Field Survey of Radioactive Cesium Contamination in Concrete After the Fukushima-Daiichi Nuclear Power Station Accident

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Abstract

The Fukushima-Daiichi nuclear power station accident in East Japan in 2011 released radioactive cesium (r-Cs) into the surrounding environment. In addition to the major contamination of soil and vegetation, surrounding concrete structures have also been contaminated. Understanding the contamination characteristics is important for the decontamination or disposal of offsite and onsite concrete structures. However, r-Cs contamination of actual concrete has not been extensively investigated. In this paper, the r-Cs contamination characteristics of actual concrete are introduced on the basis of several field surveys and analysis of concrete samples taken from actual contaminated structures in the Fukushima prefecture. As no dissolution to water was detected, the surface contamination was safely removed by water-jet abrasion. However, in KCl solution, significant dissolution of r-Cs was detected. A contamination depth analysis was performed by quantitative r-Cs concentration mapping based on β-ray radiography using imaging plates. In many cases, such as roadside gutter cover plates, r-Cs was located on the surface without significant penetration into the concrete. However, in cases such as degraded concrete, deeper penetration up to several millimeters deep was observed. In cracked concrete, the r-Cs penetrated at least 10 cm, with the contamination level decreasing one order of magnitude in the first 1 cm.

1. Introduction

Interactions between concrete or cementitious materials and radioactive cesium (r-Cs) and other radionuclides have been primarily studied for the occurrence in concrete barriers or cement solidification in the final disposal of radioactive waste (Atkinson and Nickerson 1988; Holland and Lee 1992; Kienzler et al. 2000; Kaminski et al. 2016; PNNL 2014). However, research on the contamination of actual concrete structures is limited (Warming 1984; Farfán et al. 2011; Maeda et al. 2014; Koma et al. 2017).

The 2011 Fukushima Daiichi nuclear power station (NPS) accident released various radionuclides into the environment, contaminating not only soil, vegetation, and other media but also both offsite and onsite concrete structures. Due to the nature of the accident and its half-life, r-Cs is the most significant radiation source among the released nuclides. In the accident, radionuclides were mainly released as volatiles, and thus r-Cs is dominant especially offsite. Among the released volatiles, only r-Cs has a specific half-life of decay (2 and 30 years for Cs-134 and Cs-137, respectively) and is thus a significant radiation source.

The penetration depth of r-Cs in concrete significantly affects the waste volume. The movement and solubility changes of r-Cs resulting from the degradation of the concrete (Haga et al. 2019) are critical in determining appropriate decontamination and disposal methods. Thus, understanding the degree and state of r-Cs contamination in concrete is imperative for effective decontamination and final disposal activities.

Furthermore, to estimate the state of concrete contamination by r-Cs, it is necessary to understand the contamination mechanisms (Nagoya University 2018) and to perform analysis on actual concrete. For the analysis of actual contaminated concrete, the sampling of onsite concrete structures is quite difficult to perform during the busy decommissioning activities onsite. Even when samples were obtained, it is not allowable to analyze them in general laboratories without suitable specialized facilities for nuclear fuel treatment and the analysis process itself is time consuming. Therefore, in this study, r-Cs penetration in actual offsite concrete structures was analyzed.

Even for the sampling of offsite concrete, communication is required with local governments during the difficult times only one or two years after the severe accident and special permissions is required from the administrators of the concrete structures. Several field surveys of contaminated concrete around the Fukushima...
Concrete. Table 1 Important factors for r-Cs penetration into concrete.

