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## ► To cite this version:

O. Evrard, J. Patrick Laceby, Atsushi Nakao. Strategies and effectiveness of land decontamination in the region affected by radioactive fallout from the Fukushima nuclear accident: A review: SOIL Discussions. 2019. cea-02812441

**HAL Id: cea-02812441**

**<https://cea.hal.science/cea-02812441>**

Preprint submitted on 6 Jun 2020

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1 **Strategies and effectiveness of land decontamination in the region affected by radioactive fallout**  
2 **from the Fukushima nuclear accident: A review**

3 **Olivier Evrard<sup>1</sup>, J. Patrick Lacey<sup>2</sup>, Atsushi Nakao<sup>3</sup>**

4 <sup>1</sup>Laboratoire des Sciences du Climat et de l'Environnement (LSCE/IPSL), Unité Mixte de Recherche 8212 (CEA/  
5 CNRS/UVSQ), Université Paris-Saclay, Gif-sur-Yvette, France.

6 <sup>2</sup>Environmental Monitoring and Science Division (EMSD), Alberta Environment and Parks (AEP), Calgary, Alberta,  
7 Canada.

8 <sup>3</sup>Graduate School of Life and Environmental Sciences, Kyoto Prefectural University, Kyoto, Japan.

9  
10 *Correspondence to:* Olivier Evrard (olivier.evrard@lsce.ipsl.fr).

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 km<sup>2</sup>) and for the 40 non-evacuated municipalities affected by  
20 lower, although still significant, levels of radioactivity (Intensive Contamination Survey Areas, 7836  
21 km<sup>2</sup>). 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<sup>-1</sup>. Decontamination activities generated ~20 million m<sup>3</sup> 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; nuclear accident; 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 Airborne and ground contamination surveys demonstrated that the contamination was the highest  
47 (i.e., initial  $^{137}\text{Cs}$  levels  $>100,000 \text{ Bq m}^{-2}$ ) in a plume extending to the northwest of the FDNPP covering  
48 an area of  $\sim 3000 \text{ km}^2$  (Kinoshita et al., 2011;Chartin et al., 2013;Yasunari et al., 2011). Although many  
49 radioactive substances were released into the environment by the FDNPP accident, radiocesium (i.e.,  
50  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) presents the most serious risk to the local population over the medium to long term as  
51 it was emitted in very large quantities and it has a relatively long half-life (i.e.,  $^{134}\text{Cs}$  - 2 years;  $^{137}\text{Cs}$  - 30  
52 years) (Steinhauser et al., 2014).

53 Numerous investigations have been conducted by Japanese and international researchers to improve  
54 our understanding of the fate of radiocesium in the Fukushima region (for a review, see: Evrard et al.,  
55 2015). In general, radiocesium sorption mechanisms were characterised (Fan et al., 2014;Nakao et al.,  
56 2015) and their fluxes measured in riverine systems draining the main radioactive plume (Nagao et al.,  
57 2013). Land use (Koarashi et al., 2012) and soil properties (Nakao et al., 2014) were shown to control  
58 the migration of radiocesium in soils. Accordingly, the fate of this contaminant was intensively  
59 investigated in forest ecosystems (Gonze and Calmon, 2017) and cultivated landscapes (Yoshimura et  
60 al., 2016), which are the two main land uses in the fallout-impacted region. Typhoons and other major  
61 rainfall events were also demonstrated to drive soil erosion and sediment migration processes thus  
62 directly influencing post-fallout radiocesium dynamics (Chartin et al., 2017).

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

75 Although radiocesium is mainly transported in particle-bound form in the Fukushima fallout-impact  
76 area (Konoplev et al., 2016), dissolved radiocesium was found in numerous environmental  
77 compartments, primarily during the immediate post-accidental phase (Yoshimura et al., 2014). As most  
78 of the dissolved radiocesium migrated through these landscapes immediately after the FDNPP  
79 accident, this literature review will focus on particulate radiocesium. Furthermore, as  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$   
80 were emitted in equivalent proportions into the environment in March 2011, with an initial  $^{134}\text{Cs}/^{137}\text{Cs}$   
81 activity ratio of  $\sim 1$  (Kobayashi et al., 2017), this review will focus primarily on  $^{137}\text{Cs}$  owing to its longer  
82 half-life and thus greater risk to the local population over the medium to long-term. Accordingly, the  
83 goal of this review is to examine the remediation strategies and their effectiveness for particulate  
84 bound  $^{137}\text{Cs}$  in Japan.

85 This literature review will be divided into five main sections. First, the spatial extent of the  
86 decontaminated zone and the schedule of these remediation activities will be outlined. Second, the  
87 remediation strategies in different environments (i.e., farmland, river, forests) will be presented along



88 with a summary of their cost effectiveness. Third, the impacts of remediation activities on dosimetry  
89 will be summarized. Fourth, the initiatives to manage the large volume of waste generated by  
90 remediation will be discussed. Fifth, major research questions and requirements to guide the future  
91 management of Fukushima fallout-impacted areas will be identified and presented. The objective of  
92 this review is to provide a synthesis of the remediation lessons learnt in Japan following the FDNPP  
93 nuclear accident, which are fundamental in light of the potential for a similar catastrophe to occur  
94 somewhere on Earth in the future (Christoudias et al., 2014).

95

## 96 **2. Areas targeted by decontamination**

97 In November 2011, the Japanese government adopted the *Act on Special Measures Concerning the*  
98 *Handling of Pollution by Radioactive Materials* (Japanese Ministry of the Environment, 2011a) in order  
99 to reduce the impact of radioactive substances from the FDNPP accident on human health and the  
100 environment (Yasutaka and Naito, 2016). In support of this Act, decontamination guidelines were  
101 released by the Japanese Ministry of Environment in December 2011 and updated in 2013. These  
102 guidelines outlined the methods for surveying and quantifying the levels of contamination and the way  
103 to prepare these areas targeted for remediation (Japanese Ministry of the Environment, 2013). A  
104 decontamination roadmap (*Policy for Decontamination in the Special Decontamination Area*) was  
105 implemented in January 2012 under the direct supervision of the Japanese government.

106 According to the decontamination roadmap, the remediation programme had to be implemented in  
107 'special areas' where targets were set for the exposure of the public to external dose rates in order for  
108 residents to return to their day-to-day lives (Yasutaka and Naito, 2016). Achieving pre-accident  
109 radiation levels is not the objective, rather the effectiveness of decontamination will ultimately depend  
110 upon the land use and the air dose of each particular area. Two zones were delineated with different  
111 strategies for remediation (Figure 2). First, Special Decontamination Zones (SDZ) are areas located  
112 within a 20-km radius of the FDNPP or areas where the cumulative dose one year after the accident  
113 was expected to exceed 20 mSv yr<sup>-1</sup>. SDZs occur in 11 municipalities (1117 km<sup>2</sup>) where residents were  
114 evacuated after the FDNPP accident in 2011. The central government of Japan is responsible for  
115 remediation works in SDZs. Second, Intensive Contamination Survey Areas (ICAs) refer to 102  
116 municipalities from eight Prefectures with ambient dose rates exceeding 0.23 μSv h<sup>-1</sup>, designated as  
117 ICAs by the Ministry of Environment on December 28, 2011 (Mori et al., 2017). The area of the ICAs is  
118 eight times greater than the SDZ (Yasutaka and Naito, 2016). In particular, the decontamination  
119 methods and target areas for remediation in the ICAs differ from those of the SDZ with  
120 decontamination activities for the ICAs conducted by local governments with support from the central  
121 government. In total, the SDZs and ICAs cover a surface area of 8953 km<sup>2</sup> with a population that was  
122 not evacuated after the accident of ~1.7 million (Yasutaka and Naito, 2016).

