

# RAPID REMOVAL OF CESIUM-137 FROM URBAN AREA AFTER THE FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT ACCIDENT

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## Abstract

Urbanized areas have a large impact on external exposure to residents. Diverse decontamination methods had been evaluated by the model decontamination project carried out by Japan Atomic energy Agency in 2011 and 2012 to reduce the external exposure. On the other hand, quantitative information of distribution of radioactive substances, especially  $^{137}\text{Cs}$ , in urban area is limited, although the information is crucial for decontamination planning. To provide quantitative information of the distribution of  $^{137}\text{Cs}$  in urban area, this study evaluated relative  $^{137}\text{Cs}$  inventories, which are defined as the relative values of  $^{137}\text{Cs}$  inventory on each urban component such as roof and road to that on a nearby permeable plane field, for 11 buildings in the evacuation zone in 2015 and 2016. Additionally, the results were analyzed with the data obtained in the model decontamination project to evaluate temporal trend in the distribution during the initial five years after the accident. The relative inventory obtained for paved ground were 0.2 and for other building components such as roof and wall were less than 0.04 in 2016. These values indicate the limited contamination of urban area. The relative inventories of paved ground, roof and wall in 2011 and 2012 showed similar values with those obtained in this study, suggesting that initial run-off and the following wash-off effects during the first year after the accident largely defined the distribution of  $^{137}\text{Cs}$  in urban area. Some of the component showed rapid decrease in the relative inventory more than decay during five years after the accident. The fast decrease and the obviously low relative inventories on the components in urban area demonstrate the rapid removal of  $^{137}\text{Cs}$  from urban area.

## 1. Introduction

Large amounts of radionuclides were deposited on the ground over the North Kanto and the South Tohoku regions of Japan after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. The distributions, temporal changes and migrations of the radionuclides in diverse terrestrial environments have been well studied so far (Saito and Onda, 2015). On the other hand, those studies in urban area is crucially limited, although the area has large impact on exposure of local residents.

A decontamination pilot project had been carried out at residential area in the evacuation zone in 2011 and 2012 to establish guidelines of decontamination procedures (JAEA, 2015). The project provided many important information such as horizontal/vertical distributions of radionuclides in the area. Studies after the Chernobyl nuclear power plant accident evaluated radionuclides inventory (i.e. activity per unit area,  $\text{Bq m}^{-2}$ ) on urban components such as roofs, walls and roads, and their temporal changes (IAEA, 2006 and references in there). Their studies indicated rapid removal of the radionuclides from paved surface more than that from undisturbed field. The

different temporal decreases in the radionuclide inventory, especially  $^{137}\text{Cs}$  inventory, among the urban components largely affect to decontamination planning such as a decision of priority of urban surface to be decontaminated and an estimation of efficiency of the decontamination for dose reduction. In the case of the area affected by the FDNPP accident, relative  $^{137}\text{Cs}$  inventories, which are defined as the relative values of  $^{137}\text{Cs}$  inventory on each component to initial  $^{137}\text{Cs}$  inventory on a nearby permeable plane field, had been investigated for 11 buildings in the evacuation zone in 2015 (Yoshimura et al., 2017), but the temporal trend in radionuclides activity on urban surfaces have not been shown.

This study investigated the relative  $^{137}\text{Cs}$  inventories on urban components for the 11 buildings at evacuation zone in 2016. The objects buildings were selected to be same with those reported by Yoshimura et al. (2017). To clarify the temporal trends in relative  $^{137}\text{Cs}$  inventory on urban components, the data in 2015 (Yoshimura et al., 2017), in 2016 (this study) and obtained by the decontamination pilot project (JAEA, 2015) was analyzed together.

## 2. Methods

### 2-1. Investigation

This study evaluate the relative  $^{137}\text{Cs}$  inventories on urban components for the 11 buildings in Okuma and Tomioka towns, located in the evacuation zone, from 12 to 22 January 2016 (figure 1). The buildings and the adjacent fields had not been decontaminated. Therefore, decrease in  $^{137}\text{Cs}$  deposited on the studied fields has reflected only weathering effect (wash-off due to precipitation).

The surface contamination count rate (cpm) was measured for major components (roofs, walls and paved grounds at 212, 468 and 97 points, respectively) using a Geiger-Mueller survey meter (TGS-146, Hitachi-Aloka Medical, Japan) (table 1). Descriptions of the investigation has been reported by Yoshimura et al. (2015).

