

Depth distribution of cesium-137 in paddy fields across the Fukushima pollution plume in 2013

Hugo Lepage, O. Evrard, Yuichi Onda, Irène Lefèvre, J. Patrick Laceby, Sophie Ayrault

▶ To cite this version:

Hugo Lepage, O. Evrard, Yuichi Onda, Irène Lefèvre, J. Patrick Laceby, et al.. Depth distribution of cesium-137 in paddy fields across the Fukushima pollution plume in 2013. Journal of Environmental Radioactivity, 2015, 147, pp.157 - 164. 10.1016/j.jenvrad.2015.05.003. hal-01806119

HAL Id: hal-01806119

https://hal.science/hal-01806119

Submitted on 14 May 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Depth distribution of cesium-137 in paddy fields across the Fukushima pollution plume in 2013

Hugo Lepage^{a,*}, Olivier Evrard^a, Yuichi Onda^b, Irène Lefèvre^a, J. Patrick Laceby^a, Sophie Ayrault^a

^a Laboratoire des Sciences du Climat et de l'Environnement (LSCE/IPSL), Unité Mixte de Recherche 8212 (CEA, CNRS, UVSQ), 91198, Gif-sur-Yvette Cedex, France

^b Center for Research in Isotopes and Environmental Dynamics (CRIED), University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8572, Japan

* Corresponding author.

E-mail address: hugo.lepage@lsce.ipsl.fr (H. Lepage).

Telephone: +33/1/69.82.43.33

List of authors

Hugo LEPAGE

Laboratoire des Sciences du Climat et de l'Environnement

F-91198 Gif-sur-Yvette Cedex

France

E-mail: hugo.lepage@lsce.ipsl.fr

Tel: +33/1/69 82 43 33

Olivier Evrard

Laboratoire des Sciences du Climat et de l'Environnement

F-91198 Gif-sur-Yvette Cedex

France

E-mail: olivier.evrard@lsce.ipsl.fr

Tel: +33/1/69 82 35 20

Yuichi Onda

Graduate School of Life and Environmental Sciences,

University of Tsukuba,

1-1-1 Tennodai, Tsukuba, Ibaraki 305-8572, Japan

E-mail: onda@geoenv.tsukuba.ac.jp

Tel: +81-29-853-4226

Irène Lefèvre

Laboratoire des Sciences du Climat et de l'Environnement

F-91198 Gif-sur-Yvette Cedex

France

E-mail: <u>irene.lefevre@lsce.ipsl.fr</u>

Tel: +33/1/69 82 43 65

J. Patrick Laceby

Laboratoire des Sciences du Climat et de l'Environnement

F-91198 Gif-sur-Yvette Cedex

France

E-mail: placeby@lsce.ipsl.fr

Sophie Ayrault

Laboratoire des Sciences du Climat et de l'Environnement

F-91198 Gif-sur-Yvette Cedex

France

E-mail: sophie.ayrault@lsce.ipsl.fr

Tel: +33/1/69 82 43 54

*Highlights (for review)

Highlights

- Depth migration of ¹³⁷Cs were investigated in soil cores collected on contaminated paddy field.
- Major part of ¹³⁷Cs remain in the uppermost 5 cm even 30 months after the accident.
- More than 50% of contamination could remain in tilled soil after decontamination.
- Radiocesium migrate in the uppermost 3 cm after grass cutting.

Depth distribution of cesium-137 in paddy fields across the

2 Fukushima pollution plume in 2013

3

4

1

Abstract

5 Large quantities of radiocesium were deposited across a 3000 km² area northwest of the 6 Fukushima Dai-ichi nuclear power plant after the March 2011 accident. Although many studies have investigated the fate of ¹³⁷Cs in soil in the months following the accident, the depth 7 distribution of this radioactive contaminant in rice paddy fields requires further examination 8 9 after the typhoons that occurred in this region. Such investigations will help minimize potential human exposure in rice paddy fields. Radionuclide activity concentrations, organic content and 10 particle size were analysed in 10 soil cores sampled from paddy fields in November 2013, 20 11 km north of the Fukushima power plant. Our results demonstrate limited depth migration of 12 13 ¹³⁷Cs with the majority concentrated in the uppermost layers of soils (<5 cm). More than 30 months after the accident, between 46.8 to 98.7% of the total ¹³⁷Cs inventories was found within 14 15 the top 5 cm of the soil surface, despite cumulative rainfall totalling 3300 mm. Furthermore, there were no significant correlations between ¹³⁷Cs depth distribution and the other parameters. 16 We attributed the maximum depth penetration of ¹³⁷Cs to grass cutting (73.6-98.5% of ¹³⁷Cs in 17 the upper 5 cm) and farming operations (tillage -46.8-51.6% of 137 Cs in the upper 5 cm). As 18 this area is exposed to erosive events, ongoing decontamination works may increase soil 19 20 erodibility. We therefore recommend the rapid removal of the uppermost – contaminated – 21 layer of the soil after removing the vegetation to avoid erosion of contaminated material during 22 the subsequent rainfall events. Further analysis is required to thoroughly understand the impacts 23 of erosion on the redistribution of radiocesium throughout the Fukushima Prefecture.

24

25

1 Introduction

- The Tohoku earthquake and the subsequent tsunami on March 11, 2011 resulted in the Fukushima Dai-Ichi Nuclear Power Plant (FDNPP) accident and the significant corresponding atmospheric release of radionuclides, such as ¹³⁷Cs (T_{1/2} = 30 years) (Saunier et al., 2013). Approximately 80% of the release was transported over the Pacific Ocean with the remainder
- 30 predominantly deposited on Fukushima Prefecture soils as a result of wet atmospheric fallout

(Kawamura et al., 2011). Estimations of ¹³⁷Cs total activity in the Fukushima Prefecture soils range between 10 PBq and 50 PBq, with deposition characterized by strong spatial heterogeneities (Koo et al., 2014). The highest activities are concentrated within a 70 km long radioactive plume where initial ¹³⁷Cs contamination exceeded 300 kBq.m⁻² covering an area of 3000 km². Therefore it is crucial to understand and monitor the fate of the initial radioactive deposits in order to protect the local population against exposure to high dose rates due to gamma radiation that may prevail in areas accumulating contamination.