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

128

129

130



### 131 3. Decontamination Strategies and their Cost Effectiveness

132 The effectiveness of decontamination was assumed to strongly vary depending on the remediation  
133 method and the initial radiation dose rates prior to decontamination. Different remediation  
134 techniques were proposed depending on the land use and the zone (i.e., SDZ vs. ICA). Yasutaka et al.  
135 (2013a) and Yasutaka et al. (2013b) compared the impact of four scenarios of decontamination,  
136 including two very unlikely options (i.e., minimal and maximal scenarios), in terms of effectiveness and  
137 cost according to the results of demonstration tests conducted by JAEA. These results were updated  
138 in a more recent publication (Yasutaka and Naito, 2016). Only the two scenarios following the  
139 guidelines provided by the Japanese government are assessed in this review. As such, the results of  
140 both the minimal and maximal remediation options are therefore not discussed. In the first scenario,  
141 5 cm of topsoil was removed from 50% of agricultural land in the SDZ where  $^{137}\text{Cs}$  concentrations  
142 exceed 5000 Bq kg<sup>-1</sup>, and replaced with a 5-cm layer of 'clean' soil (measure A1; Table 2). In the  
143 remaining 50% of agricultural land with  $^{137}\text{Cs}$  concentrations below 5000 Bq kg<sup>-1</sup>, topsoil was  
144 interchanged with subsoil (measure A3). Ploughing with zeolite and potassium (measure A4) was  
145 adopted for agricultural land in ICAs where the annual additional effective dose exceeded 1 mSv yr<sup>-1</sup>.  
146 In the second scenario, measure A1 was applied to all cultivated land. Similar measures (e.g. Table 2)  
147 were included in both scenarios to decontaminate forested areas, roads and houses. The total  
148 decontamination cost for implementing these remediation measures varied between 2–5.1 trillion JPY  
149 (~16–41 billion EUR), with 1.3–2 trillion JPY (~10–16 billion EUR) for the SDZ and 0.7–3.1 (~6–25 billion  
150 EUR) for the ICAs. Although the area where decontamination has been implemented in the ICAs (922–  
151 3330 km<sup>2</sup>) covers a surface 3 to 11 times larger than that of SDZ (295 km<sup>2</sup>), the decontamination costs  
152 for the SDZ and ICAs are in the same order of magnitude.

153 The decontamination program includes a variety of other activities on top of the actual on-site  
154 remediation works, including the transport of waste, the volume reduction of waste along with the  
155 temporary, the interim, and the final storage of decontamination waste and removed soil in  
156 containers. Depending on the set of measures implemented in the field, the cost of the remediation  
157 works will therefore be highly variable. A synthesis of the unit costs for waste management and storage  
158 is provided in Table 3 (Yasutaka and Naito, 2016). As shown by Yasutaka et al. (2013b), the quantity of  
159 waste generated when decontaminating agricultural land varies considerably depending on the  
160 decontamination method used. Consequently, differences in the quantity of waste generated resulted  
161 in large differences between agricultural land decontamination methods and the costs associated with  
162 storage containers, temporary storage sites and interim storage facilities.

163 According to the latest available figures from the Japanese Ministry of the Environment (2019b) at the  
164 end of 2018, the volume of soil waste generated in the SDZ was 9,100,000 m<sup>3</sup> with a remediation cost  
165 of approximately 1.5 trillion JPY (~12 billion EUR). In the ICAs, the latest figures available for March  
166 2018 showed that 7,900,000 m<sup>3</sup> of waste soil were produced with a remediation cost of approximately  
167 1.4 trillion JPY, equivalent to ~11 billion EUR (Japanese Ministry of the Environment, 2019a).

168

### 169 4. Strategies for decontamination in various environments, and their effectiveness

170 In general, owing to the strong and nearly irreversible bond of radiocesium to fine soil particles, the  
171 majority of FDNPP derived  $^{137}\text{Cs}$  is stored within the topsoil (i.e. the top 5 cm) in undisturbed soils  
172 (Lepage et al., 2014; Matsunaga et al., 2013; Takahashi et al., 2015; Mishra et al., 2015; Matsuda et al.,  
173 2015). Mishra et al. (2015) reported that for these undisturbed soils, the vertical migration of  
174 radiocesium down the soil profile was slower in forest soils compared to grassland soils. In disturbed  
175 soils, anthropogenic activities may increase the depth migration of radiocesium down the soil profile



176 (Lepage et al., 2015;Matsunaga et al., 2013). For example, Lepage et al. (2015) illustrated that 90% of  
177 the FDNPP derived radiocesium was homogeneous throughout the tilled soil layer in cultivated soils.  
178 Endo et al. (2013) reported that radiocesium concentrations were not depth dependent in cultivated  
179 soils (i.e. paddy fields) whereas they declined exponentially in uncultivated soil. Both Sakai et al.  
180 (2014b) and Tanaka et al. (2013) also demonstrated that radiocesium from the FDNPP accident was  
181 measureable at 15cm depth in rice paddy fields. As illustrated by Koarashi et al. (2012), the penetration  
182 of radiocesium in the soil differed depending on both the land use and the physicochemical properties  
183 of the soil (e.g., bulk density, clay content and organic matter content). However, remediation  
184 strategies consisting in removing the top 5 cm layer of the soil should have been effective as cultivation  
185 or other human activities that may have led to the redistribution of radiocesium with depth after the  
186 accident were prohibited in the main fallout impacted region.

#### 187 *Soil/farmland decontamination*

188 Different strategies were carried out in Japan to decontaminate soil in farmland, either removing the  
189 contaminated layer of soil or through the sowing of plants having the capacity of extracting and  
190 concentrating radiocesium from the soil. There are few publications in international journals regarding  
191 the potential or effectiveness of the latter strategy (e.g. Pareniuk et al., 2015). Among the few available  
192 studies, Kobayashi et al. (2014) grew thirteen plant species from three families (*Asteraceae*, *Fabaceae*,  
193 and *Poaceae*) in shallow and deeply cultivated fields where the 0–8 cm and 0–15 cm soil layers were  
194 respectively ploughed. The variation in plough depth was expected to reflect the impact of different  
195 contact zones between the root systems and radiocesium in the soil. Overall, 29 to 225 Bq kg<sup>-1</sup> dry  
196 weight of <sup>137</sup>Cs were found in the plants, corresponding to transfer factors ranging from 0.019 to 0.13  
197 (geometric mean (GM), 0.057) for plants growing in shallow soils, and from 0.022 to 0.13 (GM 0.063)  
198 for plants growing in deeper soils (Kobayashi et al., 2014). The authors found that none of their tested  
199 plant species resulted in a significant decrease in radiocesium in soil likely because of the strong  
200 fixation of <sup>137</sup>Cs to clay particles. This result was confirmed by Yamashita et al. (2014) who showed that  
201 99 wild plants grown in paddy and upland fields had a very low phytoextraction efficiency. Tamaoki et  
202 al. (2016) reached the same conclusions, although they suggested *Kochia* (*Bassia scoparia*) as a  
203 potential candidate for phytoremediation although its efficiency in removing <sup>137</sup>Cs would require  
204 numerous cultivation rounds.