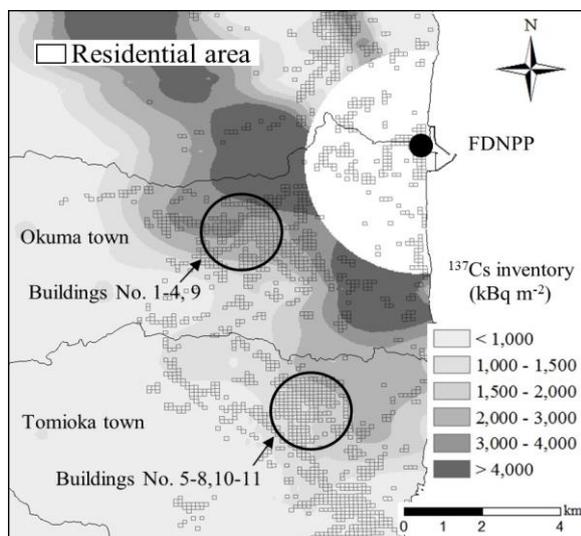


Figure 1 Study site (Yoshimura et al., 2017)

### 2-2. Evaluation of $^{137}\text{Cs}$ inventory

The  $^{137}\text{Cs}$  inventory ( $\text{Bq m}^{-2}$ ) on object component was calculated by multiplying the count rate by conversion factor, which is the ratio between  $^{137}\text{Cs}$  inventory and the count rate obtained in this study. To obtain the conversion factor,  $^{137}\text{Cs}$  inventory and the count rate were measured at 11 paved grounds (parking areas and roads) using a portable Ge gamma-ray spectrometer (Falcon-5000, CANBERRA, USA) equipped with a cylindrical collimator (30-mm-thick Pb, 60 mm height with 170 mm diameter) and a Geiger-Mueller survey meter (TGS-146, Hitachi-Aloka Medical, Japan), respectively, on 10 February 2016. The count rate was measured at 19 points in the area with 3 m in diameter below the portable gamma-ray spectrometer. Measurement description is mentioned in Yoshimura et al. (2017). Figure 2 shows the relation between the  $^{137}\text{Cs}$  activity per unit area and count rate, and a slope of 0.101 was obtained as the conversion factor.

This value is applicable in the case of that the  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio is constant. The results of in-situ

measurement using the portable Ge gamma-ray spectrometer showed no areal variation in the  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio in the studied area. However, the conversion factor have temporally increased, because the  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio decreases with time due to the different decay constant. Therefore, this study retrospectively calculated the conversion factors on the date of investigations in 2015 (conversion factor of 0.0776) and 2016 (conversion factor of 0.0993) by considering the physical decays of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ .

This study also estimated the  $^{137}\text{Cs}$  inventory on urban components from the count rate data obtained by the decontamination pilot project (JAEA, 2014<sup>a</sup>) using the Geiger-Mueller survey meter equipped with a collimator (table 2). The retrospectively estimated conversion factor was also applied to the calculation of the  $^{137}\text{Cs}$  inventory.

Table 1 Measured components and number of measurement points. W- and RC- indicate wood- or reinforced concrete-structures, respectively. The number from 1 to 3 in the “construction” column represents stories of the object building. Initial  $^{137}\text{Cs}$  inventory on permeable plane fields adjacent to the studied buildings are also shown as INV.

No.	Construction	Roof	Wall	Paved ground	INV. ( $\text{MBq m}^{-2}$ )
1	W-2	25	42	—	3.63
2	W-1	7	55	10	3.63
3	W-1	9	39	6	3.63
4	W-1	30	38	—	2.5
5	RC-1	—	46	1	2.67
6	W-1	34	33	7	3
7	W-1	32	30	—	2.07
8	RC-1	—	52	9	2.05
9	RC-2	32	47	18	3.7
10	RC-3	34	37	39	2.54
11	RC-3	9	49	7	2.2
Total		212	468	97	

### 2-3. Relative $^{137}\text{Cs}$ inventory

The relative  $^{137}\text{Cs}$  inventory, which is the ratio of  $^{137}\text{Cs}$  inventory of the component at the measured date to the initial one on permeable plane fields adjacent to each of the studied buildings, is useful to compare the  $^{137}\text{Cs}$  distribution on urban components among the area of which deposition amount is different. To calculate the relative  $^{137}\text{Cs}$  inventory for the data set obtained in this study and in Yoshimura et al. (2017), the  $^{137}\text{Cs}$  inventory on the adjacent permeable plane fields was measured using the portable Ge gamma-ray spectrometer on 4-16 February 2016.

