difficult to
430 manage.

431 Regarding waste contaminated with ¹³⁷Cs, the final objective is to bring radiocesium to the solution
432 phase and then enrich it, to reduce it to the smallest possible volume. In the Fukushima Prefecture,
433 the radiocesium concentrations found in the disaster waste are lower than other alkali metals.
434 Therefore, the treatment methods require approaches that help concentrate ¹³⁷Cs (Parajuli et al.,
435 2016a). The reduction of solid waste volume can be achieved through compaction or incineration. For
436 organic waste (i.e. forest litter, weeds, wood, or tree branches from contaminated areas), incineration
437 (‘thermal treatment’) is traditionally preferred (IAEA, 2003, 2006) as it reduces the volume of waste by
438 several orders of magnitude (Parajuli et al., 2013). The problem is that this ‘thermal treatment’ may
439 enrich contaminants and the Japanese legislation has a 8000 Bq kg⁻¹ radionuclide threshold for placing
440 waste in landfills (Japanese Ministry of the Environment, 2011a).

441 Accordingly, waste contaminated with radionuclide levels between 8000 Bq kg⁻¹ and 100,000 Bq kg⁻¹
442 needs to be disposed in designated landfills equipped with radiation level and leachate monitoring as
443 well as a treatment system in order to control the potential release of radioisotopes into the
444 environment (Parajuli et al., 2013). Therefore, either specially designed landfills need to be constructed
445 or pre-treatment methods need to be designed to remove radionuclides from the waste. This issue is
446 crucial as the construction of temporary storage sites and interim storage facilities were estimated to
447 account for 50% of the overall cost of decontamination. For example, transport, storage and
448 administrative costs were estimated to represent a cost of 1.55–2.12 trillion JPY (~12.4–17 billion EUR)
449 for the decontamination scenarios complying with the guidelines of Japanese authorities (Yasutaka

450 and Naito, 2016). Furthermore, securing routes and locations for transporting more than 20 million
451 tonnes of decontamination waste and removed soil that were generated to the interim storage
452 facilities remains a major challenge. Nevertheless, assessing the management and storage of low-
453 concentration radioactive cesium-containing soil and methods for using controlled landfill sites may
454 lead to a significant reduction in the amount of material requiring transport.

455 The combustible waste generated through decontamination was initially stored at temporary storage
456 facilities (Figure 6). The volume of this waste was to be reduced by incineration, and the incineration
457 ash was transferred to interim storage facilities. In 2013, the Japanese Ministry of Environment made
458 a plan stating that incineration ashes with high ^{137}Cs concentrations and leachable characteristics
459 should be stored in concrete shielded structures facilities. After being transferred to interim storage
460 facilities, incombustibles (e.g., soil) were planned to be stored at soil storage facilities in the interim
461 storage facilities (Yasutaka and Naito, 2016). The interim storage facilities are to be built in the areas
462 neighbouring the FDNPP (i.e., in Okuma and Futaba municipalities), while the temporary storage sites
463 were planned to be built in six municipalities in the SDZ (i.e., from North to South: Iitate, Minamisoma,
464 Katsurao, Namie, Tomioka and Naraha municipalities).

465 Contaminated soil removed by decontamination works is transported to an interim storage facility
466 where flammable decontamination waste (DW) is incinerated or melted to reduce its mass and
467 volume. Depending on its radiocesium content, this waste is either stored at an interim storage facility
468 or disposed in a leachate-controlled type of landfill site (Fujiwara et al., 2017). The total surface area
469 of the interim storage facilities in Futaba and Okuma municipalities is planned to cover ~1600 ha, and
470 by February 2019, a contract was already established between Japanese authorities and landowners
471 for ~70% of the land required for storage. Soil storage operations started in October 2017 in Okuma
472 and in December 2017 in Futaba. By March 2019, ~2.5 million m^3 of waste soil had already been
473 transported from the temporary storage facilities distributed across all the remediated area to these
474 two interim storage facilities (Japanese Ministry of the Environment, 2019b). All the soil waste is
475 planned to be transported to the Okuma and Futaba sites by the end of 2021 (Japanese Ministry of the
476 Environment, 2019a). The final disposal of this decontamination waste should take place outside of
477 the Fukushima Prefecture, within 30 years after the opening of the interim storage facilities (i.e.,
478 ~2047).