205

206 Accordingly, given the low efficiency of phytoremediation, the main remediation strategy consists of  
207 removing the surface layer of soils with the majority of radiocesium. The effectiveness of this strategy  
208 was examined by Sakai et al. (2014a) in Kawamata Town. Approximately 5-10 cm of the surface soil  
209 was removed from one rice paddy by heavy machinery, whereas a nearby paddy field was not  
210 decontaminated and used as control plot. Both of these paddies then were ploughed and planted with  
211 rice. Five surface soil samples (0-5 cm) were collected after decontamination and prior to ploughing  
212 on June 12, 2011. Thereafter five soil cores (20 cm depth) were collected on July 13, 2012 at 3 m  
213 intervals across both rice paddy fields. In 2011, the accumulation of radiocesium in the 0-5 cm surface  
214 layer of the soil in the decontaminated paddy field (170±64 Bq kg<sup>-1</sup>) was lower than the control rice  
215 paddy field (2231 ±64 Bq kg<sup>-1</sup>). However, the <sup>137</sup>Cs concentration of the surface soil layer in the  
216 decontaminated rice paddy field (753±62 Bq kg<sup>-1</sup>) was significantly higher in 2012 than in 2011. This  
217 result suggests that radiocesium is likely redistributed through the rice paddy field irrigation and  
218 drainage networks. The authors concluded that the redistribution of soil within the paddy fields may  
219 decrease the effectiveness of decontamination. A lack of replicates was outlined by the authors and  
220 prevented them to finally conclude on the effectiveness of surface removal for decontamination (Sakai  
221 et al., 2014a). In contrast, Kurokawa et al. (2019) observed a 80% decrease in <sup>137</sup>Cs activities after



222 decontamination in cultivated land of Tomioka Town, showing the efficiency of this remediation  
223 strategy.

224

225 Another study was conducted in experimental paddy fields located ~40 km from the FDNPP (Wakahara  
226 et al., 2014). Two plots were established: a paddy field where the top 5–10 cm of soil was removed  
227 before cultivation and a control paddy. The  $^{137}\text{Cs}$  soil inventory measured 3 months after the FDNPP  
228 accident was approximately 200,000 Bq  $\text{m}^{-2}$ . However, after decontamination, this inventory  
229 decreased to 5000 Bq  $\text{m}^{-2}$ . Suspended sediment and  $^{137}\text{Cs}$  fluxes were measured in the outflow of the  
230 paddy fields after puddling (i.e., the mixing of soil and water before planting rice) and they were 11.0  
231 kg and 630,000 Bq (1240 Bq  $\text{m}^{-2}$ ) respectively in the control paddy, versus 3.1 kg and 24,800 Bq (47.8  
232 Bq  $\text{m}^{-2}$ ) in the decontaminated paddy. After irrigation, 5.5 kg of particles and 51,900 Bq (102 Bq  $\text{m}^{-2}$ ) of  
233  $^{137}\text{Cs}$  were discharged from the control plot, whereas 70 kg of suspended sediment and 165,000 Bq  
234 (317 Bq  $\text{m}^{-2}$ ) of  $^{137}\text{Cs}$  were discharged from the remediated field. This 3-fold higher export of  $^{137}\text{Cs}$  from  
235 the decontaminated paddy was likely explained by the supply of contamination from upper paddy  
236 fields, which remained connected to the remediated field. This result highlights the importance of  
237 remediation strategies focusing on the entire catchment scale.

238

239 Although this practice has not been specifically investigated in the literature, decontamination of  
240 farmland in the Fukushima fallout-impacted region was not limited to the removal of the 5-cm topsoil  
241 layer concentrating the radiocesium. After this first step, a layer of crushed granite, extracted from  
242 local quarries dedicated to decontamination, is used to replace the removed soil layer (Evrard et al.,  
243 2019). The entire soil profile consisting of the residual initial soil (in depth) and this crushed granite  
244 layer (on top) is then thoroughly mixed to prepare the soil for recultivation with the objective being to  
245 further dilute the residual radiocesium activities in the soil (Figure 3).

246

247 Other studies investigated the impact of remediation works on the radiocesium levels measured in  
248 sediment transiting the river networks draining the main radioactive pollution plume. Evrard et al.  
249 (2016) modelled the progressive dilution of radiocesium concentrations measured in sediment  
250 following decontamination works. They demonstrated a 90% decrease of the contribution of upstream  
251 contaminated soils to sediment transiting the coastal plains of the Mano and Niida Rivers between  
252 2012 and 2015. Furthermore, Osawa et al. (2018) monitored the radiocesium concentrations in  
253 suspended sediment collected in two tributaries of the Mano and Niida Rivers from 2013 and 2016.  
254 They also attributed a decrease in the  $^{137}\text{Cs}$  concentrations observed in 2016 to the decontamination  
255 efforts completed in 2015 in the local catchments.

256

#### 257 *River channel decontamination*

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

265 Nishikiori and Suzuki (2017) investigated this challenge in the 13-km<sup>2</sup> Kami-Oguni River catchment, a  
266 tributary of the Abukuma River, in the Fukushima Prefecture. Decontamination of a 170-m long and 8  
267 to 13-m wide river section isolated from the floodplain with 2-m high concrete dikes was studied, as  
268 the roads located on top of the banks were used by children to go to school. First, all the plants were



269 removed from the channel. Then, the top 5-cm layer of sediment was excavated from the dike slopes  
270 and planted with turf. Afterwards, sediment was removed from the channel, and the removal depth  
271 (between 15–35 cm) was locally adjusted depending on the vertical distribution of radiocesium  
272 measured using a NaI scintillation detector before, immediately after, and then three months after the  
273 remediation campaign. In addition, sediment samples were collected along transects at various depths  
274 in the floodplain and analysed with coaxial germanium detectors. Radiocesium contamination strongly  
275 varied with depth, depending on changes in the mud versus sand fractions, the former being enriched  
276 in radiocesium. Radiocesium concentration also varied across the channel, depending on the local flow  
277 velocity, which varied depending on the flood magnitude, the plant density and the microtopography.  
278 Before decontamination, air dose rates 1 cm above the ground varied between 0.2–1.99  $\mu\text{Sv h}^{-1}$ ,  
279 demonstrating the heterogeneity of contamination. After remediation, the contamination levels  
280 decreased by a factor of approximately 2, from a mean of 0.78 ( $\pm 0.41$ )  $\mu\text{Sv h}^{-1}$  before decontamination  
281 to 0.34 ( $\pm 0.15$ )  $\mu\text{Sv h}^{-1}$  after decontamination at 1 cm above the ground. However, Nishikiori and Suzuki  
282 (2017) underlined the risk associated with the potential deposition of contaminated material  
283 originating from upstream landscapes during subsequent flood events.