<table>
<thead>
<tr>
<th>Nature of r-Cs</th>
<th>Concrete chemical characteristics</th>
<th>Physical effects</th>
</tr>
</thead>
<tbody>
<tr>
<td>Extremely low concentration of r-Cs as material</td>
<td>High K concentration in pore solution 0.1 to 1 ppm order of stable Cs Leaching of Ca by water Adsorption site change on C–S–H by carbonation</td>
<td>Cracks in concrete Water suction in dried concrete Clay particle stain on outdoor concrete</td>
</tr>
<tr>
<td>Strong interaction with clay minerals</td>
<td></td>
<td></td>
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<tr>
<td>Competitive adsorption with K</td>
<td></td>
<td></td>
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<tr>
<td>Solution or particle</td>
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Daiichi NPS were conducted. In this paper, the basic results of the surveys are described. For most of the samples, the radiation doses were measured by survey meters. For some samples, water abrasion was applied as a trial decontamination method and the dissolving behaviors of r-Cs were evaluated. Contamination depth was quantitatively evaluated by autoradiography using imaging plates (IP).

Almost no report of this kind with detailed information of concrete contamination has been published; thus, this paper is unique even without detailed discussion of contamination mechanisms. The obtained results are fundamental to gain insight and understanding of the current field contamination extent and characteristics, as required for further detailed studies.

2. Factors affecting r-Cs penetration into concrete

The possible important factors affecting phenomenon of Cs penetration into concrete are summarized in Table 1. First, the absolute physical amount of r-Cs released is extremely small: 1 g of Cs-137 releases 3.2 TBq of radiation. A radiation concentration of 100 kBq/kg is a relatively high level of offsite contamination; however, this is only 31 ng/kg (31 ppt) in terms of mass concentration, which is considerably lower than the detection limit of conventional chemical analytical techniques such as inductively coupled plasma mass spectroscopy (ICP-mass). The concentration of stable Cs in seawater is 310 ppt (JSAC 2004), whereas the r-Cs concentration in retention water in the damaged reactor immediately after the accident was estimated to be 4.4 GBq/L (3.5 ppm) (Nagoya University 2018). Such a low concentration of Cs may exhibit unforeseen characteristics. Second, Cs strongly interacts with clay minerals. Some parts of Fukushima are comprised of granite rocks along with vermiculite as a decomposed mineral from the original biotite included in granite. Third, potassium (K) behaves as a competitive adsorbate in the adsorption of Cs. Lastly, the chemical/physical form of r-Cs, that is, simple dissolved Cs ions or stable complex or particle Cs adsorbed or included (Adachi 2013), is a determining factor of r-Cs penetration in concrete.

Some chemical characteristics of the concrete also affect r-Cs penetration in concrete. In general, a K concentration in the pore solution of concrete on the order of 0.1 M reduces adsorption of Cs. In addition, approximately 0.1 to 1 ppm and 0.1 to 10 ppm of stable Cs are expected to be present in the concrete pore solution and soil or municipal solid waste incineration ashes, respectively (Kuramochi 2018 - personal communication on unpublished data in National Institute for Environmental Studies); these concentrations are significantly higher than the concentration of r-Cs released into the environment during the accident.

Furthermore, Cs adsorption by calcium silicate hydrate (C–S–H) in concrete is higher at lower Ca/Si ratios (Viallis et al. 1999). A low Ca/Si ratio is attained by the addition of pozzolan materials such as fly ash from coal-fired power stations, leaching of Ca via rainwater, or carbonation (Haga et al. 2019).

In terms of important physical characteristics that affect r-Cs concrete penetration, the concrete of large structures tends to crack due to dry shrinkage, leading to deep penetration of Cs ions. Drying of concrete is also important for water suction for rapid movement of contaminated water by rainfall (Sato et al. 2015). Furthermore, the staining of an outdoor concrete surface by clay dust promotes Cs adsorption.

3. Samples

Through several surveys, concrete samples were collected from various existing concrete structures in the Fukushima prefecture. Therefore, no information is available regarding the materials used, mix proportion, strength, etc. However, the samples were obtained from the usual site casted civil engineering works, buildings, or concrete products, and are not considered unique. A list of the samples obtained for this study is provided in Table 2.

The initial samples were obtained in December 2012 from relatively less contaminated road structures in the village of Iitate within a sampling area of 1 km. Six cores were drilled from the upper and side surfaces of the concrete walls as shown in Fig. 1. Three concrete cover plates of roadside gutters were sampled and replaced with new ones as shown in Fig. 2.