The  $^{137}\text{Cs}$  deposition patterns due to FDNPP accident was largely determined until 23 March 2011

(UNSCEAR, 2013). Since the decrease rate of  $^{137}\text{Cs}$  inventory on permeable plane fields has been reported to be same with its physical decay constant (Mikami et al., 2015), the initial  $^{137}\text{Cs}$  inventory on the permeable plane fields at March 23, 2011 was estimated by only physical decay correction of the data obtained in this study. The estimated initial  $^{137}\text{Cs}$  inventories on the permeable plane fields are shown in Table 1.

For the data of decontamination pilot project, the  $^{137}\text{Cs}$  inventory on the permeable plane fields adjacent to the object component was estimated from the results of the Forth Airborne Monitoring Survey by MEXT (2011) as the averaged value within an area with radius 150 m from the object component. The results of Airborne Monitoring Survey is well correspondent with the inventory evaluated by soil sampling at 2,200 points on permeable plane fields. Since the Forth Airborne Monitoring Survey provides the data as the value on 7 November 2011, initial  $^{137}\text{Cs}$  inventory was estimated by physical decay correction.

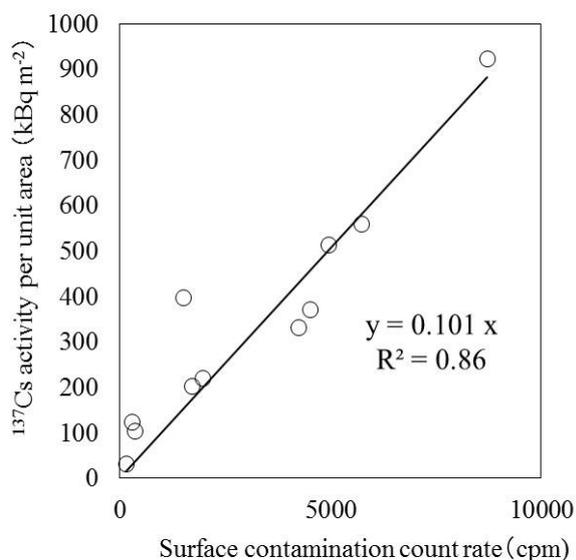


Figure 2 Relation between the  $^{137}\text{Cs}$  activity per unit area and count rate.

Table 2 Measured components and the data number referring in the decontamination pilot project (JAEA, 2014).

Site	Components			Measured date
	Roof	Wall	Paved ground	
Tomiooka	75	9	6	15 Jan.-3 Feb. 2012
Katsurao	—	—	12	18 Dec. 2011
Tamura	—	—	12	24 Dec. 2011
Okuma	20	2	22	30 Jan.-10 Apr. 2012
Kawauchi	5	2	—	9 Jan. 2012
Namie	—	1	—	6 Dec. 2011

### 3. Results and Discussions

#### 3-1. Relative $^{137}\text{Cs}$ inventories on urban components

The relative  $^{137}\text{Cs}$  inventory was summarized in table 3. The relative  $^{137}\text{Cs}$  inventory on the permeable plane fields is estimated values by physical decay correction of initial  $^{137}\text{Cs}$  inventory. The relative  $^{137}\text{Cs}$  inventory on paved ground was 0.18, accounting for 20% of the relative  $^{137}\text{Cs}$  inventory on the permeable plane fields, in 2016. Wall and roof showed obviously low values comparing to paved ground even when considering the standard deviation of the relative inventories. These low relative  $^{137}\text{Cs}$  inventories indicates that the contamination of urban area due to the  $^{137}\text{Cs}$  fallout was limited.

Table 3 Relative  $^{137}\text{Cs}$  inventories on urban components and their standard deviation (SD) estimated for 2011, 2015 and 2016.

Components	Dec.					
	2011-Apr.		Jan. 2015		Jan. 2016	
	Ave	SD	Ave	SD	Ave	SD
Permeable plane field	0.98	-	0.92	-	0.90	-
Paved ground	0.22	0.07	0.18	0.06	0.18	0.05
Wall	0.09	0.12	0.01	0.00	0.01	0.00
Roof	0.14	0.13	0.02	0.02	0.02	0.03

In the case of the FDNPP accident, wet deposition was the major process of  $^{137}\text{Cs}$  fallout on the ground (Morino et al., 2013). Therefore, the vertical structure of wall was inferred to be little-affected by the wet deposition, resulting in the low relative inventory. On the other hand, paved ground and roof are horizontal surface and directly affected by the wet deposition, but low relative inventory was also obtained. This was probably resulted by the low fixation capacity of  $^{137}\text{Cs}$  by the components due to their impermeability.

