Reviewer's comment	Authors' reply
Anonymous reviewer # 1	
SOIL-2019-43 Strategies and effectiveness of land decontamination in the region affected by radioactive fallout from the Fukushima nuclear accident: A review This review describes the effects of the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in March 2011. This review describes the spatial extent of the decontaminated zone and the remediation strategies in different environments (including schedule and costs). The issues of the impacts of remediation activities on dosimetry and large volume of waste generated are also discussed. They conclude that the strategy of removing the surface layer of the soil concentrating 137Cs was effective in cultivated land at a catchment scale avoiding its transformation into source of contamination. Discussion and conclusion they are extremely interesting and provocative. This manuscript adheres to the journal's standards. The research meets the applicable standards for the research integrity. The research output, in terms of novelty, scores good uniqueness. The level of clarity is well above the threshold of acceptability. Potentially, its potential impact upon the international scientific community of reference is good. The article is presented in an intelligible manner. This work is interesting and deserves to be published.	Many thanks for these encouraging general comments!
Title: 20 words. It can be shortened	As recommended, we have shortened the title of the manuscript (P.1 L.3).
Abstract: OK.	OK
Keywords: REVISE. The keywords, together with title and abstract function in a system comparable to a chain reaction. Once the keywords have assisted the Reader find the suitable paper and its title has fruitfully drawn in the attention, it is up to the abstract to further activate the interest and keep their curiosity. So, these three elements must work together and not replicate each other.	Thank you for this comment - the 'nuclear accident' keyword has been removed as it duplicated the information contained in the title. It was replaced with 'phytoextraction' (P. 1 L. 43).
Introduction: OK	OK
Conclusion: OK. If confirmed.	OK
Figures: REVISE. Figure 1 unnecessary (in case, move to Supplementary Material), I would suggest to shift Figure 6 here.	We somewhat disagree with the reviewer, and we think that providing Fig. 1 is informative for the readership. Furthermore, the other

Figure 2 is quite useless (please, add geographical coordinates). This figure is rather unnecessary, a kmz file would be more useful.	reviewers/commenters did not request to remove Fig. 1. We also do think that Fig. 2 is important. Accordingly, we have updated figure 2 to now include geographic coordinates. In addition, we now also provide a kmz file as Supplementary Material. Many thanks for this suggestion.
References: REVISE.	
In particular (page.row): 2.63 This section, including Figure 1, it is purely of a methodological nature, being a review, and, in my opinion, it takes away the bite from the paper.	We agree with the reviewer that this is not the main message of the paper, although we do think that this section sets the scene and puts this research in context. Furthermore, this approach of grounded a review in the literature is quite common in review papers. Of note, the other reviewers did not request to change or remove this paragraph.
2.72 Also in my opinion, a review is the right interface where 'grey literature' becomes scientific literature, sensu stricto. Precisely, it must be the responsibility of the authors to insert unpublished and reliable information. The fact that a job has been peer-reviewed, per se, is not a guarantee of total reliability. As, on the other hand, official documents (published in languages other than English, as mostly in Japanese in this case) are certainly not unreliable. My advice is to insert that grey literature useful to enrich the review of data and interpretations. Converting Table 1 into individual references.	Of note, we integrated in the text the most up-to-date information from these grey sources that is of interest to the readership of our manuscript (more than 10 references to grey literature documents were made in this manuscript). Most information available in the grey literature sources referred to in Table 1 consist of guidelines provided by the Japanese authorities.
3.96 I would begin by telling, briefly, what happened. Mentioning, for instance, the International Nuclear and Radiological Event Scale (INES) introduced by the International Atomic Energy Agency. And, discussing, comparatively (e.g. Ivanov et al. 1997, Rosén et al. 1999. And the, already quoted, doi>10.1016/j.scitotenv.2013.10.029), the two Level 7 INES Major accidents.	We added a short description of the events that occurred at FDNPP in 2011 (P.3, LL.109-116). We agree that the INES level associated with the accident is very important to mention. This was added to the very beginning of the manuscript (P.1 LL.48-49). As the objective of the current manuscript is not to compare the Chernobyl and the Fukushima accidents, we added this information in the abstract with relevant references for those readers who would be interested in this topic (PP.2-3; LL.89-95).
3.113 I would mention that the sievert is a derived unit of ionizing radiation dose in the International System of Units and is a measure of the human health effect of ionizing radiation	Done, on P.3 LL. 131-132.