In the coastal catchments affected by the FDNPP accident, Chartin et al. (2013) showed that

39

40

41

42

43

44

45

46

47

48

49

50

51

52

53

54

55

56

57

58

59

60

61

In the coastal catchments affected by the FDNPP accident, Chartin et al. (2013) showed that paddy fields are one of the major sources of ¹³⁷Cs mobilization and export by soil erosion. A significant proportion of paddy fields are located in the upstream area of the contaminated catchments and they were shown to supply large quantities of contaminated sediment to rivers during typhoons and snowmelt events (Evrard et al., 2013; Evrard et al., 2014). Dispersion of contamination originating from paddy fields along the rivers of the region could therefore contaminate downstream areas that were affected to a limited extent by the initial fallout. In this region, background levels of ¹³⁷Cs from fallout from atmospheric nuclear weapons testing were estimated to be under 100Bq.kg⁻¹ (Fukuyama et al., 2005). Several studies have shown that ¹³⁷Cs has a low mobility in most soils (Beck 1966, Ivanov et al., 1997) and is rapidly fixed to fine particles, especially clay minerals (Sawhney, 1972; He & Walling, 1996). These findings were confirmed in the vicinity of the main contamination plume in the Fukushima Prefecture where Saito et al. (2014) reported that ¹³⁷Cs was concentrated in the silt and clay fractions. Also, it was reported that the majority of ¹³⁷Cs remained in the first centimetre of the soil profile (Fujiwara et al., 2012; Kato et al., 2012; Koarashi et al., 2012; Lepage et al., 2014). However, it was also shown that in soils with high levels of organic matter, radiocesium may migrate down the soil profile as organic matter may reduce its affinity with clay minerals (Kamei-Ishikawa et al., 2008; Koarashi et al., 2012; Staunton et al., 2002; Szenknect et al., 2003). Koarashi et al. (2012) analysed several soil profiles sampled in the vicinity of Fukushima City and showed that clay content and Total Organic Carbon (TOC) were the main factors controlling radiocesium depth migration. They also found a strong negative correlation (r = -0.79, p < 0.005) between TOC content divided by clay content (TOC/Clay) and the percentage of retention of ¹³⁷Cs suggesting that the presence of organic matter inhibits the adsorption of ¹³⁷Cs on clay minerals.

- Thirty months after the accident, it is important to investigate the depth distribution of ¹³⁷Cs in
- a selection of paddy fields located within the main contamination plume of Fukushima
- Prefecture. The concentration of ¹³⁷Cs in the uppermost layers of the soil could make it available
- 65 for erosion and delivery to nearby rivers. This investigation of ¹³⁷Cs depth distribution in paddy
- 66 fields is particularly timely in the current post-accidental phase characterised by the
- 67 implementation of large-scale remediation efforts targeting paddy fields. The implications of
- these findings for potential soil erosion will be specifically discussed.

2 Materials and methods

2.1 Study area

69

70

- 71 The study was conducted in Fukushima Prefecture, located in North-Eastern Japan, 30 km
- 72 northwest of FDNPP (Fig. 1). We focused our work on two coastal catchments (i.e. Mano and
- Niida River catchments 450km²) draining the main part of the radioactive plume. These
- catchments extend from the coastal mountain range (approximately 30 km from the coast) to
- 75 the Pacific Ocean, and their elevation ranges from 0 to 900 m. Mean annual rainfall was 1320
- 76 mm according to Japanese Meteorological Agency (2014) measured over 37 years at the rainfall
- station located in the upper part of the Nitta catchment (Fig. 1). Our study was conducted in
- November 2013 and cumulative rainfall reached 3300mm (max = 35mm.h⁻¹) between the
- accident and our field survey (32 months) with the occurrence of 4 typhoons (Songda and Roke
- 80 in 2011, Man-Yi and Wipha in 2013). In these catchments, paddy fields are predominantly
- 81 located along the rivers (Tanaka et al., 2013).
- 82 Remediation works implemented since July 2012 under the supervision of the Japanese
- 83 Ministry Of Environment, Government of Japan (MOE, 2012a; MOE, 2013) are concentrated
- 84 in upper parts of the Nitta River catchment and consist of removing the five uppermost
- 85 centimetres of the soil (Mizoguchi, 2013; Sakai et al., 2014) to decrease the radioactive dose
- level in order to avoid exceeding the permissible level (1 mSv y⁻¹) determined by Japanese
- authorities (MOE, 2012b).

88

2.2 Sample collection and preparation

- 89 A radiameter (LB123 D-H10, Berthold Technologies) was used to measure radiation dose rates
- at the ground level in the paddy fields (Table 1). To be representative, dose levels were
- 91 measured at 5 different locations on each field within 10m². The formula Eq. (1) proposed by

- 92 the Ministry of Environment MOE (2012b) was then applied to convert these data into annual
- 93 dose rates.

$$94 D_{an} = \frac{(D_{amb} - 0.04)* (8 + 16 * 0.4)* 365}{1000} (1)$$

- Where D_{amb} is the ambient dose rate, and D_{an} is the annual dose rates.
- Soil cores (Table 1) were collected in paddy fields across the two selected catchments (Fig 1).
- 97 We selected the fields to cover a range of dose rates (low, medium and high) in order to
- 98 investigate migration of ¹³⁷Cs in fields with different levels of contamination (Table 1).
- A soil auger (diameter 45 mm) was used to sample soil cores to a depth of 10 cm from 10 fields.
- The soil cores were sub-sectioned into 1 cm increments for the uppermost 5 cm (0-1; 1-2; 2-3;
- 3-4; 4-5 cm) and an additional 5 cm increment was taken from 5 cm to 10 cm (5-10 cm).
- Because of the very high radioactive dose rate measured at the location where core P9 was
- sampled (5.5 µsv.h⁻¹), two additional layers were sampled at greater depths (10-15 cm and 15-
- 104 20 cm). Density of the soil was determined by dividing the dry soil mass in each layer by its
- volume determined from the diameter of the soil auger and the thickness of the layer. Mean
- 106 compaction in the cores was estimated to be 13% using soil density. Soil TOC content was
- measured with the dry combustion method (VarioTOC, Elementar, LAME, IRSN, Fontenay
- aux Roses, France) and clay content (particle size < 10 µm) of the samples was measured using
- a laser diffraction system (Malvern Mastersizer 2000) coupled to a liquid dispersing unit (Hydro
- 110 2000G, University of Paris Sud, Orsay, France). XRD measurement were performed on a
- diffractometer (INEL XRG 3000, L3MR, CEA, Saclay, France) equipped with a curved
- position sensitive detector CPS 120 and radiation were produced at 30 kV and 30 mA.