#### 284 *Forest decontamination*

285 The guidelines for the decontamination of forested areas in the Fukushima Prefecture (Table 2)  
286 indicate that only those areas lying within 20 m of houses should be targeted for remediation (Yasutaka  
287 and Naito, 2016) (Figure 4). Although the remediation in forests has not been a priority for the  
288 Japanese authorities during the early post-accidental phase, pilot studies were conducted to quantify  
289 the potential effectiveness of wider remediation programmes. Ayabe et al. (2017) investigated the  
290 impact of local-scale decontamination including the removal of the litter layer, the superficial soil layer,  
291 and the understorey in a secondary mixed forest with a cover of bamboo grass, *Sasa nipponica*, as  
292 understorey located in Kawamata Town. Although the total  $^{137}\text{Cs}$  contamination in soil and litter was  
293 reduced by  $\sim 20\%$  after decontamination compared to an adjacent untreated area, the radioactive  
294 contamination levels returned to their initial level four months after the completion of remediation  
295 works. This was likely due to the occurrence of a torrential rainfall event and the supply of  
296 contaminated foliage to the ground by litterfall. These results suggest that the removal of the litter  
297 and superficial soil layers in a contaminated forest may have limited effectiveness.

298 In another study by Lopez-Vicente et al. (2018), several different forest decontamination practices  
299 were compared through the monitoring of  $^{137}\text{Cs}$  contamination in soil and leaf samples in 10 plots  
300 installed in the evacuation zone, 16 km to the southwest of FDNPP, between May 2013 and July 2015  
301 (i.e. 27 months monitoring). Four potential forest remediation strategies were assessed. First, the  
302 combination of tree thinning and litter removal provided the best results to reduce  $^{137}\text{Cs}$  export (350–  
303 380  $\text{Bq m}^{-2} \text{day}^{-1}$ ), followed by the application of tree thinning only (163–174  $\text{Bq m}^{-2} \text{day}^{-1}$ ). Clearcutting  
304 and litter removal provided limited results (92–104  $\text{Bq m}^{-2} \text{day}^{-1}$ ), with higher  $^{137}\text{Cs}$  export rates than  
305 those observed from the control plots (52  $\text{Bq m}^{-2} \text{day}^{-1}$ ). Finally, plots where tree matting was  
306 conducted had lower  $^{137}\text{Cs}$  export rates (19–25  $\text{Bq m}^{-2} \text{day}^{-1}$ ) than control plots. Overall, the decreasing  
307 trend in radiocesium concentrations measured in the plot outflow was high in 2013, moderate in 2014  
308 and low in 2015 owing to the vegetation recovery after the countermeasures.

309

#### 310 **5. Impact of decontamination on dosimetry**

311 Two parameters are assessed before authorizing evacuees to return home: the prevalent dose rate  
312 and the cumulative dose. Importantly, background radiation levels need to be incorporated into this  
313 assessment. In the Fukushima Prefecture, background dose rates before the FDNPP accident were





314 estimated to be  $\sim 0.04\text{--}0.05$  mSv h<sup>-1</sup> (National Institute of Advanced Industrial Science and Technology,  
315 2011).

316 Individual external radiation doses (mSv day<sup>-1</sup>) may not be directly related to outdoor air doses (mSv  
317 day<sup>-1</sup>) as people do not spend 24 hours a day outside. When people are inside, the distances from the  
318 sources of radiation are greater and walls generate a shielding effect (IAEA, 2000). In Japan, when the  
319 Ministry of the Environment estimated daily individual external effective dose rates, it was assumed  
320 that people spent 8 hours outdoors and 16 hours indoors, with the indoor air dose rate being 40% of  
321 the outdoor air dose rate (Japanese Ministry of the Environment, 2013). Based on these assumptions,  
322 the external radiation dose rate is 60% of the air dose rate. Several researches have estimated external  
323 conversion coefficients according based on data provided by the Ministry of the Environment  
324 (Yasutaka and Naito, 2016).

325 Figure 5 compares the annual and individual dose rates that the global population may be exposed to  
326 in order to help facilitate a comparison with levels in the FDNPP fallout-impacted region. In Japan, a  
327 long-term dosimetric target of 1 mSv yr<sup>-1</sup> was adopted by the Nuclear Emergency Response  
328 Headquarters. Accordingly, a guidance value of 0.23  $\mu$ Sv h<sup>-1</sup> was proposed to achieve the target by  
329 implementing decontamination measures. In particular, areas with ambient dose rates exceeding this  
330 value were defined as ICAs. This guidance value is based on a simplified deterministic model assuming  
331 that inhabitants again spend 8 hours outdoors and 16 hours indoors (i.e. a shielding factor of 0.4) per  
332 day and that the contribution of natural radiation is 0.04 mSv h<sup>-1</sup> (IAEA, 2013). According to Mori et al.  
333 (2017), this model has three main challenges. First, the same behavioural pattern is assumed for the  
334 entire population. Second, the radiation exposure is assumed to be uniform. Third, conservative  
335 assumptions are adopted when converting the ambient dose into an effective dose. For instance, the  
336 time spent outside is assumed to be 8 hours, which is more than anticipated for the majority of the  
337 population and likely results in an overestimation of the actual measured doses (Nomura et al., 2015).

338 Although this approach is effective for the immediate post-accidental context, more sophisticated  
339 approaches are required to estimate doses over the longer term. Therefore, a probabilistic method  
340 that accounts for spatial variations (i.e., houses, workplaces, and other environments) of the  
341 contamination and for inter-population variations (i.e., indoor workers, outdoor workers,  
342 pensioners) in behavioural patterns was developed by Mori et al. (2017). For this approach, the 95<sup>th</sup>  
343 percentile doses for outdoor workers were above 1 mSv yr<sup>-1</sup> in 25 of the 59 municipalities in Fukushima  
344 Prefecture (1–35 mSv yr<sup>-1</sup>). In particular, the doses to more than 90% of the outdoor workers in Okuma  
345 town, Futaba town, Tomioka town, Namie town, and litate village were over 1 mSv yr<sup>-1</sup>. Furthermore,  
346 the 95<sup>th</sup> percentile doses for indoor workers were above 1 mSv yr<sup>-1</sup> in Okuma town, Futaba town,  
347 Tomioka town, Namie town, and litate village. If people return home in these municipalities, it is  
348 possible that they would be exposed to doses exceeding 1 mSv yr<sup>-1</sup> for all population groups. However,  
349 the results indicate that the same behavioural patterns and contamination levels should not be  
350 assumed for all inhabitants nor all municipalities. Based on the different behaviour of the local  
351 population, the 95<sup>th</sup> percentile doses of indoor workers and pensioners in 53 of the 59 municipalities  
352 were below the dosimetric target of 1 mSv yr<sup>-1</sup> (0.026–0.73 mSv yr<sup>-1</sup>) (Mori et al., 2017). Radiation dose  
353 rates were also measured among different types of workers having professional activities in the village  
354 of Kawauchi where the annual doses of foresters (range: 0.7–1.9 mSv yr<sup>-1</sup>) were not significantly higher  
355 than those of farmers (0.7–1.5), builders (0.6–1.5), office workers (0.5–1.5) and unemployed  
356 individuals (0.5–1.7). In contrast, decontamination workers (0.5–7.1) were found to have significantly  
357 higher dose rates (Orita et al., 2017).