The relative  $^{137}\text{Cs}$  inventory on paved ground, wall and roof were 0.22, 0.09 and 0.14, respectively, in 2011-2012, and the values were obviously lower than that of permeable plane field even when considering the uncertainty due to the difference in investigation site and methods. This results suggest that the initial run-off and the following wash-off effects during the first year after the accident largely defined the distribution of  $^{137}\text{Cs}$  in urban area. The rapid removal of  $^{137}\text{Cs}$  from urban area during the initial period after the deposition is well consistent with the result of studies in Europe after the Chernobyl Nuclear Power Plant accident (Roed, 1987; Roed and Andersson, 2000<sup>a</sup>, 2000<sup>b</sup>).

Temporal change in the relative  $^{137}\text{Cs}$  inventories are described in figure 3. The proportions of decrease in relative  $^{137}\text{Cs}$  inventories on paved ground, wall and roof between 2011 and 2016 were 18%, 94% and

85%, respectively. These proportions were larger than that of permeable plane field (8%), of which decrease was represented by physical decay. This implies that the weathering effect one year after the accident also facilitate the removal of  $^{137}\text{Cs}$  from urban area. JAEA (2014) reported that the  $^{137}\text{Cs}$  removal rate, which was defined as the ratio between the  $^{137}\text{Cs}$  inventory in the catchment and discharged amount of  $^{137}\text{Cs}$  from the catchment, was less than several percentages during 3.5 years after the accident. Therefore, it is suggested that the urban area has large self-cleaning capacity more than that in other terrestrial environment.

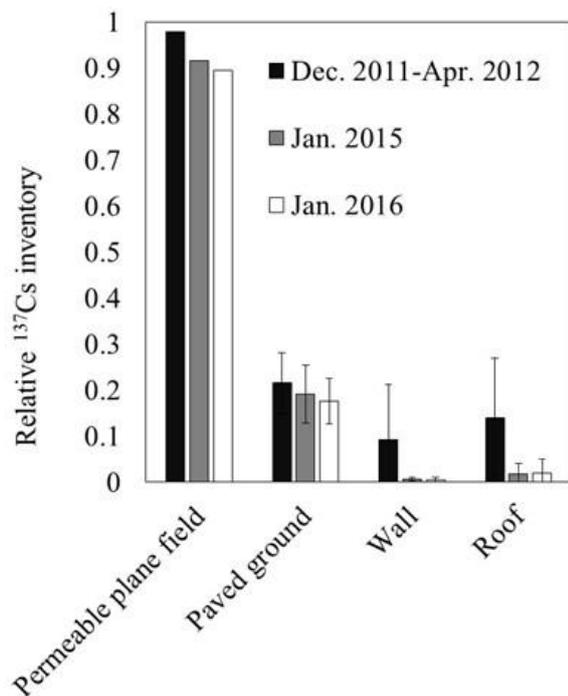


Figure 3. Comparison of relative  $^{137}\text{Cs}$  inventories among years and with that on permeable plane field.

### 3-2. Comparison of temporal trend of relative $^{137}\text{Cs}$ inventory on paved ground

The temporal trend of relative  $^{137}\text{Cs}$  inventory on paved ground obtained this study was compared with the results of studies in Europe after the Chernobyl Nuclear Power Plant accident (Andersson et al., 2002; Jacob and Meckbach, 2000) (figure 4). The relative  $^{137}\text{Cs}$  inventory estimated from the data of decontamination pilot project (about a year after the accident) fell in the range of the relative  $^{137}\text{Cs}$  inventory reported by the studies in Europe. This suggests that the initial behaviors (initial wash-off and weathering during the first year after the accident) of  $^{137}\text{Cs}$  deposited on paved ground was similar between this study and the case in Europe. On the other hand, the following decrease in the relative  $^{137}\text{Cs}$  inventory after 2012 was faster in Europe (decreasing rate of  $0.075\text{--}0.33\text{ yr}^{-1}$ ) than that of this study (decreasing rate of  $0.049\text{ yr}^{-1}$ ), suggesting small

weathering effect in this study more than that in the case in Europe. This difference in decreasing rate is probably caused by the human activities such as traffic and road cleaning. The studies in Europe was carried out in residential area, whereas the results in this study was obtained in the evacuation zone. Kinase et al. (2015) reported that the decrease in dose rate on roads in the evacuation zone is slower than that outside of the zone. Therefore,  $^{137}\text{Cs}$  on paved ground outside of evacuation zone probably removed rapidly more than the trend observed in this study, although further study is necessary.