113 **2.3 Gamma spectrometry measurements**

- Before measurement, samples were dried in an oven at 40°C for a week, ground to a fine powder
- in an agate mortar, and then packed into 15 mL polyethylene specimen containers. Cesium-137
- activities were determined by gamma spectrometry using low-background coaxial N- and P-
- types HPGe detectors (Canberra/Ortec). Counting times of samples varied between 80 000 s
- and 150 000 s. The ¹³⁷Cs activities were measured at the 661 keV emission peak. Counting
- efficiencies and energy calibration were monitored using internal, national (IRSN) and certified
- 120 International Atomic Energy Agency (IAEA) reference materials (i.e., IAEA-135, IAEA-375,
- 121 IAEA-CU-2006-03, IAEA-Soil-6, RGU-1, and RGTh-1) prepared in the same specimen

- 122 containers as the samples. Uncertainties were estimated by combining counting statistics and
- 123 calibration uncertainties. Summing and self-absorption effects were taken into account by
- measuring reference materials with similar densities and characteristics as the collected
- samples. All activities were decay corrected to the date of 14 March 2011 corresponding to the
- date of the main radionuclide deposits on soils (Kinoshita et al., 2011; Shozugawa et al., 2012).

2.4 Distribution of ¹³⁷Cs with depth

- Radiocesium profiles in undisturbed soils are expected to display an exponential decline with
- depth (He & Walling, 1997; Walling & He, 1999), which can be described by the following
- 130 function (Beck, 1966):

131
$$C_{(x)} = C_0 * \exp^{-\alpha x}$$
 (2)

- Where C(x) is the concentration activity of a radionuclide in Bq.kg⁻¹ at the depth x (cm) and C₀
- at x=0, and α (cm⁻¹) is a coefficient representing the characteristics of the radionuclide
- distribution and depends on different characteristics of the soil (e.g. pH, CEC, TOC, clay
- 135 content).

127

- In addition, a more specific model can be used to describe ¹³⁷Cs mobility in soils (Kato et
- 137 al., 2012; Koarashi et al., 2012; Miller et al., 1990):

138
$$I_{(x)} = I_t (1 - \exp^{-z/h_0})$$
 (3)

- Where $I_{(z)}$ is the ¹³⁷Cs inventory (Bq.m⁻²) at the z (kg.m⁻²) depth, I_t is the total ¹³⁷Cs inventory
- and h_0 is the relaxation mass depth (kg.m²), an index characterising the ¹³⁷Cs penetration in the
- soil. The greater the value of the relaxation mass depth h₀, the deeper the ¹³⁷Cs penetrates into
- the soil profile (Kato et al., 2012).
- In order to use this formula, we calculated the ¹³⁷Cs inventory using ¹³⁷Cs activities (Bq.kg⁻¹)
- (Lee et al., 2013) at the successive (x) depths (Eq. 4):

145
$$I(x) = \rho(x) * h(x) * A(x)$$
 (4)

- Where ρ is the dry bulk density (g.m⁻³), h is the thickness (cm) of the layer and A the activity
- 147 (Bq.kg⁻¹) of this layer.

3 Results and discussion

3.1 Depth distribution

- 150 Based on the distribution of ¹³⁷Cs activity with depth, soil cores were classified into four groups
- (undisturbed, decontaminated, tilled and managed) (Table 2 Fig 2):
- Undisturbed: distribution shown an exponential decrease of ¹³⁷Cs activity with depth (Fig.
- 153 3),

148

149

- Decontaminated: very low level of ¹³⁷Cs activity in all layers,
- Tilled: similar level of ¹³⁷Cs activity in each layer,
- Managed: similar level of contamination in the upper 2-3cm then exponential decrease
- with depth of the ¹³⁷Cs activity.
- 158 The group of undisturbed fields (P6, P7 and P9) remained abandoned by the end of 2013 as
- they were protected by a dense grass cover (Fig 4a) and they showed an exponential decrease
- of ¹³⁷Cs activities with depth (Fig 3). Furthermore, our results on relaxation mass depths h₀ in
- undisturbed soils (Table 2) varied from 5.4 to 8.3 and remained in the same range as previous
- results found for soils collected in this area (Table 3). Although this result indicates a limited
- migration of ¹³⁷Cs, contamination from the FDNPP accident could still be found at the 10-15
- cm and 15-20 cm layers in P9 (respectively 370 and 170 Bq.kg⁻¹, Fig 2). This is likely due to
- the natural migration of ¹³⁷Cs as this soil core was heavily contaminated (155 kBq.kg⁻¹) and
- 166 contained 8.5% of TOC. Nevertheless, level of contamination corresponded to less than 10%
- of total contamination of P9, as observed in the other undisturbed soil cores.
- 168 The core P5 did not show evidence of additional contamination of ¹³⁷Cs due to Fukushima
- accident (Fig 2) as at this site the ¹³⁷Cs concentration levels remained similar to pre-accident
- 170 conditions, which were estimated not to exceed 100 Bq.kg⁻¹ in Japanese soils (Fukuyama et al.,
- 171 2005). However, the detection of ¹³⁴Cs confirms that this field was contaminated by the
- 172 Fukushima fallout and subsequently decontaminated by remediation efforts (Fig 4b -
- 173 Mizoguchi, 2013; Sakai et al., 2014).
- P2 and P4 were disturbed after the initial radionuclide deposition, resulting in a homogenization
- of activities in successive soil layers (Fig 2; Fig 3). These fields were most likely tilled by
- farmers (Endo et al., 2013; Matsunaga et al., 2013; Yamaguchi et al., 2012) and now around

51% of the total contamination inventory is found deeper in the soil (i.e. 5-10 cm) (Table 2). This percentage could be higher (\approx 66%) as tilled soils generally showed a similar level of contamination until 15 cm (Endo et al., 2013; Yamaguchi et al., 2012). Thus, a greater sampling depth should be considered for future investigations on 137 Cs migration in Fukushima soils. The ongoing farming operations in this area can be explained by the fact that those fields are not located in the evacuation-prepared area and that cultivation is allowed (Fig 1).

In contrast, managed fields (P1, P3, P8 and P10) showed a similar level of contamination in the upper 2-3 centimetres and then a decrease (Fig 2). These four fields have been continuously managed since the accident, as illustrated by our field observations during previous campaigns (Nov. 2011, April 2012 and May 2013). Grass was cut each year using heavy machinery (Fig 4c), which may explain the mixing of soil and associated ¹³⁷Cs in these fields due to the compaction of the first few centimetres of the soil (Jagercikova et al., 2014; Matsunaga et al., 2013). Takahashi et al. (2014) also reported the same migration in the uppermost 3-centimetres and concluded that it was caused by the repeated formation and melting of needle ice in the surface soil during winter and spring. Further investigation is required to clarify the process involved in this migration. However, our results demonstrated that 73.6 to 98.5% of the ¹³⁷Cs contamination was concentrated in the upper 5 cm (Table 2).