479 *Incineration*

480 The temperature used in the furnaces used for incineration of radioactive waste is similar to that used
481 in the plants treating municipal waste (870–882°C). The incinerators for radioactive waste are
482 radiation-controlled areas, with workers following protocols in accordance with the Ordinance on
483 Prevention of Ionizing Radiation Hazards (Act No. 134 of the 2015 amendment of Law No. 41 of the
484 Japanese Ministry of Labour in 1972). The heavier particles are collected at the bottom of the furnace,
485 generating the so-called bottom ash (BA), while the lighter particles pass to a bag-filter where the
486 temperatures are kept lower (250–300°C) and where the so-called fly ash (FA) and the vaporised
487 cesium are collected (Figure 7). The exhaust gas is filtered to trap the residual fine particles, generating
488 several types of FA. Measurements made on incineration products since 2015 showed that BA and FA
489 are produced with similar levels of radiocesium, both with low radiocesium leachability (<1%) (Fujiwara
490 et al., 2017). Radiocesium levels in the exhaust gases were found to be lower than method detection
491 limits (Parajuli et al., 2013).

492

493

494 *Incineration ash treatment*

495 The chemical form and the leachability of radiocesium depends on the type of waste incinerated.
496 Results observed for three different types of ash samples suggest that ^{137}Cs along with other alkali
497 metals in wood bark and household garbage ashes, originated from burnable materials, were mostly
498 washed out with water even at ambient temperatures. However, municipal sewer sludge was
499 different, with potential ^{137}Cs elution only occurring under very specific conditions (i.e., with acid
500 treatment and under high temperatures). Acid treatment at high temperatures was found to be
501 inappropriate for treating wood bark and household garbage ashes because of the generation of a Ca
502 excess leading to gypsum formation and complexifying the subsequent treatment process (Parajuli et
503 al., 2013).

504 *Soil recycling*

505 As 22 million m^3 of decontamination soil (i.e. 90% of the total) and incineration ash waste (10%) is
506 expected to be produced through remediation of the fallout impacted region, recycling may be
507 instrumental for reducing this volume (Takai et al., 2018). The Japanese Ministry of Environment
508 developed a policy to separate decontamination soil into low- and high-activity soils, the former being
509 'recycled' in public projects. In these uses, decontamination soil will be used for the basic structure
510 and will be covered by uncontaminated soil or concrete. In theory, the unconditional 'clearance level'
511 defined by IAEA for the use of recycled material is fixed to 100 Bq kg^{-1} for radiocesium. However, as
512 disaster waste was found with higher ^{137}Cs levels, the Japanese Ministry of Environment decided that
513 those materials with radiocesium levels up to 3000 Bq kg^{-1} can be reused at a minimum depth of 30
514 cm underground (reference level assessed for recycling of concrete for the road subbase course). For
515 decontamination soil recycling, the radioactivity level had to be reanalysed for a different type of
516 engineering structures (deterministic estimation of radiation dose rates). The corresponding level of
517 radiocesium concentrations in the soil was estimated to 6000 Bq kg^{-1} . To confine doses to levels below
518 $10 \mu\text{Sv yr}^{-1}$ based on the derived radioactivity level, an additional layer of soil slope protection of 40
519 cm or more was needed. Accordingly, the Japanese Ministry of Environment determined the maximum
520 radioactivity level to be $6,000 \text{ Bq kg}^{-1}$ for embankments covered with 50 cm uncontaminated soil.
521 Overall, the recycling of decontaminated soil is limited to civil engineering structures in public projects,
522 such as road embankments and coastal levees. Takai et al. (2018) evaluated the associated additional
523 doses to workers and the public using these structures and demonstrated that additional dose rates
524 would remain below the 1 mSv yr^{-1} threshold corresponding to 6000 Bq kg^{-1} .