4.144 what interchanged means, exactly?	Replaced (modified on P. 4 L. 163).
4.163 A term of comparison would help the Reader. For instance, the whole EU budget was at some C37 billion in 2017	A comparison with the EU budget in 2017 was added (P.5, LL.186-188).
5.192 Please, discuss this issue comparatively (e.g. doi> 10.1021/es980788+)	Again, the main goal was to review the current strategies and effectiveness of decontamination efforts after the Fukushima accident. Adding in a comprehensive comparison to Chernobyl is beyond the scope of this review. To address these and other comments, we did note how the fallout situations are different in these two regions which would complicate a direct comparison of the remediation and decontamination efforts (PP.2-3; LL.89-95).
6.258 Please, discuss this issue comparatively (e.g. Santschl et al. 1990)	Please see our other comments regarding the comparison with the Chernobyl situation.
12.505 This section deserve more room and comparative discussion (e.g. Rosén et al. 1999)	Please see our other comments regarding the comparison with the Chernobyl situation.
References Ivanov YA et al. 1997. Migration of 137Cs and 90Sr from Chernobyl fallout in Ukrainian, Belarussian and Russian soils. J. Environ. Radioactivity 35, I-21 Rosén K et al. 1999. Migration of radiocaesium in Swedish soil profiles after the Chernobyl accident, 1987-1995. J. Environ. Radioactivity 46, 45-66 Santschl PH et al. 1990. The self-cleaning capacity of surface waters after radioactive fallout evidence from European waters after Chernobyl, 1986-1988. Environ. Sci. Techn. 24, 519-527	Again, although we acknowledge that these articles are of a wide interest, the objective of this review was to focus on the lessons learnt from the Fukushima case study, which is very different on many aspects from the Chernobyl experience. Although we agree that a comparison of the strategies of remediation and their effectiveness in Fukushima vs. in Chernobyl would have a wide interest, our opinion is that it falls beyond the scope of the current review. However, these references were added to the current manuscript (PP.2-3; LL.89-95).
The manuscript represent a substantial contribution to scientific progress within the scope of SOIL (interdisciplinary, mainly). The results are discussed in a thorough and balanced way (consideration of related and relevant work, including appropriate references need some reworking). Scientific results and conclusions are presented in a clear, concise, and quite well-structured way. Recommendation: Major revision is required	Many thanks for these encouraging general comments!
Yves Thiry's comments (annotated manuscript)	
Nice review at the good level for an educational communication and popularization on the issue and the challenging questions that remain. I included directly in the PDF file some suggestions of additional precisions or questions that would merit clarifications.__Please also note the supplement to this comment:https://www.soil-discuss.net/soil-2019-43/soil-2019-43-SC1-supplement.pdf	Many thanks for these positive comments and for sharing these suggestions in the pdf file. All the points were addressed in the text (except the comparison with the Chernobyl situation, please see our reply to Reviewer 1 on this topic).

Anonymous reviewer # 2	
The review paper entitle "Strategies and effectiveness of land decontamination in the region affected 1 by radioactive fallout 2 from the Fukushima nuclear accident: A review" It is a very well written review, very informative with plenty of details. A large number of publications are given. With this review, it is becoming obvious how the remediated actions taken from Japanese authorities had crucially resulted in low effective doses to the population Furthermore, the remediated actions taken in Japan will remain a great lesson in case of a similar accident. All aspects or remediation actions in various environments are covered as well as their effectiveness. Furthermore, perspective of future research is given too. The manuscript can be published as it is.	Many thanks for this positive review!

Anonymous reviewer # 3	
Manuscript "Strategies and effectiveness of land decontamination in the region affected by radioactive fallout from the Fukushima nuclear accident: A review", by Evrard et al is a very interesting report of the nuclear accident occurred in Japan eight years ago and, in particular, of the countermeasures adopted to safeguard the well-being of the populations involved. I liked reading it, also because it is well written and contains a lot of ancillary information to the main subject however useful to understand the enormity of the problem.	Many thanks for these positive comments.
I suggest only the following few additions, which I think can further improve the quality of work: - without absolutely making a comparison between the two events, at least in terms of soil contamination levels could some Chernobyl data be cited?	Added (P.2, LL. 55-58).
- Lines 507-509: please provide an explanation of why hot acid treatment does not work with Andisols, and why these soils need lime to be reused for cultivation.-	Lime is needed to readjust the pH to reasonable levels after acid treatment for allowing recultivation on these soils, Andisols were more sensitive to this treatment than Cambisols (P.12, LL.539-540).
Lines 526-528: can you say something about the restrictions to which people in the contaminated areas has had to adhere in terms of collection of mushrooms and other products of the understory or use of firewood?	Added (P.13; LL.559-563).