358 The workers involved in decontamination activities were often directly exposed to internal irradiation  
359 through inhalation, which is much more difficult to measure than the external irradiation. Accordingly,



360 eighty-three people who worked in highly contaminated areas where surface  $^{137}\text{Cs}$  deposition density  
361 was over  $100 \text{ kBq m}^{-2}$  were enrolled in a study (Tsubokura et al., 2013). Using a database on internal  
362 exposure from the Hirata Central Hospital in Fukushima Prefecture, data were compiled on age,  
363 gender, body weight, equipment used in decontamination activity, total working period, duration  
364 between the final working day and the day of an examination, and  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  body burden. Hirata  
365 Central Hospital was also equipped with a permanent whole body counter with detection limits of 300  
366 Bq per individual for both  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  measurements following a 2-min scan. The levels of internal  
367 radiocesium exposure among all the decontamination workers were below the detection limits. No  
368 other radionuclides besides natural  $^{40}\text{K}$  were detected. No acute health problems had been reported.  
369 However, levels of external exposure were not assessed, as individual data on dose rates were not  
370 available. This study suggests that the resuspension of radioactive materials may cause a minimal  
371 internal contamination during decontamination works (Yamaguchi et al., 2012). Other studies  
372 calculated that radiation doses from internal exposure were marginal (Hayano et al., 2013; Tsubokura  
373 et al., 2015). As such, remediation efforts should be concentrated on reducing the external exposure  
374 of the local population.

375 According to the decontamination scenarios described in section 3, the reduction in annual individual  
376 additional Effective Dose (ED) for all decontamination scenarios was 1666 person-Sv for the SDZ and  
377 876-1245 person-Sv for the ICAs (Yasutaka and Naito, 2016). Despite the higher reduction rate  
378 achieved in the SDZ compared to the ICAs, they remained in the same order of magnitude although  
379 the decontamination efficiencies were very different in both areas. This result may be directly  
380 attributed to the differences in population density in SDZ and ICAs, with 90,000 inhabitants living in  
381 SDZs in 2010 versus approximately 1.5 million inhabitants living in ICAs areas exposed to over  $1 \text{ mSv}$   
382  $\text{yr}^{-1}$ . This strong dependence of ED on population densities may lead the authorities to concentrate  
383 their remediation efforts in the most densely populated areas. The results obtained also depend on  
384 the effectiveness of these decontamination programmes. For instance in ICAs, where approximately 1  
385 million inhabitants reside in areas exposed to  $1\text{--}5 \text{ mSv yr}^{-1}$ , the reduction in annual individual additional  
386 ED was much larger in those areas where the full decontamination scenario would be implemented.

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

396

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

### 398 *Waste management*

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



405 Regarding waste contaminated with  $^{137}\text{Cs}$ , the final objective is to bring radiocesium to the solution  
406 phase and then enrich it, to reduce it to the smallest possible volume. In the Fukushima Prefecture,  
407 the radiocesium concentrations found in the disaster waste are lower than other alkali metals.  
408 Therefore, the treatment methods require approaches that help concentrate  $^{137}\text{Cs}$  (Parajuli et al.,  
409 2016a). The reduction of solid waste volume can be achieved through compaction or incineration. For  
410 organic waste (i.e. forest litter, weeds, wood, or tree branches from contaminated areas), incineration  
411 ('thermal treatment') is traditionally preferred (IAEA, 2003, 2006) as it reduces the volume of waste by  
412 several orders of magnitude (Parajuli et al., 2013). The problem is that this 'thermal treatment' may  
413 enrich contaminants and the Japanese legislation has a  $8000 \text{ Bq kg}^{-1}$  radionuclide threshold for placing  
414 waste in landfills (Japanese Ministry of the Environment, 2011b).

415 Accordingly, waste contaminated with radionuclide levels between  $8000 \text{ Bq kg}^{-1}$  and  $100,000 \text{ Bq kg}^{-1}$   
416 needs to be disposed in designated landfills equipped with radiation level and leachate monitoring as  
417 well as a treatment system in order to control the potential release of radioisotopes into the  
418 environment (Parajuli et al., 2013). Therefore, either specially designed landfills need to be constructed  
419 or pre-treatment methods need to be designed to remove radionuclides from the waste. This issue is  
420 crucial as the construction of temporary storage sites and interim storage facilities were estimated to  
421 account for 50% of the overall cost of decontamination. For example, transport, storage and  
422 administrative costs were estimated to represent a cost of 1.55–2.12 trillion JPY (~12.4–17 billion EUR)  
423 for the decontamination scenarios complying with the guidelines of Japanese authorities (Yasutaka  
424 and Naito, 2016). Furthermore, securing routes and locations for transporting more than 20 million  
425 tonnes of decontamination waste and removed soil that were generated to the interim storage  
426 facilities remains a major challenge. Nevertheless, assessing the management and storage of low-  
427 concentration radioactive cesium-containing soil and methods for using controlled landfill sites may  
428 lead to a significant reduction in the amount of material requiring transport.

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

439 Contaminated soil removed by decontamination works is transported to an interim storage facility  
440 where flammable decontamination waste (DW) is incinerated or melted to reduce its mass and  
441 volume. Depending on its radiocesium content, this waste is either stored at an interim storage facility  
442 or disposed in a leachate-controlled type of landfill site (Fujiwara et al., 2017). The total surface area  
443 of the interim storage facilities in Futaba and Okuma municipalities is planned to cover ~1600 ha, and  
444 by February 2019, a contract was already established between Japanese authorities and landowners  
445 for ~70% of the land required for storage. Soil storage operations started in October 2017 in Okuma  
446 and in December 2017 in Futaba. By March 2019, ~2.5 million  $\text{m}^3$  of waste soil had already been  
447 transported from the temporary storage facilities distributed across all the remediated area to these  
448 two interim storage facilities (Japanese Ministry of the Environment, 2019a). All the soil waste is  
449 planned to be transported to the Okuma and Futaba sites by the end of 2021 (Japanese Ministry of the  
450 Environment, 2019b). The final disposal of this decontamination waste should take place outside of



451 the Fukushima Prefecture, within 30 years after the opening of the interim storage facilities (i.e.,  
452 ~2047).

#### 453 *Incineration*

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

#### 466 *Incineration ash treatment*

467 The chemical form and the leachability of radiocesium depends on the type of waste incinerated.  
468 Results observed for three different types of ash samples suggest that <sup>137</sup>Cs along with other alkali  
469 metals in wood bark and household garbage ashes, originated from burnable materials, were mostly  
470 washed out with water even at ambient temperatures. However, municipal sewer sludge was  
471 different, with potential <sup>137</sup>Cs elution only occurring under very specific conditions (i.e., with acid  
472 treatment and under high temperatures). Acid treatment at high temperatures was found to be  
473 inappropriate for treating wood bark and household garbage ashes because of the generation of a Ca  
474 excess leading to gypsum formation and complexifying the subsequent treatment process (Parajuli et  
475 al., 2013).