525 In Japan, contaminated wastes are disposed under the standard of $8,000 \text{ Bq kg}^{-1}$. The volume of
526 decontamination soil having a radioactivity concentration of $8,000 \text{ Bq kg}^{-1}$ or below is estimated to be
527 approximately 10 million m^3 , which corresponds to half of the total amount of decontamination soil
528 generated. The radioactivity concentration of $8,000 \text{ Bq kg}^{-1}$ will decrease to $6,000 \text{ Bq kg}^{-1}$ in 5 years.
529 Therefore, more than half of the total decontamination soil should become recyclable in at least 5
530 years. Through the use of pre-treatment activities, such as classification processing, even more
531 decontaminated soil may become recyclable in the non-too distant future (Takai et al., 2018).

532 *Soil remediation*

533 Remediation of contaminated soil based on a hot acid treatment was tested for the two most common
534 soil groups found in Fukushima (Parajuli et al., 2016b): Cambisols (i.e. brown forest soils) and Andisols
535 (i.e. soils developed on volcanic ash). Although this method was shown to be effective for the former
536 soil type, this was not the case for the latter. In particular, lime must be added to readjust the pH of
537 Andisols after their treatment with acid, and the soil must be mixed with untreated and
538 uncontaminated soil prior to being reused for cultivation. Furthermore, to avoid the transfer of

539 residual radiocesium to plants, additives such as zeolite or Prussian blue adsorbents need to be
540 incorporated into the Andisols. The problem associated with this strategy is that, through their ageing,
541 zeolites may increase ^{137}Cs exchangeability with potassium and accelerate ^{137}Cs transfer to the
542 cultivated plants over longer time periods (Yamaguchi et al., 2019). These restrictions illustrate the
543 difficulty of finding alternatives to the storage of decontamination soil waste in interim facilities.

544

545 **7. Perspectives for future research**

546 The total estimated decontamination cost would exceed 16 trillion JPY (~128 billion EUR) if all forested
547 areas exposed to radiation dose rates exceeding 1 mSv yr^{-1} were decontaminated. However,
548 decontaminating all of the forested areas would not result in a major ED reduction for the average
549 inhabitant (Yasutaka and Naito, 2016). As almost 70% of the surface area of Fukushima Prefecture is
550 covered with forests (Hashimoto et al., 2012) and forestry is a significant economic activity in the
551 region, future research should prioritize investigating radiocesium dynamics in these regions. In
552 particular, the biological cycling of ^{137}Cs in forests has now been affected by the decomposition of litter
553 where radiocesium was concentrated shortly after the FDNPP accident (Koarashi et al., 2012).
554 Furthermore, the local population in rural areas of the Fukushima Prefecture enjoy 'satoyama', or the
555 collection of vegetation, including mushrooms, edible wild plants, and firewood from forested
556 landscapes (Prand-Stritzko and Steinhäuser, 2018; Nihei, 2016). The collection and shipping of
557 mushrooms remains prohibited in the main fallout-affected areas (Fukushima Prefecture, 2018).
558 Furthermore, the low permissible levels for radiocesium contamination in wood (e.g. 40 Bq kg^{-1} in
559 wood for cooking or eating) will restrict the use of this commodity for at least several decades in the
560 region (Ohashi et al., 2017). In addition, approximately 1800 workers are employed by the forest
561 industry in the region (Yasutaka and Naito, 2016). For many of the local inhabitants, the forest, the
562 satoyama, and its harvest are inseparable from their daily lives.

563 Forest sources were also shown to deliver a significant proportion of contaminated material to the
564 river systems draining the fallout-impact region. The analysis of deposited particulate matter collected
565 in three fallout-contaminated coastal catchments between November 2012 and November 2014
566 demonstrated that forest sources supplied a mean of 17 % (standard deviation, SD, 10 %) of the
567 sediment transiting these river systems (Lacey et al., 2016). Huon et al. (2018) obtained similar results
568 through the analysis of sediment cores collected between November 2014 and April 2015 in a dam
569 reservoir draining fallout-impacted cultivated and forested landscapes, with the latter supplying a
570 mean of 27% (SD, 6%) of the material deposited in the lake. These conclusions were validated through
571 an analysis of a larger number of sediment samples ($n=400$) collected in coastal river systems in the
572 Fukushima region over a longer time period (from November 2011 to November 2017) where a mean
573 of 24% (SD, 21%) of the material transiting these systems was modelled to be derived from forested
574 landscapes (Evrard et al., 2019). Cumulatively, these results demonstrate that forested landscapes
575 represent a potential long-term source of particulate contaminated matter that likely will require
576 diligent management for the foreseeable future.