A second set of samples was obtained in November 2014. These included three cores from the concrete facilities of an incineration plant in the town of Namie, six cores from collapsed columns of a destroyed building by tsunami on the south side of the Fukushima Daiichi NPS, and eight cores from the town of Okuma from various concrete products for road-side gutter cover plates or construction blocks and porous tuff blocks called “Oya-ishi”.

The third set of samples was obtained from two cores taken in December 2015 from cracked portions of a
building surveyed in 2014. Before sampling, several studies were conducted to decide the sampling positions.

The cores were 4 or 10 cm in diameter and 5 to 10 cm in length. As the concrete had already been exposed to rainfall and the concentration of water-soluble r-Cs (if present) was estimated to be reduced, water was used as the cooling medium during coring.

4. Measurements

4.1 Radiation dose rate by survey meters

To estimate the r-Cs penetration into the concrete, radiation dose rates were measured at the sample surface by using two different survey meters: the Geiger-Muller (GM) tube and the NaI scintillator survey meter. The measurement values (dose rate) directly obtained from the survey meters were used in this study. The GM tube mainly counts β-rays and the NaI scintillator mainly counts γ-rays. β-rays can penetrate up to only 0.1 mm into the concrete (the stopping range, or the penetration limit, for the highest energy β-rays from Cs-137 is 0.2 mm); however, γ-rays can penetrate deeper, up to tens of centimeters into concrete (with 45-cm-thick concrete, γ-rays of Cs-137 decrease to 1/100 intensity). By using these characteristics of β-rays and γ-rays and by comparing the relative dose rate strengths, it is possible to understand the relative penetration depth of r-Cs. When r-Cs is located at the concrete surface, the dose rates obtained by the two methods are expected to be linearly proportional to the r-Cs concentrations. For greater r-Cs penetration, the dose rate measured by the NaI scintillator will be relatively higher than that as measured by the GM tube. Therefore, by comparing the measurement results from the two methods, it is possible to estimate whether r-Cs is located on the surface or has penetrated deeper.

For measurement of the radiation dose rate, it is important to consider the background level. When the measurement is carried out in contaminated environments, the effect of environmental radiation on the measurement accuracy of the GM tube can be eliminated to some extent by impeding β-rays with a thick lead shield for the tube because β-rays are easy to shield. However, this is difficult for the NaI scintillator because...
γ-rays are relatively difficult to shield efficiently. One may think it is possible to know the radiation of the measurement point by subtracting the background radiation level; however, it is quite difficult to determine the background level at the measurement location due to radiation from inhomogeneous contamination of surrounding substances such as soil and vegetation and special arrangement of radiation sources. Therefore, concrete sample evaluation using the NaI scintillator should be carried out in low background radiation environments, making it an effective method for cored surface contamination samples while it may be ineffective for use in a contaminated field. Further difficulties arise when using this method for a block-shaped concrete sample destroyed from some original structures by tsunami and its various surfaces contaminated afterwards, as the measurement results of one surface will be affected by the γ-rays from the other surfaces.

Depending on the purpose of measurements, some dose rate measurements were carried out onsite and others were carried out in the low background radiation environment at the National Institute for Environmental Studies in the city of Tsukuba.

4.2 Autoradiography measurements
For conducting autoradiography on the samples, imaging plates (IP; BAS-SR2040, GE Healthcare) were used to accumulate data, which were then retrieved by a laser scanner and fluorescence signal collector, Typhoon FLA 7000 (GE Healthcare), with a spatial resolution of 50 μm. Samples were cored from the field concrete and processed into plates having depth profiles and thickness of a few millimeters. The original surfaces of the concrete structure specimens were covered by a plastic bar to prevent scattering of β-rays from the surface perpendicular to the IP. While the thickness of the sample plate was limited, β-ray scattering from the surface made identification of the position of the surface with respect to the IP difficult. In the case of cracked concrete, the surface of the core sample was first covered with resin containing iron powder to prevent the scattering of β-rays from the concrete surface and then the sample was cross-sectioned without coolant by a wire saw into two pieces perpendicular to the crack.