Contrary to the results obtained by Koarashi et al. (2012), no significant correlation was found between TOC, clay content, TOC/Clay and both the α coefficient and the relaxation mass depth h_0 (Table 4) in both managed and undisturbed soils (number of samples = 7). As the depth distribution of 137 Cs in soils does not vary with the soil parameters, the different 137 Cs migration observed between undisturbed and managed fields is most likely explained by the type and frequency of farming operations carried out between the nuclear accident and the sampling campaign. The limited migration of 137 Cs could be attributed to the presence of phyllosilicate minerals as identified by XRD analyses (data not presented). These results confirm those found for undisturbed soils located under different land uses in the vicinity of the FDNPP by previous studies (Table 3). Most of these studies concluded that 137 Cs was exclusively found in the upper 5 cm for undisturbed soil in forests (mean of 137 Cs inventory = $94.8\pm2.5\%$) and in fields ($98.7\pm1.8\%$). Our results on tilled soils are also consistent with those from previous publications (Endo et al., 2013; Koarashi et al., 2012; Matsunaga et al., 2013; Tanaka et al., 2013).

To complement the published research conducted a few months after the accident (Fig 5 – Table 4), our results showed that even more than 30 months after the accident and after the occurrence of several typhoons (Fig 5), the depth migration of 137 Cs was very limited with the majority (73.6 – 98.7%) of this radionuclide still found in the upper 5 centimetres of undisturbed and managed soils. These results are complementary with the study of Mastunaga et al. (2013) who concluded that radiocesium did not migrate with depth even after rainfalls, 5 months after the accident. Tilling is the main contributor of the migration of radiocesium in soil as the main part of contamination is under the first layers (48.4 – 53.2%) in tilled fields (Table 3).

208

209

210

211

212

213

214

215

216

217

218

219

220

221

222

223

224

225

226

227

228

229

230

231

232

233

234

235

236

3.2 Implications for contamination redistribution across the catchments

In most of the investigated soil cores, contamination was concentrated in the upper layers of the soils and is therefore potentially available for soil erosion (Motha & Wallbrink, 2002; Walling & Woodward, 1992). However, in abandoned fields, the dense grass cover will protect the soil against erosion as soil erodibility is mainly controlled by the vegetation cover, surface roughness and crusting characteristics (Evrard et al., 2008; Le Bissonnais et al., 2005). During decontamination operations, the grass is cut and vegetation is removed leaving the soil bare and exposed to the erosive impact of intense rainfall. The specific supply of contaminated sediment from fields where remediation efforts were concentrated to nearby rivers was demonstrated by Evrard et al (2014) who showed an increase of radioactive dose rates in recent sediment drape deposits collected in upper parts of the Nitta River catchment after the heavy typhoons that occurred during summer in 2013. In this context, we recommend rapidly removing the first few centimetres of the soil immediately after removing the vegetation to avoid erosion of contaminated material during the subsequent rainfall events. Alternatively, remediation efforts could be concentrated before July or after October, when typhoons are unlikely to occur. We do not recommend mixing soil with water (puddling) in contaminated fields, as this could export the contamination contained in the uppermost layers of the soil (Wakahara et al., 2013). Furthermore, contaminated soils

that have been eroded and transported by the river could be deposited on top of already

decontaminated soil (Sakai et al. 2014). To avoid this recontamination, we recommend to start

the decontamination works in the catchment headwaters and to proceed downstream.

4 Concluding remarks

Depth distributions of ¹³⁷Cs investigated in 10 paddy fields varied with land management. The maximum depth penetration of ¹³⁷Cs collected in November 2013 was attributed to maintenance and farming operations in paddy fields contaminated after the FDNPP accident (March 2011). In tilled fields, the contamination showed a similar level in each layer (0-10cm) while in managed field, where vegetation was removed, contamination only migrated down the first 2-3 cm. In undisturbed soils, ¹³⁷Cs inventory decreased with depth following an exponential function. Our results confirm the overall limited migration of ¹³⁷Cs concentrated in the first few centimetres (<5 cm) of soil in undisturbed and managed paddy fields located within the main Fukushima contamination plume.

247

248

237

238

239

240

241

242

243

244

245

246

Author contribution

- 249 H.L. and O.E. wrote the main manuscript; O.E. and Y.O. designed research; H.L. and O.E.
- conducted field sampling; H.L. and I.L. conducted laboratory measurements. Y.O., J.P. L. and
- 251 S.A. participated to the redaction and reviewed the manuscript.

252

253

Acknowledgements

- 254 This work has been supported by ANR (French National Research Agency) and JST (Japan
- 255 Science and Technology agency) in the framework of the joint TOFU ANR Flash/J-RAPID
- Franco-Japanese project (ANR-11-JAPN-001). This work has also been supported by CNRS in
- 257 the framework of the NEEDS-Environnement HAMUSUTA project, and by the Institute of
- 258 Environmental Radioactivity (IER) in Fukushima University. Finally, funding from ANR in the
- 259 framework of the AMORAD project (ANR-11-RSNR-0002) is gratefully acknowledged. Hugo
- 260 Lepage received a PhD fellowship from CEA (Commissariat à l'Energie Atomique et aux
- 261 Energies Alternatives, France). The authors would like to thank A. Ritt for conducting TOC
- measurement and C. Latrille for XRD measurement.