577 In cultivated landscapes where the remediation activities were concentrated, the main question is
578 whether or not to restart agricultural production. The removal of the topsoil layer concentrating the
579 radiocesium, the replacement of this material with crushed granite extracted from local quarries and
580 the final mixing of the entire profile to prepare the soils for re-cultivation raises several important
581 questions. For example, to what extent will the residual radiocesium in the soil be transferrable to the
582 plants cultivated on these soils? How will the crushed granite, which was homogenized into the soil,
583 affect the soils fertility? Recent research showed that potassium fertilization is required to maintain
584 productivity when restarting cultivation after decontamination (Kurokawa et al., 2019). Indeed, as it

585 was demonstrated in the current literature review, the reopening of the region after the completion
586 of remediation activities represents a unique situation in history, coupled with unprecedented
587 challenges that require further ongoing investigations.

588 Although previous dosimetric studies demonstrated that currently the internal exposure of both the
589 local population and the decontamination workers remains minimal, both internal and external
590 exposures of these groups should be studied over longer temporal periods to help understand the
591 long-term impacts of this accident on exposed population groups. More research is also required to
592 understand the fate and dynamics of other longer-lived radionuclides in the Fukushima region
593 including radiocarbon (Paterne et al., 2018; Povinec et al., 2016; Xu et al., 2016), plutonium and uranium
594 isotopes (Jaegler et al., 2018; Zheng et al., 2013; Steinhauser, 2014) as they may be persistent in the
595 environment even though many were emitted only at the trace and ultra-trace levels.

596 **Conclusions**

597 The quick and early decision of the Japanese authorities to decontaminate FDNPP fallout impacted
598 landscapes was unprecedented. Decontamination activities were rapidly implemented in agricultural
599 and residential areas covering a surface of ~9000 km². These remediation activities produced ~20
600 million m³ of soil waste in less than 6 years (2013–2019) with an approximate cost of 3 trillion JPY (~24
601 billion EUR). The strategy of removing the surface layer of the soil concentrating ¹³⁷Cs was shown to be
602 effective in cultivated land when the strategy was applied at a catchment scale to avoid the supply of
603 mobilized contamination from the headwaters. The main current challenges are associated with the
604 treatment and the transport of this waste to the interim storage facilities for the next ~30 years that
605 are being built near the FDNPP. The re-cultivation of the soils after decontamination also raises several
606 concerns. In particular, more information is required regarding soil fertility after decontamination and
607 the potential transfer of the residual ¹³⁷Cs to the plants cultivated on decontaminated fields.

608 The risks of internal and external radiation dose exposures of the decontamination workers and the
609 local population to exceed the target of 1mSv yr⁻¹ appeared to be low during the early post-accidental
610 phase. However, dosimetric monitoring programmes should be carried out to confirm this result over
611 the longer term, particularly after local population returns to the region as a risk of internal
612 contamination remains if these inhabitants consume local food. Furthermore, as ~75% of the surface
613 exposed to the highest ¹³⁷Cs fallout levels in the Fukushima Prefecture are covered with forests where
614 decontamination was not implemented, the potential long-lasting contribution of radiocesium to the
615 river systems draining these mountainous, forested landscapes exposed to typhoons should be
616 investigated. The behaviour and the dynamics of longer-lived radionuclides such as plutonium isotopes
617 remains poorly documented and they should also be studied in the future as they may persist in the
618 environment for long timescales even though they were emitted at trace and ultra-trace levels.

619 **Data availability**

620 All the data provided in this review paper can be accessed directly in the referred publications or URL.

621 **Competing interests**

622 The authors declare that they have no conflict of interest.

623 **Supplement**

624 A kmz file with the location of the Intensive Contamination Survey Areas (ICAs) in Japan is provided.

625

626 **Acknowledgements**

627 This research was funded by the AMORAD project (ANR-11-RSNR-0002), supported by the French
628 National Research Agency (ANR, Agence Nationale de la Recherche, Programme des Investissements
629 d'Avenir). The support of CNRS (Centre National de la Recherche Scientifique, France) and JSPS
630 (Japan Society for the Promotion of Science) in the framework of the Franco-Japanese collaboration
631 project framework (PRC, CNRS-JSPS) is also gratefully acknowledged.