#### 476 *Soil recycling*

477 As 22 million m<sup>3</sup> of decontamination soil (i.e. 90% of the total) and incineration ash waste (10%) is  
478 expected to be produced through remediation of the fallout impacted region, recycling may be  
479 instrumental for reducing this volume (Takai et al., 2018). The Japanese Ministry of Environment  
480 developed a policy to separate decontamination soil into low- and high-activity soils, the former being  
481 'recycled' in public projects. In these uses, decontamination soil will be used for the basic structure  
482 and will be covered by uncontaminated soil or concrete. In theory, the unconditional 'clearance level'  
483 defined by IAEA for the use of recycled material is fixed to 100 Bq kg<sup>-1</sup> for radiocesium. However, as  
484 disaster waste was found with higher <sup>137</sup>Cs levels, the Japanese Ministry of Environment decided that  
485 those materials with radiocesium levels up to 3000 Bq kg<sup>-1</sup> can be reused at a minimum depth of 30  
486 cm underground (reference level assessed for recycling of concrete for the road subbase course). For  
487 decontamination soil recycling, the radioactivity level had to be reanalysed for a different type of  
488 engineering structures (deterministic estimation of radiation dose rates). The corresponding level of  
489 radiocesium concentrations in the soil was estimated to 6000 Bq kg<sup>-1</sup>. To confine doses to levels below  
490 10 μSv yr<sup>-1</sup> based on the derived radioactivity level, an additional layer of soil slope protection of 40  
491 cm or more was needed. Accordingly, the Japanese Ministry of Environment determined the maximum  
492 radioactivity level to be 6,000 Bq kg<sup>-1</sup> for embankments covered with 50 cm uncontaminated soil.  
493 Overall, the recycling of decontaminated soil is limited to civil engineering structures in public projects,  
494 such as road embankments and coastal levees. Takai et al. (2018) evaluated the associated additional



495 doses to workers and the public using these structures and demonstrated that additional dose rates  
496 would remain below the 1 mSv yr<sup>-1</sup> threshold corresponding to 6000 Bq kg<sup>-1</sup>.

497 In Japan, contaminated wastes are disposed under the standard of 8,000 Bq kg<sup>-1</sup>. The volume of  
498 decontamination soil having a radioactivity concentration of 8,000 Bq kg<sup>-1</sup> or below is estimated to be  
499 approximately 10 million m<sup>3</sup>, which corresponds to half of the total amount of decontamination soil  
500 generated. The radioactivity concentration of 8,000 Bq kg<sup>-1</sup> will decrease to 6,000 Bq kg<sup>-1</sup> in 5 years.  
501 Therefore, more than half of the total decontamination soil should become recyclable in at least 5  
502 years. Through the use of pre-treatment activities, such as classification processing, even more  
503 decontaminated soil may become recyclable in the non-too distant future (Takai et al., 2018).

#### 504 *Soil remediation*

505 Remediation of contaminated soil based on a hot acid treatment was tested for the two most common  
506 soil groups found in Fukushima (Parajuli et al., 2016b): Cambisols (i.e. brown forest soils) and Andisols  
507 (i.e. soils developed on volcanic ash). Although this method was shown to be effective for the former  
508 soil type, this was not the case for the latter. In particular, lime must be added to treat Andisols which  
509 must be mixed with untreated and uncontaminated soil prior to being reused for cultivation.  
510 Furthermore, to avoid the transfer of residual radiocesium to plants, additives such as zeolite or  
511 Prussian blue adsorbents need to be incorporated into the Andisols. The problem associated with this  
512 strategy is that, through their ageing, zeolites may increase <sup>137</sup>Cs exchangeability with potassium and  
513 accelerate <sup>137</sup>Cs transfer to the cultivated plants over longer time periods (Yamaguchi et al., 2019).  
514 These restrictions illustrate the difficulty of finding alternatives to the storage of decontamination soil  
515 waste in interim facilities.

516

#### 517 **7. Perspectives for future research**

518 The total estimated decontamination cost would exceed 16 trillion JPY (~128 billion EUR) if all forested  
519 areas exposed to radiation dose rates exceeding 1 mSv yr<sup>-1</sup> were decontaminated. However,  
520 decontaminating all of the forested areas would not result in a major ED reduction for the average  
521 inhabitant (Yasutaka and Naito, 2016). As almost 70% of the surface area of Fukushima Prefecture is  
522 covered with forests (Hashimoto et al., 2012) and forestry is a significant economic activity in the  
523 region, future research should prioritize investigating radiocesium dynamics in these regions. In  
524 particular, the biological cycling of <sup>137</sup>Cs in forests has now been affected by the decomposition of litter  
525 where radiocesium was concentrated shortly after the FDNPP accident (Koarashi et al., 2012).  
526 Furthermore, the local population in rural areas of the Fukushima Prefecture enjoy 'satoyama', or the  
527 collection of vegetation, including mushrooms, edible wild plants, and firewood from forested  
528 landscapes (Prand-Stritzko and Steinhauser, 2018; Nihei, 2016). In addition, approximately 1800  
529 workers are employed by the forest industry in the region (Yasutaka and Naito, 2016). For many of the  
530 local inhabitants, the forest, the satoyama, and its harvest are inseparable from their daily lives.

531 Forest sources were also shown to deliver a significant proportion of contaminated material to the  
532 river systems draining the fallout-impact region. The analysis of deposited particulate matter collected  
533 in three fallout-contaminated coastal catchments between November 2012 and November 2014  
534 demonstrated that forest sources supplied a mean of 17 % (standard deviation, SD, 10 %) of the  
535 sediment transiting these river systems (Lacey et al., 2016). Huon et al. (2018) obtained similar results  
536 through the analysis of sediment cores collected between November 2014 and April 2015 in a dam  
537 reservoir draining fallout-impacted cultivated and forested landscapes, with the latter supplying a  
538 mean of 27% (SD, 6%) of the material deposited in the lake. These conclusions were validated through  
539 an analysis of a larger number of sediment samples (n=400) collected in coastal river systems in the



540 Fukushima region over a longer time period (from November 2011 to November 2017) where a mean  
541 of 24% (SD, 21%) of the material transiting these systems was modelled to be derived from forested  
542 landscapes (Evrard et al., 2019). Cumulatively, these results demonstrate that forested landscapes  
543 represent a potential long-term source of particulate contaminated matter that likely will require  
544 diligent management for the foreseeable future.