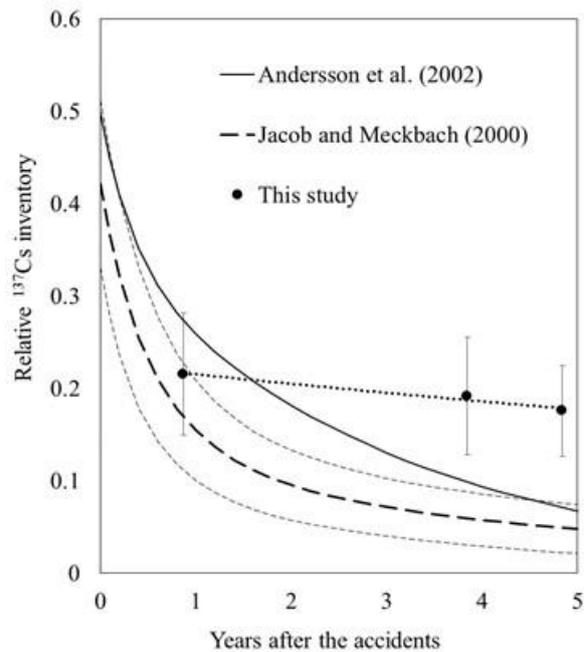


Figure 4 Temporal trend of relative  $^{137}\text{Cs}$  inventory on paved ground obtained in this study and observed in Europe after the Chernobyl Nuclear Power Plant accident.

### References

- Andersson, K.G., Roed, J., Fogh, C.L., 2002. Weathering of radiocesium contamination on urban streets, walls and roofs. *J. Environ. Radioact.* 62, 49-60.
- IAEA, 2006. Environmental Consequences of the Chernobyl Accident and Their Remediation: Twenty Years of Experience. Vienna, Austria, Reports of the Chernobyl Forum Experts Group 'Environment'.
- Jacob, P., Meckbach, R., 2000. Measurements after the Chernobyl Accident Regarding the Exposure of an Urban Population, pp. 34-41. In IAEA-TECDOC-1131.
- JAEA, 2014. Report for the Project Entrusted from the Nuclear Regulation Authority. <http://fukushima.jaea.go.jp/initiatives/cat03/pdf/07/Part1-6.pdf>. (in Japanese).

- JAEA, 2015. Remediation of contaminated areas in the aftermath of the accident at the Fukushima Daiichi nuclear power station: overview, analysis and lessons learned Part 1: a report on the “decontamination pilot project”. JAEA-Review 2014-2051.
- Kinase, S., Takahashi, T., Sato, S., Yamamoto, H., Saito, K., 2015. Prediction of Ambient Dose Equivalent Rates for the Next 30 Years After the Accident. Proceedings of International Symposium on Radiological Issues for Fukushima's Revitalized Future. 40-43.
- MEXT, 2011. Results of the Fourth Airborne Monitoring Survey by MEXT. [http://radioactivity.nsr.go.jp/en/contents/4000/3179/24/1270\\_1216.pdf](http://radioactivity.nsr.go.jp/en/contents/4000/3179/24/1270_1216.pdf).
- Mikami, S., et al., 2015. Spatial distributions of radionuclides deposited onto ground soil around the Fukushima Dai-ichi Nuclear Power Plant and their temporal change until December 2012. *J. Environ. Radioact.* 139, 320-343.
- Morino, Y., Ohara, T., Watanabe, M., Hayashi, S., Nishizawa, M., 2013. Episode analysis of deposition of radiocesium from the Fukushima Daiichi nuclear power plant accident. *Environ. Sci. Technol.* 47, 2314-2322.
- Roed, D.J., 1987. Run-off from and weathering of roof material following the Chernobyl accident. *Radiat. Prot. Dosim.* 21, 59-63.
- Roed, J., Andersson, K.G., 2000<sup>a</sup>. Development of a Strategy for Decontamination of an Urban Area, pp. 92-98. In IAEA-TECDOC-1131.
- Roed, J., Andersson, K.G., 2000<sup>b</sup>. Early Measurements in Scandinavia Following the Chernobyl Accident, pp. 71-75. IAEA-TECDOC -1131.
- Saito, K., Onda, Y., 2015. Outline of the national mapping projects implemented after the Fukushima accident. *J. Environ. Radioact.* 139, 240-249.
- UNSCEAR, 2013. Sources, Effects and Risks of Ionizing Radiation. Report to the General Assembly with Scientific Annexes, vol. I. Scientific Annex A.
- Yoshimura, K., Saito, K., Fujiwara, K., 2017. Distribution of <sup>137</sup>Cs on components in urban area four years after the Fukushima Dai-ichi Nuclear Power Plant accident. *J. Environ. Radioact.* 178-179, 48-54.