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

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

4

5 **Olivier Evrard[1], J. Patrick Laceby[2], Atsushi Nakao[3]**

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7 CNRS/UVSQ), Université Paris-Saclay, Gif-sur-Yvette, France.
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11
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13 **Abstract**

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

42

44

1. Introduction

Large quantities of radiocesium (12–62 PBq) were released into the environment by the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in March 2011 (Stohl et al., 2012;Chino et al., 2011). Accordingly, this accident hit the maximum level (i.e. 7) on the International Nuclear and Radiological Event Scale (INES)(IAEA, 2013a). Airborne and ground contamination surveys demonstrated that the contamination was the highest (i.e., initial ^{137}Cs levels >100,000 Bq m^{-2}) in a plume extending to the northwest of the FDNPP covering an area of ~3000 km² (Kinoshita et al., 2011;Chartin et al., 2013;Yasunari et al., 2011). Although many radioactive substances were released into the environment by the FDNPP accident, radiocesium (i.e., ^{134}Cs and ^{137}Cs) presents the most serious risk to the local population over the medium to long term as it was emitted in very large quantities and it has a relatively long half-life (i.e., ^{134}Cs - 2 years; ^{137}Cs - 30 years) (Steinhauser et al., 2014). The highest contamination radiocesium levels observed in Fukushima (>185 kBq m^{-2}) are similar to those recorded in the area impacted by Chernobyl fallout, although on a much smaller land surface area in Japan (1700 km²) compared to eastern Europe (29,400 km²)(Steinhauser et al., 2014).

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

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

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

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

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

107

2. Areas targeted by decontamination

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

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

126 According to the decontamination roadmap, the remediation programme had to be implemented in
127 'special areas' where targets were set for the exposure of the public to external dose rates in order for
128 residents to return to their day-to-day lives (Yasutaka and Naito, 2016). Achieving pre-accident
129 radiation levels is not the objective, rather the effectiveness of decontamination will ultimately depend
130 upon the land use and the air dose of each particular area. Two zones were delineated with different
131 strategies for remediation (Figure 2), based on different values of dose equivalent (i.e. the biological
132 effect of ionizing radiation) expressed in Sievert (Sv) and its sub-units. First, Special Decontamination
133 Zones (SDZ) are areas located within a 20-km radius of the FDNPP or areas where the cumulative dose
134 one year after the accident was expected to exceed 20 mSv yr^{-1}. SDZs occur in 11 municipalities (1117
135 km²) where residents were evacuated after the FDNPP accident in 2011. The central government of

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

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

149

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

151 The effectiveness of decontamination was assumed to strongly vary depending on the remediation
152 method and the initial radiation dose rates prior to decontamination. Different remediation
153 techniques were proposed depending on the land use and the zone (i.e., SDZ vs. ICA). Yasutaka et al.
154 (2013a) and Yasutaka et al. (2013b) compared the impact of four scenarios of decontamination,
155 including two very unlikely options (i.e., minimal and maximal scenarios), in terms of effectiveness and
156 cost according to the results of demonstration tests conducted by JAEA. These results were updated
157 in a more recent publication (Yasutaka and Naito, 2016). Only the two scenarios following the
158 guidelines provided by the Japanese government are assessed in this review. As such, the results of
159 both the minimal and maximal remediation options are therefore not discussed. In the first scenario,
160 5 cm of topsoil was removed from 50% of agricultural land in the SDZ where ^{137}Cs concentrations
161 exceed 5000 Bq kg^{-1}, and replaced with a 5-cm layer of 'clean' soil (measure A1; Table 2). In the
162 remaining 50% of agricultural land with ^{137}Cs concentrations below 5000 Bq kg^{-1}, topsoil was
163 ~~interchanged~~ replaced with subsoil (measure A3). Ploughing with zeolite and potassium (measure A4)
164 was adopted for agricultural land in ICAs where the annual additional effective dose exceeded 1 mSv
165 yr^{-1}. In the second scenario, measure A1 was applied to all cultivated land. Similar measures (e.g. Table
166 2) were included in both scenarios to decontaminate forested areas, roads and houses. The total
167 decontamination cost for implementing these remediation measures varied between 2–5.1 trillion JPY
168 (~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
169 EUR) for the ICAs. Although the area where decontamination has been implemented in the ICAs (922–
170 3330 km²) covers a surface 3 to 11 times larger than that of SDZ (295 km²), the decontamination costs
171 for the SDZ and ICAs are in the same order of magnitude.