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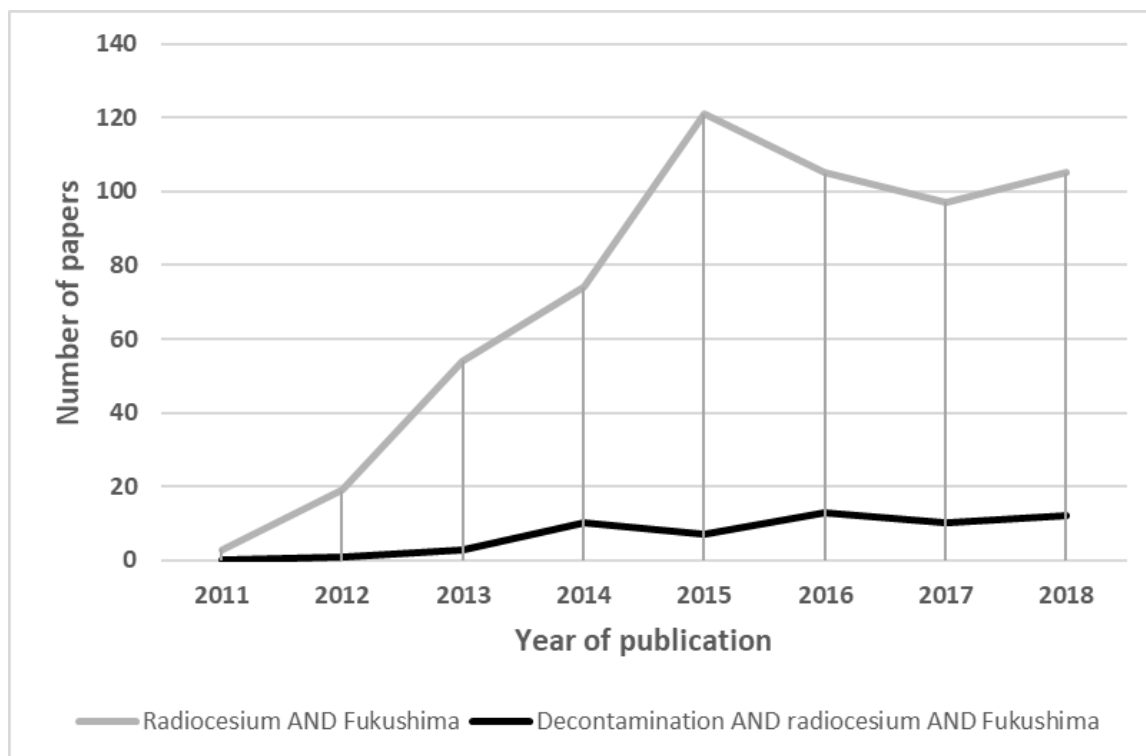
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931 **Figures**

932 Figure 1. Evolution of the number of studies published on radiocesium and Fukushima (including or
933 not a reference to decontamination) in the literature between 2011–2018, according to the Scopus
934 search engine.