263

264 References

- 265 Beck, H.L., 1966. Environmental gamma radiation from deposited fission products. Health
- 266 Phys. 12, 313.
- 267 Chartin, C., Evrard, O., Onda, Y., Patin, J., Lefèvre, I., Ottlé, C., Ayrault, S., Lepage, H., Bonté,
- 268 P., 2013. Tracking the early dispersion of contaminated sediment along rivers draining the
- Fukushima radioactive pollution plume. Anthropocene 1, 23–34.
- 270 Endo, S., Kajimoto, T., Shizuma, K., 2013. Paddy-field contamination with 134Cs and 137Cs
- due to Fukushima Dai-ichi Nuclear Power Plant accident and soil-to-rice transfer coefficients.
- 272 J. Environ. Radioact. 116, 59-64
- Evrard, O., Vandaele, K., Bielders, C., Van Wesemael, B., 2008. Seasonal evolution of runoff
- 274 generation on agricultural land in the Belgian loess belt and implications for muddy flood
- triggering. Earth Surf. Process. Landforms 1301, 1285–1301.
- Evrard, O., Chartin, C., Onda, Y., Patin, J., Lepage, H., Lefèvre, I., Ayrault, S., Ottlé, C., Bonté,
- 277 P., 2013. Evolution of radioactive dose rates in fresh sediment deposits along coastal rivers
- draining Fukushima contamination plume. Sci. Rep. 3, 3079.
- Evrard, O., Chartin, C., Onda, Y., Lepage, H., Cerdan, O., Lefèvre, I., Ayrault, S., 2014.
- 280 Renewed soil erosion and remobilisation of radioactive sediment in Fukushima coastal rivers
- 281 after the 2013 typhoons. Sci. Rep. 4, 4574.
- Fujiwara, T., Saito, T., Muroya, Y., 2012. Isotopic ratio and vertical distribution of
- radionuclides in soil affected by the accident of Fukushima Dai-ichi nuclear power plants. J.
- 284 Environ. Radioact. 113, 37–44.
- Fukuyama, T., Takenaka, C., Onda, Y., 2005. 137Cs loss via soil erosion from a mountainous
- headwater catchment in central Japan. Sci. Total Environ. 350, 238–47.
- 287 He, Q., Walling, D., 1996. Interpreting particle size effects in the adsorption of 137Cs and
- unsupported 210Pb by mineral soils and sediments. J. Environ. Radioact. 30, 117–137.
- He, O., Walling, D., 1997. The distribution of fallout 137 Cs and 210 Pb in undisturbed and
- 290 cultivated soils. Appl. Radiat. Isot. 48, 677–690.
- Ivanov, Y., Lewyckyj, N., Levchuk, S., 1997. Migration of 137 Cs and 90 Sr from Chernobyl
- fallout in Ukrainian, Belarussian and Russian soils. J. Environ. Radioact. 35.

- Jagercikova, M., Evrard, O., Balesdent, J., Lefèvre, I., Cornu, S., 2014. Modeling the migration
- of fallout radionuclides to quantify the contemporary transfer of fine particles in Luvisol
- 295 profiles under different land uses and farming practices. Soil Tillage Res. 140, 82–97.
- 296 Japanese Meteorological Agency:
- 297 http://www.data.jma.go.jp/obd/stats/etrn/view/annually_a.php?prec_no=36&block_no=1150
- 298 &year=2013&month=4&day=&view=, last access: 28 July 2014 (in Japanese)
- 299 Kamei-Ishikawa, N., Uchida, S., Tagami, K., 2008. Distribution coefficients for 85Sr and
- 300 137Cs in Japanese agricultural soils and their correlations with soil properties. J. Radioanal.
- 301 Nucl. Chem. 277, 433–439.
- Kato, H., Onda, Y., Teramage, M., 2012. Depth distribution of 137Cs, 134Cs, and 131I in soil
- profile after Fukushima Dai-ichi Nuclear Power Plant Accident. J. Environ. Radioact. 111, 59–
- 304 64.
- 305 Kawamura, H., Kobayashi, T., Furuno, A., In, T., Ishikawa, Y., Nakayama, T., Shima, S.,
- 306 Awaji, T., 2011. Preliminary numerical experiments on oceanic dispersion of 131I and 137Cs
- discharged into the ocean because of the Fukushima Daiichi nuclear power plant. J. Nucl. Sci.
- 308 Technol. 48, 1349–1356.
- Kinoshita, N., Sueki, K., Sasa, K., Kitagawa, J., Ikarashi, S., Nishimura, T., Wong, Y.-S., Satou,
- 310 Y., Handa, K., Takahashi, T., Sato, M., Yamagata, T., 2011. Assessment of individual
- 311 radionuclide distributions from the Fukushima nuclear accident covering central-east Japan.
- 312 Proc. Natl. Acad. Sci. U. S. A. 108, 19526–9.
- Koarashi, J., Atarashi-Andoh, M., Matsunaga, T., Sato, T., Nagao, S., Nagai, H., 2012. Factors
- 314 affecting vertical distribution of Fukushima accident-derived radiocesium in soil under
- different land-use conditions. Sci. Total Environ. Environ. 431, 392–401.
- 316 Koo, Y.-H., Yang, Y.-S., Song, K.-W., 2014. Radioactivity release from the Fukushima
- accident and its consequences: A review. Prog. Nucl. Energy 74, 61–70.
- Le Bissonnais, Y., Cerdan, O., Lecomte, V., Benkhadra, H., Souchère, V., Martin, P., 2005.
- 319 Variability of soil surface characteristics influencing runoff and interrill erosion. Catena 62,
- 320 111–124.

- Lee, S.H., Oh, J.S., Lee, J.M., Lee, K.B., Park, T.S., Lujaniene, G., Valiulis, D., Sakalys, J.,
- 322 2013. Distribution characteristics of 137Cs, Pu isotopes and 241Am in soil in Korea. Appl.
- 323 Radiat. Isot. 81, 315–20.
- Lepage, H., Evrard, O., Onda, Y., Patin, J., Chartin, C., Lefèvre, I., Bonté, P., Ayrault, S., 2014.
- 325 Environmental mobility of 110mAg: lessons learnt from Fukushima accident (Japan) and
- potential use for tracking the dispersion of contamination within coastal catchments. J. Environ.
- 327 Radioact. 130, 44–55.
- 328 Matsunaga, T., Koarashi, J., Atarashi-Andoh, M., Nagao, S., Sato, T., Nagai, H., 2013.
- 329 Comparison of the vertical distributions of Fukushima nuclear accident radiocesium in soil
- before and after the first rainy season, with physicochemical and mineralogical interpretations.
- 331 Sci. Total Environ. 447, 301–14.
- 332 Ministry of Economy, Trade and Industry (METI), 2014. Areas to which evacuation orders
- 333 have been issued (April 1, 2014),
- http://www.meti.go.jp/english/earthquake/nuclear/roadmap/pdf/140401MapOfAreas.pdf
- 335 Ministry of Education, Culture, Sports, Science and Technology (MEXT), 2012. MEXT, Japan
- http://radioactivity.nsr.go.jp/ja/contents/7000/6289/24/203_0928.pdf (In Japanese)
- 337 Miller, K.M., Kuiper, J.L., Heifer, I.K., 1990. 137Cs Fallout Depth Distributions in Forest
- Versus Field Sites: Implications for External Gamma Dose Rates. J. Environ. Radioact. 12, 23–
- 339 47.
- 340 Mizoguchi, M., 2013. Remediation of Paddy Soil Contaminated by Radiocesium in litate
- Village in Fukushima Prefecture. Agric. Implic. Fukushima Nucl. 131–142.
- Ministry of the Environment, Government of Japan (MOE), 2012a, http://josen.env.go.jp/en/
- 343 Ministry of the Environment, Government of Japan (MOE), 2012b,
- 344 http://josen.env.go.jp/osen/osen_05.html (In Japanese)
- 345 Ministry of the Environment, Government of Japan (MOE), 2013. Progress on Off-site Cleanup
- efforts in Japan. http://josen.env.go.jp/en/documents/pdf/workshop_july_17-18_2013_01.pdf
- 347 Motha, J., Wallbrink, P., 2002. Tracer properties of eroded sediment and source material.
- 348 Hydrol. Process. 16, 1983–2000.