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

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

182 According to the latest available figures from the Japanese Ministry of the Environment (2019b) at the
183 end of 2018, the volume of soil waste generated in the SDZ was 9,100,000 m^3 with a remediation cost
184 of approximatively 1.5 trillion JPY (~12 billion EUR). In the ICAs, the latest figures available for March
185 2018 showed that 7,900,000 m^3 of waste soil were produced with a remediation cost of approximately
186 1.4 trillion JPY, equivalent to ~11 billion EUR (Japanese Ministry of the Environment, 2019b). This
187 amount corresponds to ~8% of the total annual expenses of the European Union (~134 billion EUR in
188 2017) (European Union, 2018).

189

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

191 *Radiocesium distribution with depth in the soil*

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

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

210 Different strategies were carried out in Japan to decontaminate soil in farmland, either removing the
211 contaminated layer of soil or through the sowing of plants having the capacity of extracting and
212 concentrating radiocesium from the soil. There are few publications in international journals regarding
213 the potential or effectiveness of the latter strategy (e.g. Pareniuk et al., 2015). Among the few available
214 studies, Kobayashi et al. (2014) grew thirteen plant species from three families (*Asteraceae*, *Fabaceae*,
215 and *Poaceae*) in shallow and deeply cultivated fields where the 0–8 cm and 0–15 cm soil layers were
216 respectively ploughed. The variation in plough depth was expected to reflect the impact of different
217 contact zones between the root systems and radiocesium in the soil. Overall, 29 to 225 Bq kg^{-1} dry
218 weight of ^{137}Cs were found in the plants, corresponding to transfer factors ranging from 0.019 to 0.13
219 (geometric mean (GM), 0.057) for plants growing in shallow soils, and from 0.022 to 0.13 (GM 0.063)
220 for plants growing in deeper soils (Kobayashi et al., 2014). The authors found that none of their tested
221 plant species resulted in a significant decrease in radiocesium in soil likely because of the strong
222 fixation of ^{137}Cs to clay particles. This result was confirmed by Yamashita et al. (2014) who showed that
223 99 wild plants grown in paddy and upland fields had a very low phytoextraction efficiency. Tamaoki et

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

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

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

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

271 Other studies investigated the impact of remediation works on the radiocesium levels measured in
272 sediment transiting the river networks draining the main radioactive pollution plume. Evrard et al.
273 (2016) modelled the progressive dilution of radiocesium concentrations measured in sediment
274 following decontamination works. They demonstrated a 90% decrease of the contribution of upstream
275 contaminated soils to sediment transiting the coastal plains of the Mano and Niida Rivers between
276 2012 and 2015. Furthermore, Osawa et al. (2018) monitored the radiocesium concentrations in
277 suspended sediment collected in two tributaries of the Mano and Niida Rivers from 2013 and 2016.
278 They also attributed a decrease in the ^{137}Cs concentrations observed in 2016 to the decontamination
279 efforts completed in 2015 in the local catchments.
280
281 *River channel decontamination*

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

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

309 *Forest decontamination*

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

317 understorey located in Kawamata Town. Although the total ^{137}Cs contamination in soil and litter was
318 reduced by ~20% after decontamination compared to an adjacent untreated area, the radioactive
319 contamination levels returned to their initial level four months after the completion of remediation
320 works. This was likely due to the occurrence of a torrential rainfall event and the supply of
321 contaminated foliage to the ground by litterfall. These results suggest that the removal of the litter
322 and superficial soil layers in a contaminated forest may have limited effectiveness if these operations
323 are conducted too early after the initial radionuclide deposition. Decontamination should take place
324 after the peak of humus contamination occurring typically ~5 years after the initial fallout although
325 temporal variations were observed depending on the tree and humus types (Thiry et al., 2018).