545 In cultivated landscapes where the remediation activities were concentrated, the main question is  
546 whether or not to restart agricultural production. The removal of the topsoil layer concentrating the  
547 radiocesium, the replacement of this material with crushed granite extracted from local quarries and  
548 the final mixing of the entire profile to prepare the soils for re-cultivation raises several important  
549 questions. For example, to what extent will the residual radiocesium in the soil be transferrable to the  
550 plants cultivated on these soils? How will the crushed granite, which was homogenized into the soil,  
551 affect the soils fertility? Recent research showed that potassium fertilization is required to maintain  
552 productivity when restarting cultivation after decontamination (Kurokawa et al., 2019). Indeed, as it  
553 was demonstrated in the current literature review, the reopening of the region after the completion  
554 of remediation activities represents a unique situation in history, coupled with unprecedented  
555 challenges that require further ongoing investigations.

556 Although previous dosimetric studies demonstrated that currently the internal exposure of both the  
557 local population and the decontamination workers remains minimal, both internal and external  
558 exposures of these groups should be studied over longer temporal periods to help understand long-  
559 term impacts of this and potentially other nuclear accidents on exposed population groups. More  
560 research is also required to understand the fate and dynamics of other longer-lived radionuclides in  
561 the Fukushima region including radiocarbon (Paterne et al., 2018; Povinec et al., 2016; Xu et al., 2016),  
562 plutonium and uranium isotopes (Jaegler et al., 2018; Zheng et al., 2013; Steinhäuser, 2014) as they may  
563 be persistent in the environment even though many were emitted only at the trace and ultra-trace  
564 levels.

## 565 **Conclusions**

566 The quick and early decision of the Japanese authorities to decontaminate FDNPP fallout impacted  
567 landscapes was unprecedented. Decontamination activities were rapidly implemented in agricultural  
568 and residential areas covering a surface of ~9000 km<sup>2</sup>. These remediation activities produced ~20  
569 million m<sup>3</sup> of soil waste in less than 6 years (2013–2019) with an approximate cost of 3 trillion JPY (~24  
570 billion EUR). The strategy of removing the surface layer of the soil concentrating <sup>137</sup>Cs was shown to be  
571 effective in cultivated land when the strategy was applied at a catchment scale to avoid the supply of  
572 mobilized contamination from the headwaters. The main current challenges are associated with the  
573 treatment and the transport of this waste to the interim storage facilities for the next ~30 years that  
574 are being built near the FDNPP. The re-cultivation of the soils after decontamination also raises several  
575 concerns. In particular, more information is required regarding soil fertility after decontamination and  
576 the potential transfer of the residual <sup>137</sup>Cs to the plants cultivated on decontaminated fields.

577 The risks of internal and external radiation dose exposures of the decontamination workers and the  
578 local population to exceed the target of 1mSv yr<sup>-1</sup> appeared to be low during the early post-accidental  
579 phase. However, dosimetric monitoring programmes should be carried out to confirm this result over  
580 the longer term, particularly after local population returns to the region. Furthermore, as ~75% of the  
581 surface exposed to the highest <sup>137</sup>Cs fallout levels in the Fukushima Prefecture are covered with forests  
582 where decontamination was not implemented, the perennial contribution of radiocesium to the river  
583 systems draining these mountainous, forested landscapes exposed to typhoons should be  
584 investigated. The behaviour and the dynamics of longer-lived radionuclides such as plutonium isotopes



585 should also be studied in the future as they may persist in the environment for long timescales even  
586 though they were emitted at trace and ultra-trace levels.

587

#### 588 **Acknowledgements**

589 This research was funded by the AMORAD project (ANR-11-RSNR-0002), supported by the French  
590 National Research Agency (ANR, Agence Nationale de la Recherche). The support of CNRS (Centre  
591 National de la Recherche Scientifique, France) and JSPS (Japan Society for the Promotion of Science)  
592 in the framework of the Franco-Japanese collaboration project framework (PRC, CNRS-JSPS) is also  
593 gratefully acknowledged.

594

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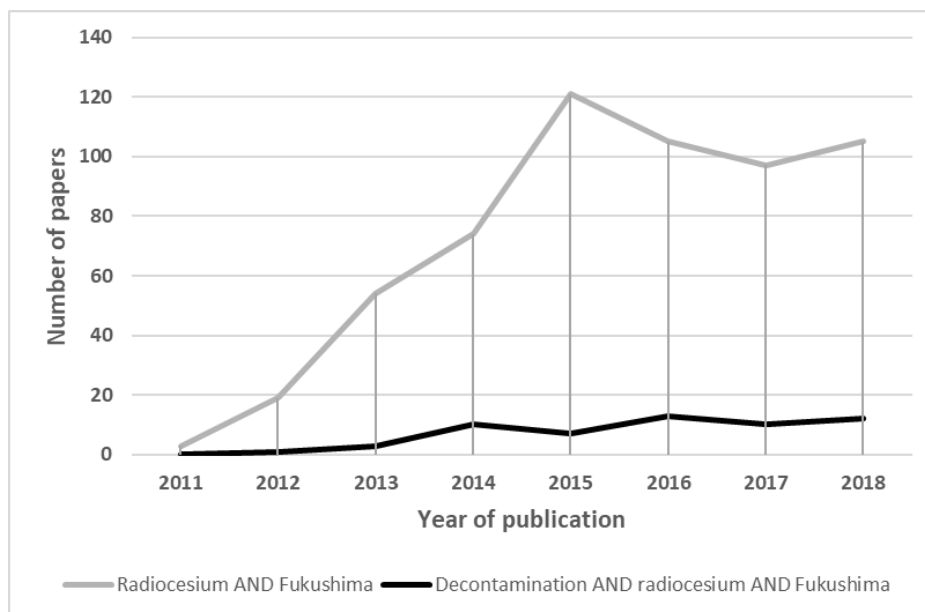
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880 **Figures**