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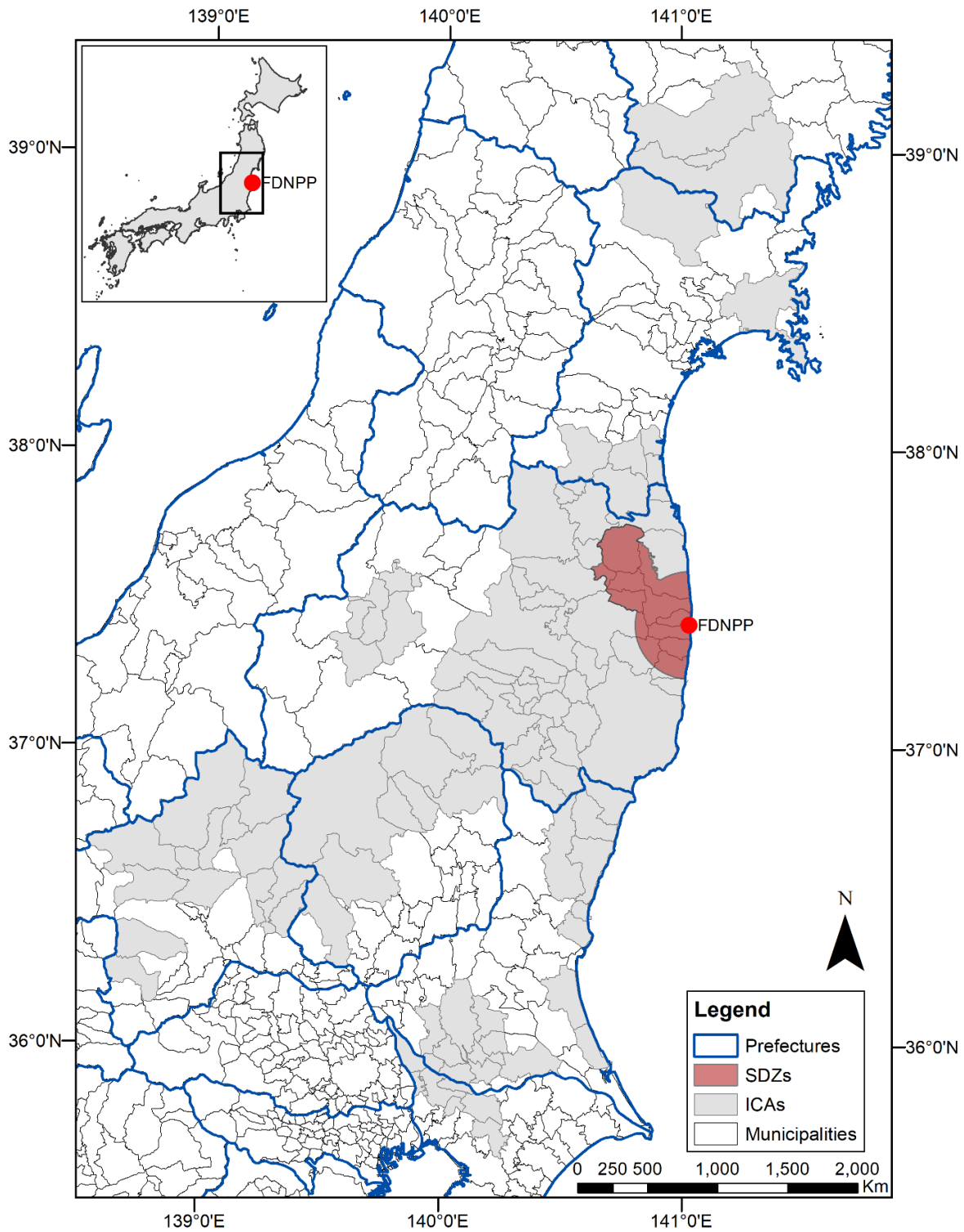
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950 Figure 2. Location of the Fukushima Prefecture in Japan (Inset Map) and the location of the Special
951 Decontamination Zones (SDZs) and the Intensive Contamination Survey Areas (ICAs).

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954 Figure 3. Illustration of the different steps of remediation activities in cultivated land in Fukushima: (a)
955 removal of the 5-cm topsoil layer concentrating most of the radiocesium (November 2013); (b)
956 addition of a crushed granite layer on top of the residual soil profile (May 2014); (c) final mixing of the
957 entire profile to prepare re-cultivation (March 2019). Pictures were taken by the authors in the litate
958 Village.



959 (a)



960 (b)



(c)

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964 Figure 4. Illustration of the 20-m buffer zone decontaminated in forested areas in the vicinity of houses.