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

338

5. Impact of decontamination on dosimetry

340 Two parameters are assessed before authorizing evacuees to return home: the prevalent dose rate
341 and the cumulative dose. Importantly, background radiation levels need to be incorporated into this
342 assessment. In the Fukushima Prefecture, background does rates before the FDNPP accident were
343 estimated to be ~0.04-0.05 mSv h^{-1} (National Institute of Advanced Industrial Science and Technology,
344 2011).

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

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

362 al. (2017), this model has three main challenges. First, the same behavioural pattern is assumed for
363 the entire population. Second, the radiation exposure is assumed to be uniform. Third, conservative
364 assumptions are adopted when converting the ambient dose into an effective dose. For instance, the
365 time spent outside is assumed to be 8 hours, which is more than anticipated for the majority of the
366 population and likely results in an overestimation of the actual measured doses (Nomura et al., 2015).

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

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

404 According to the decontamination scenarios described in section 3, the reduction in annual individual
405 additional Effective Dose (ED) for all decontamination scenarios was 1666 person-Sv for the SDZ and
406 876-1245 person-Sv for the ICAs (Yasutaka and Naito, 2016). Despite the higher reduction rate
407 achieved in the SDZ compared to the ICAs, they remained in the same order of magnitude although

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

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

425

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

427 *Waste management*

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

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

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

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

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

468 Contaminated soil removed by decontamination works is transported to an interim storage facility
469 where flammable decontamination waste (DW) is incinerated or melted to reduce its mass and
470 volume. Depending on its radiocesium content, this waste is either stored at an interim storage facility
471 or disposed in a leachate-controlled type of landfill site (Fujiwara et al., 2017). The total surface area
472 of the interim storage facilities in Futaba and Okuma municipalities is planned to cover ~1600 ha, and
473 by February 2019, a contract was already established between Japanese authorities and landowners
474 for ~70% of the land required for storage. Soil storage operations started in October 2017 in Okuma
475 and in December 2017 in Futaba. By March 2019, ~2.5 million m^3 of waste soil had already been
476 transported from the temporary storage facilities distributed across all the remediated area to these
477 two interim storage facilities (Japanese Ministry of the Environment, 2019b). All the soil waste is
478 planned to be transported to the Okuma and Futaba sites by the end of 2021 (Japanese Ministry of the
479 Environment, 2019a). The final disposal of this decontamination waste should take place outside of
480 the Fukushima Prefecture, within 30 years after the opening of the interim storage facilities (i.e.,
481 ~2047).

482 *Incineration*

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

495

496

497 *Incineration ash treatment*

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

507 *Soil recycling*

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

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

535 *Soil remediation*

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

542 residual radiocesium to plants, additives such as zeolite or Prussian blue adsorbents need to be
543 incorporated into the Andisols. The problem associated with this strategy is that, through their ageing,
544 zeolites may increase ^{137}Cs exchangeability with potassium and accelerate ^{137}Cs transfer to the
545 cultivated plants over longer time periods (Yamaguchi et al., 2019). These restrictions illustrate the
546 difficulty of finding alternatives to the storage of decontamination soil waste in interim facilities.
547
548 **7. Perspectives for future research**