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

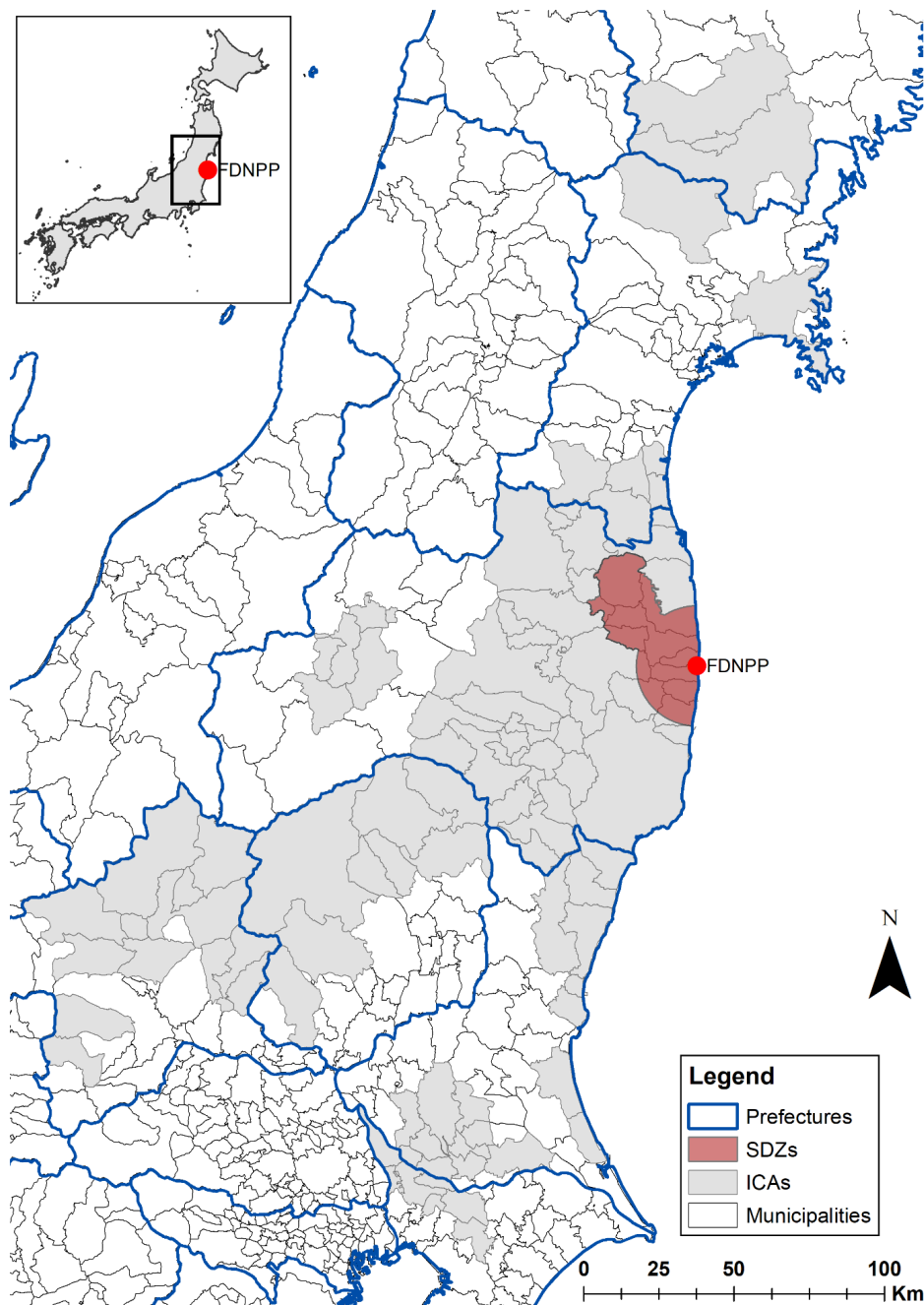


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899 Figure 2. Location of the Fukushima Prefecture in Japan (Inset Map) and the location of the Special  
900 Decontamination Zones (SDZs) and the Intensive Contamination Survey Areas (ICAs).

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



908 (a)



909 (b)



(c)

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

914 Example from litate Village (Sasu district).



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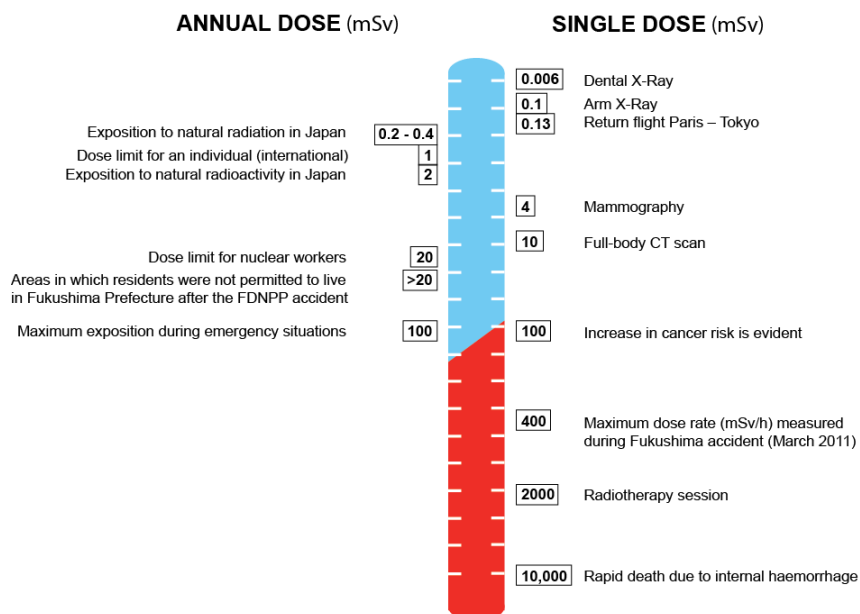
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918 Figure 5. Comparison of annual and individual radioactive dose rates to which the population may be  
 919 exposed, based on a compilation of data (Commissariat à l'Energie Atomique et aux Energies  
 920 Alternatives, 2016; Harada et al., 2014).



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



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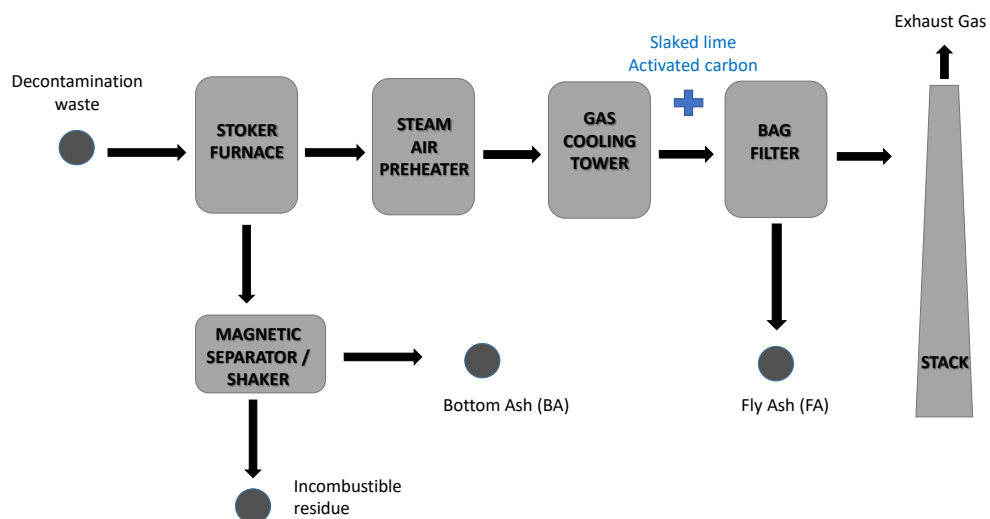
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944 Figure 7. Simplified diagram showing the functioning of an incineration plant treating decontamination  
945 waste in Fukushima, modified after Parajuli et al. (2013) and Fujiwara et al. (2017).



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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	<a href="https://www.pref.fukushima.lg.jp/site/portal-english/list382.html">https://www.pref.fukushima.lg.jp/site/portal-english/list382.html</a>
Ministry of Environment	Environmental Remediation	<a href="http://iosen.env.go.jp/en/">http://iosen.env.go.jp/en/</a>
Ministry of Economy, Trade and Industry	Fukushima Today webpage	<a href="http://www.meti.go.jp/english/earthquake/index.html">http://www.meti.go.jp/english/earthquake/index.html</a>
Nuclear Regulation Authority	Monitoring information	<a href="https://radioactivity.nsr.go.jp/en/">https://radioactivity.nsr.go.jp/en/</a>



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

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



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

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