965 Example from Iitate Village (Sasu district).

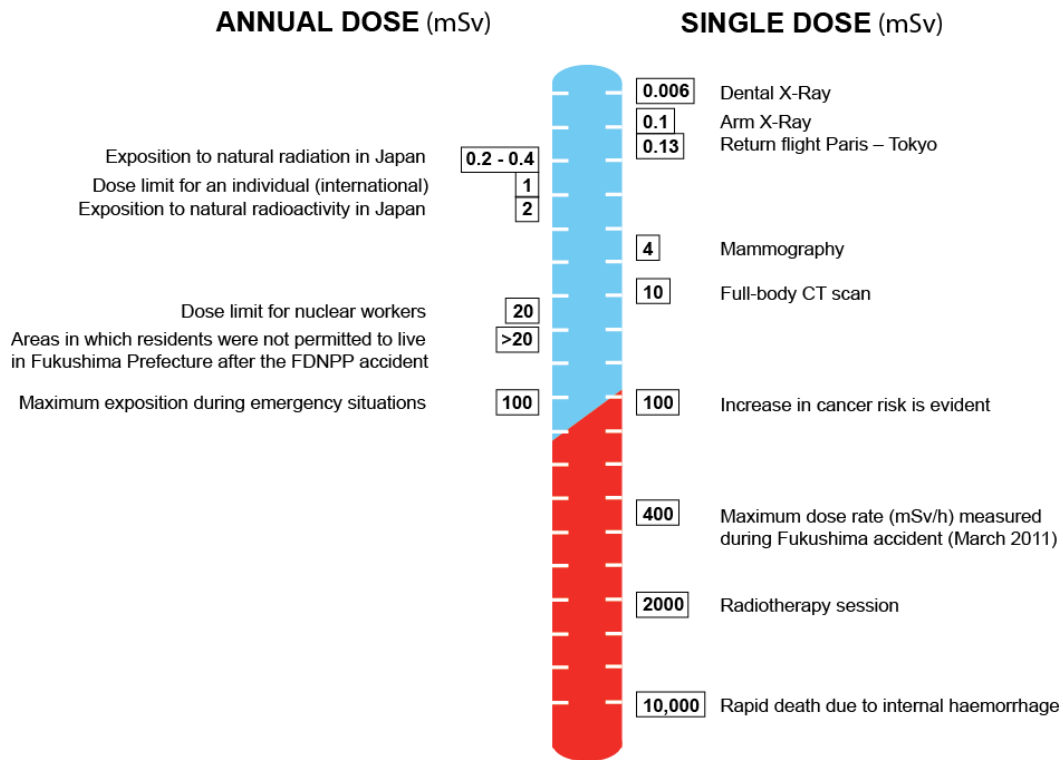


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969 Figure 5. Comparison of annual and individual radioactive dose rates to which the population may be
 970 exposed, based on a compilation of data (Commissariat à l'Energie Atomique et aux Energies
 971 Alternatives, 2016;Harada et al., 2014).



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988 Figure 6. Temporary storage facilities for radioactive waste in Iitate Village, in the Fukushima
989 Prefecture.



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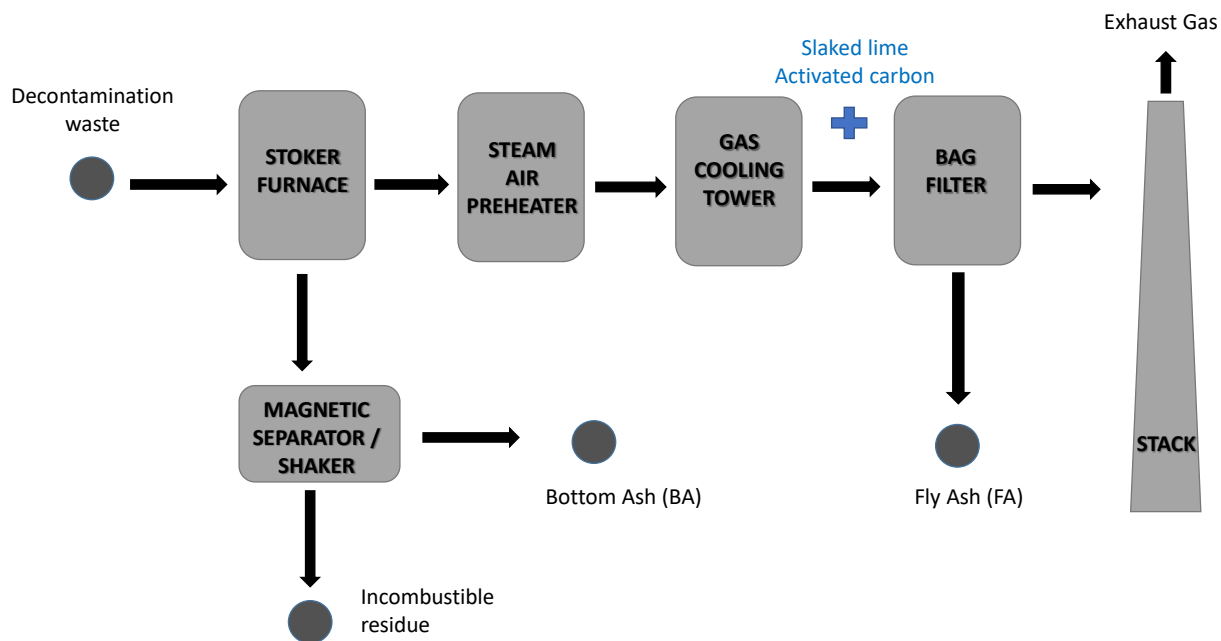
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995 Figure 7. Simplified diagram showing the functioning of an incineration plant treating decontamination waste in Fukushima, modified after Parajuli et al. (2013) and Fujiwara et al. (2017).
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Tables