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

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

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

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

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

Conclusions

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

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

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Figures

Figure 1. Evolution of the number of studies published on radiocesium and Fukushima (including or
not a reference to decontamination) in the literature between 2011–2018, according to the Scopus
search engine.
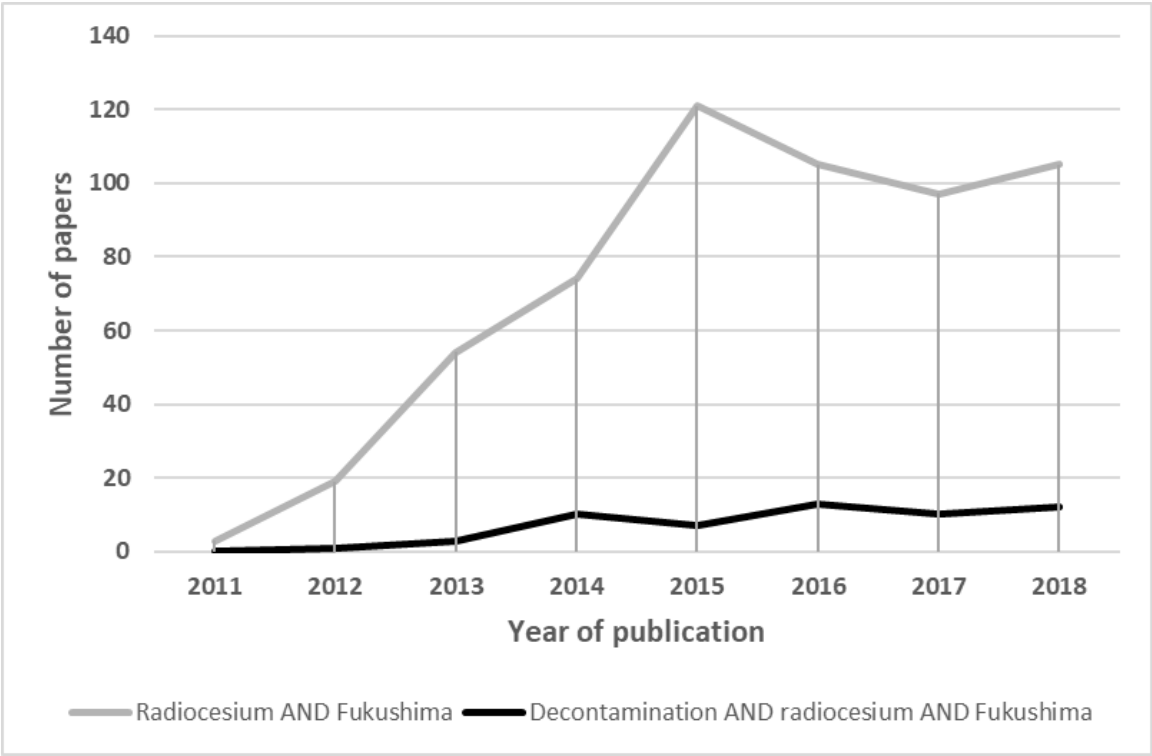
952 Figure 2. Location of the Fukushima Prefecture in Japan (Inset Map) and the location of the Special
953 Decontamination Zones (SDZs) and the Intensive Contamination Survey Areas (ICAs).

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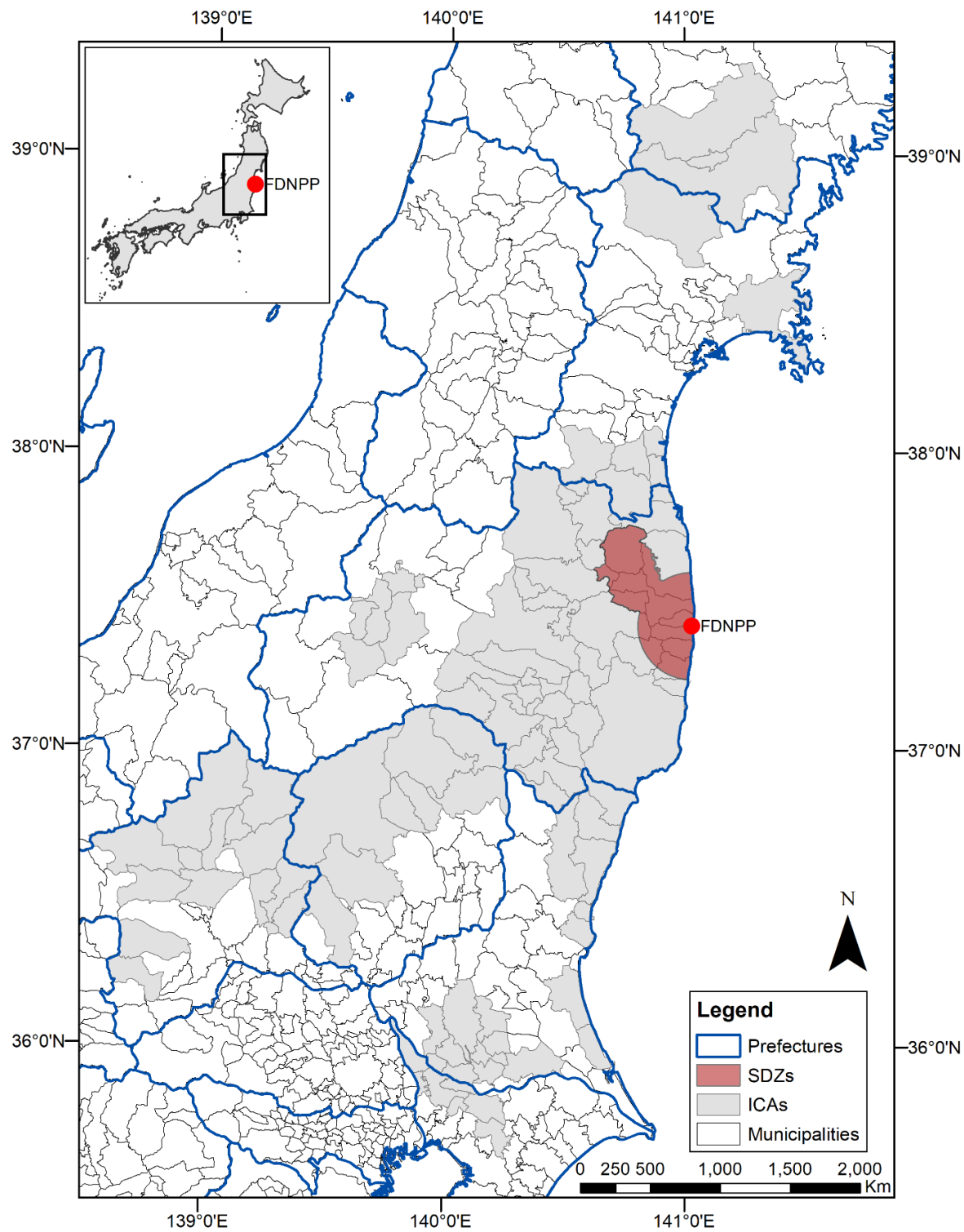
956 Figure 3. Illustration of the different steps of remediation activities in cultivated land in Fukushima: (a)
957 removal of the 5-cm topsoil layer concentrating most of the radiocesium (November 2013); (b)
958 addition of a crushed granite layer on top of the residual soil profile (May 2014); (c)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