- National Institute for Agro-Environmental Sciences (NIAES), 1996. Classification of cultivated
- soils in Japan: third approximation, Classification Committee of Cultivated Soils.
- 352 National Institute for Agro-Environmental Sciences (NIAES), digital cropland soil map of the
- Fukushima prefecture. http://agrimesh.dc.affrc.go.jp/soil_db/map_select_figure.phtml (Last
- 354 access 05/01/2015 In Japanese)
- Saito, T., Makino, H., Tanaka, S., 2014. Geochemical and grain-size distribution of radioactive
- and stable cesium in Fukushima soils: implications for their long-term behavior. J. Environ.
- 357 Radioact. 138C, 11–18.
- 358 Sakai, M., Gomi, T., Nunokawa, M., Wakahara, T., Onda, Y., 2014. Soil removal as a
- 359 decontamination practice and radiocesium accumulation in tadpoles in rice paddies at
- 360 Fukushima. Environ. Pollut. 187, 112–5.
- 361 Saunier, O., Mathieu, a., Didier, D., Tombette, M., Quélo, D., Winiarek, V., Bocquet, M., 2013.
- 362 An inverse modeling method to assess the source term of the Fukushima Nuclear Power Plant
- accident using gamma dose rate observations. Atmos. Chem. Phys. 13, 11403–11421.
- 364 Sawhney, B., 1972. Selective sorption and fixation of cations by clay minerals: a review. Clays
- 365 and Clay Minerals. 20, 93–100.
- 366 Shozugawa, K., Nogawa, N., Matsuo, M., 2012. Deposition of fission and activation products
- after the Fukushima Dai-ichi nuclear power plant accident. Environ. Pollut. 163, 243–7.
- 368 Staunton, S., Dumat, C., Zsolnay, a, 2002. Possible role of organic matter in radiocaesium
- adsorption in soils. J. Environ. Radioact. 58, 163–73.
- 370 Szenknect, S., Gaudet, J.P., Dewiere, L., 2003. Evaluation of distribution coefficients for the
- prediction of strontium and cesium migration in a natural sand at different water contents. J.
- 372 Phys. IV 107, 1279–1282.
- Takahashi, J., Tamura, K., Suda, T., 2014. Vertical distribution and temporal changes of 137
- 374 Cs in soil profiles under various land uses after the Fukushima Dai-ichi Nuclear Power Plant
- accident. J. Environ. Radioact. 1-11
- 376 Tanaka, K., Takahashi, Y., Sakaguchi, A., 2012. Vertical profiles of iodine-131 and cesium-
- 377 137 in soils in Fukushima prefecture related to the Fukushima Daiichi Nuclear Power Station
- 378 accident. Geochem. J. 46, 73–76.

- Tanaka, K., Iwatani, H., Takahashi, Y., Sakaguchi, A., Yoshimura, K., Onda, Y., 2013.
- 380 Investigation of spatial distribution of radiocesium in a paddy field as a potential sink. PLoS
- 381 One 8, e80794.
- Teramage, M.T., Onda, Y., Patin, J., Kato, H., Gomi, T., Nam, S., 2014. Vertical distribution
- of radiocesium in coniferous forest soil after the Fukushima nuclear power plant accident. J.
- 384 Environ. Radioact. 137, 37–45.
- Wakahara, T., Onda, Y., Kato, H., 2013. Estimation of radionuclide discharge from paddy fields
- in two experimental plots with different initial scrapings after the Fukushima Dai-ichi Nuclear
- Power Plant accident 1–3.
- Walling, D.E., He, Q., 1999. Improved models for estimating soil erosion rates from cesium-
- 389 137 measurements. J. Environ. Qual. 28, 611–622.
- Walling, D.E., Woodward, J.C., 1992. Use of radiometric fingerprints to derive information on
- 391 suspended sediment sources. Eros. Sediment Transp. Monit. Program. River Basin 210, 153-
- 392 164.
- 393 Yamaguchi, N., Eguchi, S., Fujiwara, H., Hayashi, K., Tsukada, H., 2012. Radiocesium and
- 394 radioiodine in soil particles agitated by agricultural practices: field observation after the
- 395 Fukushima nuclear accident. Sci. Total Environ. 425, 128–34.

Figure captions

Click here to download Figure: Figure captions.docx

Figure captions

Figure 1. Map of the study area with location of the soil cores collected within Mano and Nitta River catchments including ¹³⁷Cs soil inventory decay corrected to the date of 14 June 2011 based on the Japanese Ministry of Education, Culture, Sports, Science and Technology data (MEXT, 2012) and April 2014 restricted access areas delineated by Ministry of Economy, Trade and Industry (METI, 2014).

Figure 2. Depth distribution of 137 Cs activity in soil cores. Data on the 4-5 cm layer for P6 were not available (n/a).

Figure 3. Depth distribution of ¹³⁷Cs inventory in the different groups of contaminated soil cores.

Figure 4. Pictures taken during the sampling campaign (November 2013) and illustrating the difference of land management practices in the field, a) P9 - dense cover of grass on the field showed an absence of land management, b) P5 - land management in the field showed by tractor tracks and c) P8 - grass recently cut and presence of straw residues on the field.

Figure 5. Daily rainfall between FDNPP accident and this sampling campaign. Occurrence of typhoons is indicated on the graph. Timing of sampling campaigns of previous studies dealing with ¹³⁷Cs migration in soils is also indicated. Takahashi et al. (2014) also sampled at (1) (2) and (3).

Figure 1.