Table 1. Selection of official websites from the Japanese authorities providing information on the remediation works and their impact. (Last access to these websites on March 8, 2019).

Authority	Type of information	Webpage
Fukushima Prefecture	Environmental restoration	https://www.pref.fukushima.lg.jp/site/portal-english/list382.html
Ministry of Environment	Environmental Remediation	http://josen.env.go.jp/en/
Ministry of Economy, Trade and Industry	Fukushima Today webpage	http://www.meti.go.jp/english/earthquake/index.html
Nuclear Regulation Authority	Monitoring information	https://radioactivity.nsr.go.jp/en/

1 Table 2. Unit costs and effectiveness of decontamination measures implemented in the SDZ and ICAs after Yasutaka and Naito (2016).

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3	Code	Measure	Effectiveness range	Unit cost 10kJPY/ha (EUR)	Nb containers/ha	Target area
4						
5						
6	Agricultural land					
7	A1	Cut weeds/remove 5cm topsoil/cover soil	0.34 – 0.80	950 (7600)	815	SDZ
8	A2	Cut weeds/remove 5 cm topsoil	0.34 – 0.80	625 (5000)	815	SDZ
9	A3	Interchange topsoil and subsoil/add zeolite – K	0.34 – 0.80	310 (2500)	0	SDZ and ICA
10	A4	Ploughing with zeolite and K	0.21 – 0.50	33 (265)	0	SDZ and ICA
11	Forest					
12	F1	Remove litter and humus	0.19 – 0.59	745 (6000)	530	SDZ
13	F2	Remove litter	0.10 – 0.30	280 (2250)	260	ICA
14	Roads					
15	R1	Shot-blasting, cleaning ditches	0.15 – 0.66	480 (4000)	30	SDZ
16	R2	Cleaning roads and ditches	0.08 – 0.33	240/km (2000)	88	ICA
17	Buildings					
18	B1	Full decontamination	0.29 – 0.70	1750–3500	150	SDZ and ICA
19				(14,000–30,000)		
20	B2	Local decontamination	0.15 – 0.35	125–250	11	ICA
21				(1000–2000)		

22 Table 3. Unit costs estimated for waste management and storage after Yasutaka and Naito (2016) in the ICAs.

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24 Measure	Unit cost (JPY)	Unit cost (EUR)
26 Storage container	8000	65
27 Transport from decontamination site to temporary storage site	3100 / container	25 / container
28 Temporary storage site	20,000 / container	160 /container
29 Transport from temporary storage site to interim storage facility	3800 – 16,000 / container	30 – 130 /container
30 <i>Treatment at interim storage facility</i>		
31 Combustible volume reduction	2000 / container	16 /container
32 Storage of combustible incineration residue	100,000 / container	800 /container
33 Storage of incombustibles	30,000 / container	240 /container

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