961 (a)

962 (b)

963   (c)

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966   Figure 4. Illustration of the 20-m buffer zone decontaminated in forested areas in the vicinity of houses.
967   Example from Iitate Village (Sasu district).

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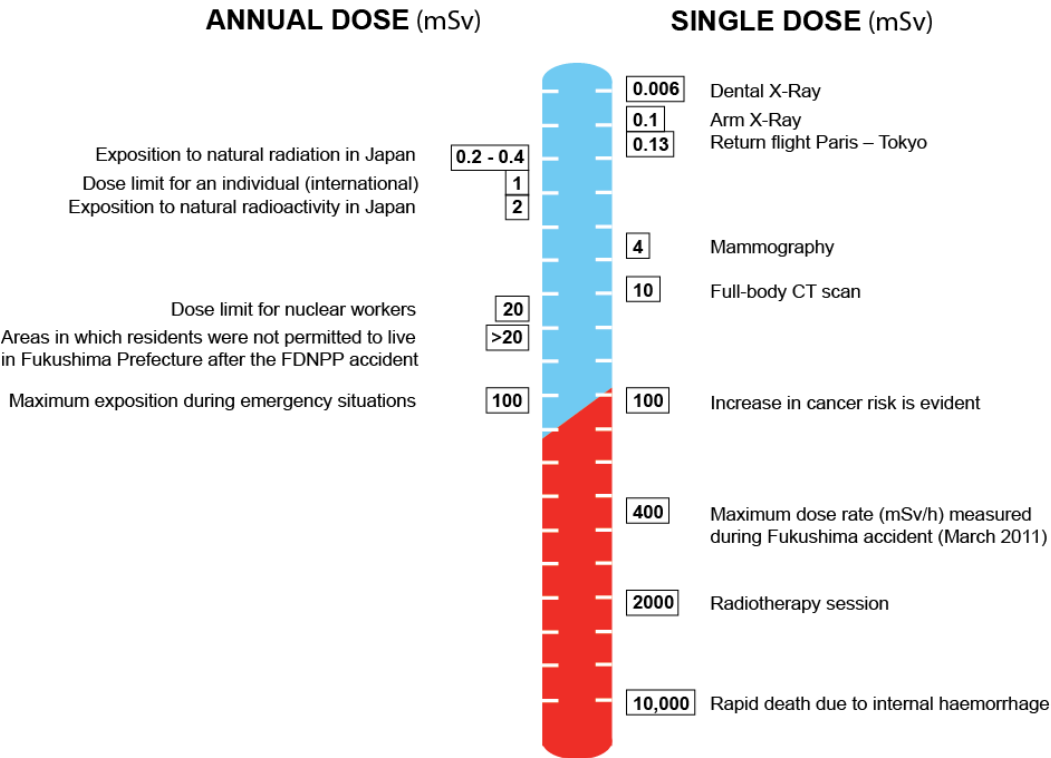972   exposed, based on a compilation of data (Commissariat à l'Energie Atomique et aux Energies
973   Alternatives, 2016;Harada et al., 2014).