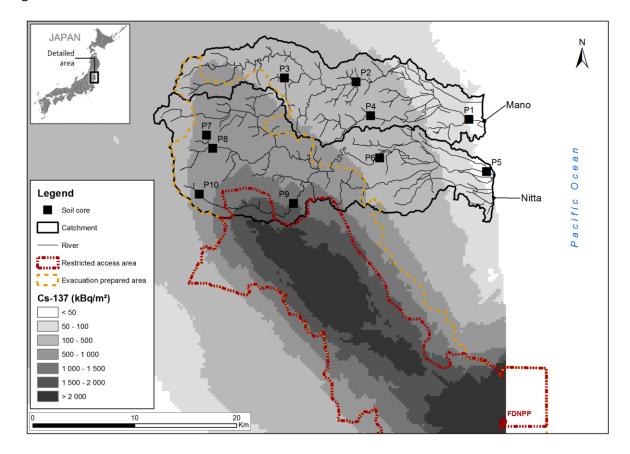


Figure2
Click here to download high resolution image

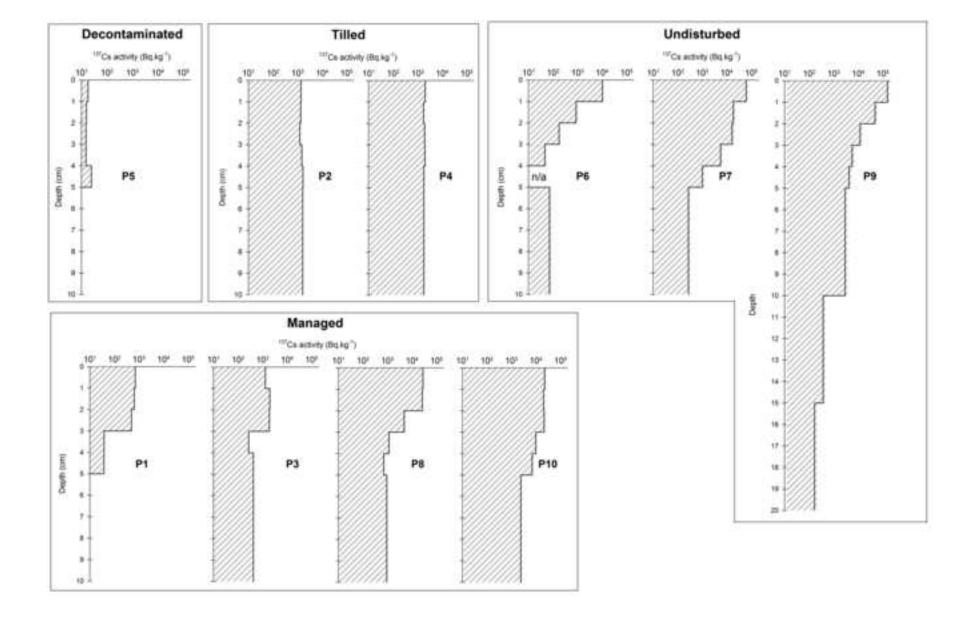


Figure3
Click here to download high resolution image

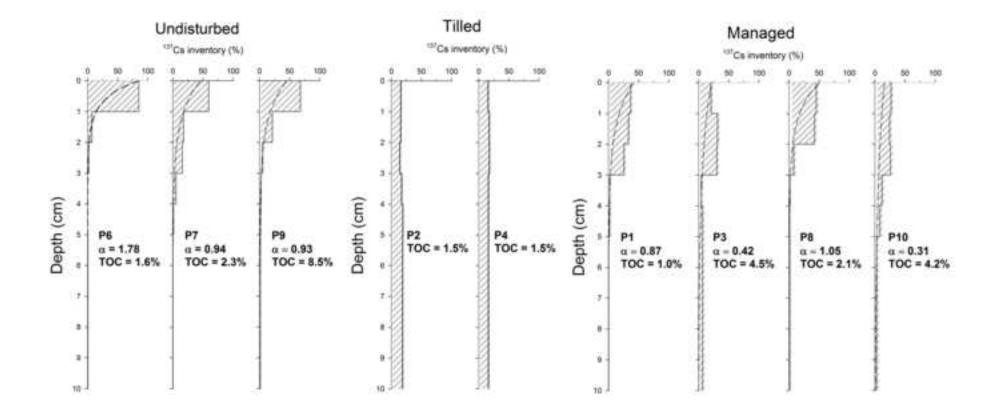


Figure 4.

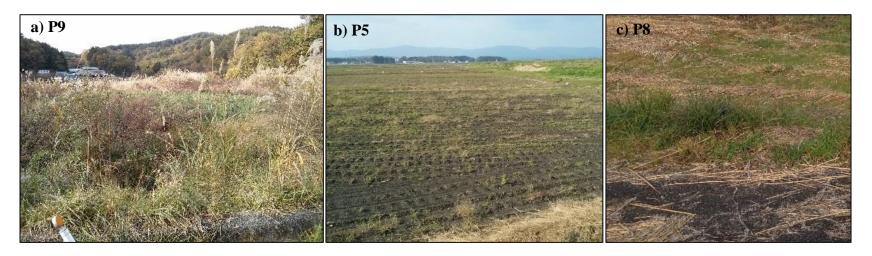


Figure5
Click here to download Figure: Figure 5.pdf

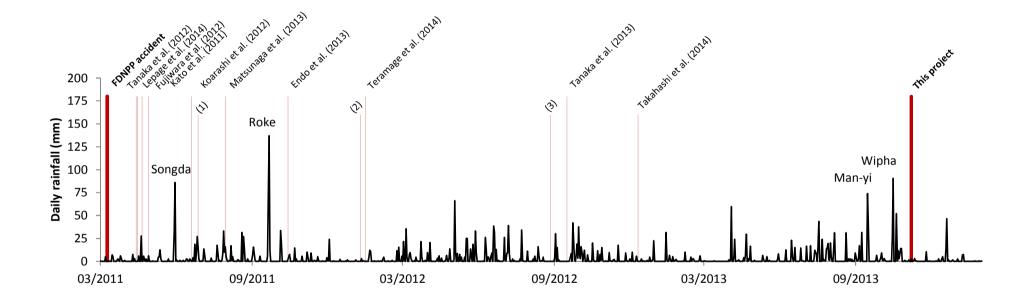


Table1 Click here to download Table: Table 1.docx

Table 1. Location of investigated soil cores and ambient radioactive dose rates measured at the ground level. Annual dose rates exceeding the permissible level of 1mSv.y⁻¹ are indicated in bold (MOE, 2012b).