ANNUAL DOSE (mSv)   **SINGLE DOSE** (mSv)

	0.006 — Dental X-Ray
	0.1 — Arm X-Ray
	0.13 — Return flight Paris – Tokyo
Exposition to natural radiation in Japan — 0.2 - 0.4	
Dose limit for an individual (international) — 1	
Exposition to natural radioactivity in Japan — 2	
	4 — Mammography
	10 — Full-body CT scan
Dose limit for nuclear workers — 20	
Areas in which residents were not permitted to live in Fukushima Prefecture after the FDNPP accident — >20	
Maximum exposition during emergency situations — 100	100 — Increase in cancer risk is evident
	400 — Maximum dose rate (mSv/h) measured during Fukushima accident (March 2011)
	2000 — Radiotherapy session
	10,000 — Rapid death due to internal haemorrhage

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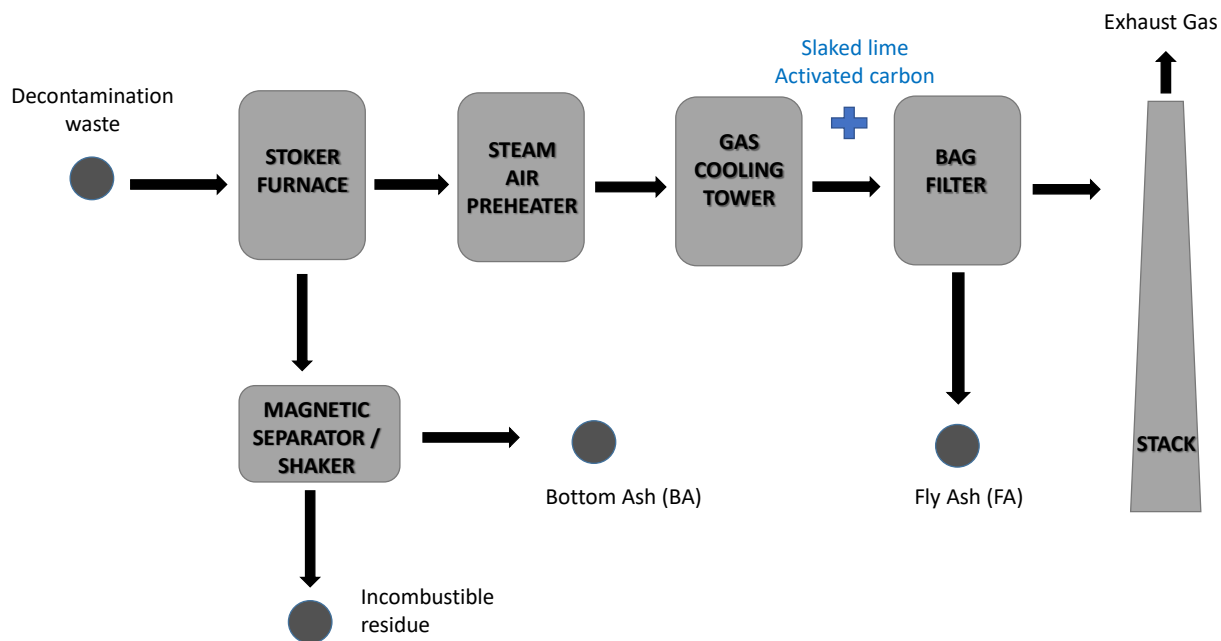991   Prefecture.

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997   Figure 7. Simplified diagram showing the functioning of an incineration plant treating decontamination
998   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).

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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/

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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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Code	Measure	Effectiveness range	Unit cost 10kJPY/ha (EUR)	Nb containers/ha	Target area
Agricultural land					
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
Forest					
F1	Remove litter and humus	0.19 – 0.59	745 (6000)	530	SDZ
F2	Remove litter	0.10 – 0.30	280 (2250)	260	ICA
Roads					
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
Buildings					
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
Treatment at interim storage facility		
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