Latitude	Longitude	Profile label	Dose rate (µSv.h ⁻¹)	Annual dose rate (mSv.y ⁻¹)	Japanese soil type (WRB)*
37.688264	140.995708	P1	0.2	0.8	Unknown
37.721432	140.870119	P2	0.4	1.9	Gray lowland soil (Gleyic Fluvisol)
37.724665	140.790469	P3	1.2	6.1	Gray lowland soil (Gleyic Fluvisol)
37.691504	140.886210	P4	0.5	2.4	Gray lowland soil (Gleyic Fluvisol)
37.642013	141.015405	P5	0.1	0.3	Unknown
37.654186	140.896448	P6	1.5	7.7	Gray lowland soil (Gleyic Fluvisol)
37.674029	140.703817	P7	2.7	14.0	Peat soil (Histosol)
37.662245	140.710906	P8	2.3	11.9	Gray lowland soil (Gleyic Fluvisol)
37.613850	140.800832	P9	5.5	28.7	Gray lowland soil (Gleyic Fluvisol)
37.621797	140.695852	P10	2.5	12.9	Gley soil (Gleysol)

^{*} Determined using National Institute for Agro-Environmental Sciences (NIAES) cropland soil map (NIAES, 1996; NIAES)

Table2

Click here to download Table: Table 2.docx

Table 2. Characteristics of the soil cores calculated for the uppermost 5 cm incremental layers. More detail is provided in the Supplementary material

Core	Class	¹³⁷ Cs inventory (%)	Bulk density (g.cm ⁻³)	h0 (kg.m ⁻²)	α (cm ⁻¹)	Mean TOC (%)	Clay (%) d < 10µm
P1	Managed	98.5	1.3 ± 0.3	20.4	0.87	1.0 ± 0.1	14.1 ± 1.1
P2	Tilled	46.8	1.2 ± 0.2	n/a	n/a	1.5 ± 0.2	8.2 ± 0.2
P3	Managed	73.6	0.8 ± 0.2	17.4	0.42	4.5 ± 0.2	10.6 ± 0.9
P4	Tilled	51.6	0.8 ± 0.2	n/a	n/a	1.5 ± 0.1	11.9 ± 2.3
P5	Decontaminated	72.9	1.2 ± 0.2	n/a	n/a	n/a	n/a
P6	Undisturbed	97.1	1.2 ± 0.1	6.3	1.78	1.6 ± 0.3	13.7 ± 2.3
P7	Undisturbed	98.7	0.9 ± 0.2	8.3	0.94	2.3 ± 0.2	10.8 ± 1.1
P8	Managed	92.9	1.1 ± 0.2	10.4	1.05	2.1 ± 0.4	8.3 ± 0.2
P9	Undisturbed	92.8	0.7 ± 0.1	5.4	0.93	8.5 ± 0.3	14.6 ± 1.2
P10	Managed	87.1	0.8 ± 0.1	16.8	0.31	4.2 ± 0.4	16.2 ± 0.6

n/a: not available

Table3
Click here to download Table: Table 3.docx

Table 4. Literature review of studies investigating evolution of radiocesium activities with depth in soils contaminated by FDNPP radioactive fallout.

Shape	Reference	Sampling period (mm/yy)	Number of samples	Type of land use investigated	Mean of % upper 5 cm	α (cm ⁻¹)	h ₀ (kg.m ⁻²)
	Endo et al. (2013)	10/11	3	Paddy field	≈50	n/a	n/a
	Koarashi et al. (2012)	06/11	6	Croplands (with one paddy field)	81	n/a	n/a
Disturbed (Tilled)	Matsunaga et al. (2013)	07/11	6	Croplands (with one paddy field)	80	n/a	n/a
	Tanaka et al. (2013)	09/12	3	Cultivated paddy field	55	n/a	n/a
	This study	11/13	2	Paddy field	49	n/a	n/a
		06/11	2	Land	99	0.3	n/a
	Takahashi et al. (2014)		3	Field	98	1.0	n/a
		01/12	2	Land	99	0.5	n/a
Disturbed			3	Field	98	0.6	n/a
(managed,		08/12	2	Land	100	0.7	n/a
grazing)			3	Field	94	0.5	n/a
		12/12	2	Land	98	0.4	n/a
			3	Field	88	0.6	n/a
	This study	11/13	4	Paddy field	88	0.7	18.2
		04/11	1	Brown forest soil	n/a	0.7	n/a
	Fujiwara et al. (2012)		1	Fluvisol	n/a	0.9	n/a
			1	Vegetable field	n/a	2	n/a
Undisturbed	Kato et al. (2011)	04/11	1	Cultivated soil (home garden)	99	1.2	9.1
Undisturbed	W 1 (2012)	06/11	6	Croplands (with one paddy field)	99	1.3	5.3
	Koarashi et al. (2012)		4	Grassland	99	1.2	4.9
			5	Forest	97	0.6	8.4
	Lepage et al. (2014)	04/11	1	Cropland	n/a	1,9	7.1

1 (2012)	05/44	6	Croplands (with one paddy field)	99	n/a	n/a
Matsunaga et al. (2013)	07/11	4	Grassland	99	n/a	n/a
		5	Forest	97	n/a	n/a
	06/11	3	Forest	98	0.7	n/a
	06/11	2	Land	100	2.0	n/a
	01/12	3	Forest	92	0.6	n/a
T.1.1.1.1.1.(2014)	01/12	2	Land	100	1.7	n/a
Takahashi et al. (2014)	08/12	3	Forest	96	0.7	n/a
		2	Land	100	1.4	n/a
	12/12	3	Forest	94	0.7	n/a
		2	Land	100	1.3	n/a
Tamalas et al. (2012)	04/11	2	Field	95	n/a	n/a
Tanaka et al. (2012)		2	Fruit trees field	92	n/a	n/a
Teramage et al. (2014)	01/12	1	Coniferous forest	92	0.6	11.1
This study	11/13	3	Paddy field	96	1.2	6.7

n/a : not available

Table4 Click here to download Table: Table 4.docx

Table 4. Correlation coefficients (r) obtained between the vertical distribution parameters and soil properties measured in samples collected in both undisturbed and managed fields

	α	h_0	TOC (%)	Clay (%)	TOC/Clay
α	1				
h0	-0.66	1			
TOC (%)	-0.35	-0.30	1		
Clay (%)	-0.11	0.13	0.30	1	
TOC/Clay	-0.38	-0.29	0.95*	-0.01	1

^{*} Significant at the 0.01 probability level