Following the method of Yamada et al. (2018), the Cs-137 concentration of each pixel was obtained via the measurement signal was performed by standard samples prepared separately containing known concentrations of Cs-137. The size of the original pixel was 50 μm × 50 μm and for the quantification, from 10 × 10 pixels, one pixel was averaged to be 0.5 mm × 0.5 mm and used for the concentration mapping. In this calculation, r-Cs is assumed to contain only Cs-137 although a minor amount of Cs-134 is included in the contaminated concrete because the initial concentrations of Cs-134 and Cs-137 were similar in the reactor (Nagoya University 2018) and the half-life of Cs-137 is 30 years and that of Cs-134 is 2 years.

4.3 Radiation concentration measurement
The radiation concentration of r-Cs was measured by a Ge-type detector (ORTEC GMX-15190) for Cs-137 and Cs-134. The total concentration at the time of measurement without decay calibration was used in this study. For concrete, samples were crushed to particles with a size less than 1.25 mm in a homogeneous condition and placed in a 100 cm³ plastic vessel (U-8 vessel). For solution, samples were directly placed in a U-8 vessel.

4.4 Water-jet abrasion
Abrasion by waterjet (WJ) shown in Fig. 3 was used to gradually remove concrete from the sample surface (Yamada et al. 2013). Prior to WJ abrasion, the concrete surface was cleaned by high-speed application of high-pressure water at 3 MPa. Then, WJ was applied at 150 MPa and the surface of concrete was swept by WJ for several seconds. The sample mass reduction was converted to the average abrasion depth by WJ.

4.5 Water-soluble content
The concrete sample was crushed to a size of less than 2 mm, and 50 g of this powder was mixed with 500 g of pure water or a 1 M potassium chloride (KCl) solution. This mixture was shaken at 200 rpm with a 4 cm amplitude for 24 hours or 7 days. Then, the solution was filtered, and the radiation concentration was measured by a Ge-type detector.

5. Survey Results

5.1 Evaluation by survey meters
All cores and gutter cover plates were evaluated by survey meters in low-background radiation environments; the measurement results are shown in Fig. 4. The γ-ray dose rate has a lower limit than the β-ray dose rate, as shown by the line in Fig. 4, which can be considered as representative of the surface-level contamination as explained in Section 4.1. The porous tuff stone block and concrete block are significantly above the trend line, which indicate considerably deeper r-Cs penetration. Several concrete samples from collapsed columns and road concrete from the village of Iitate show deeper r-Cs penetration.

The radiation concentrations of concrete samples from Iitate were measured by a Ge-type detector. The relat...
tionship between r-Cs concentration and surface (β-ray) dose rate obtained by the GM tube is shown in Fig. 5. A linear correlation, except for one data point, is observed, indicating that the contamination is primarily limited to the surface as only surface-level β-rays can be detected by the GM tube. One exception indicates that r-Cs penetrated deeper in one sample than the other samples. A detailed explanation for this exception is provided in the following sections.

5.2 Water-jet abrasion decontamination profile
An example of decontamination trial by WJ abrasion for the three concrete gutter cover plates obtained from the village of Iitate is introduced on the basis of Yamada et al. (2013). Figure 6 shows the appearance of the samples before WJ abrasion. High-pressure water cleaning prior to WJ abrasion reduced 30% to 40% of the dose rate, indicating that significant amounts of r-Cs could have been loosely adsorbed as dust such as clay. After this cleaning, WJ was applied, with the results shown in Fig. 7. Through WJ abrasion, the surface dose rate decreased, indicating the contamination profiles. The contamination was limited to depths of less than 1 mm, although the depth depends on the sample type. Clearly, the order of surface degradation corresponds to the contamination depth. The maximum contamination depth was observed for sample I-6, which had the roughest surface, with the cement paste peeled off and coarse aggregate exposed. This case corresponds to the deep r-Cs penetration data point indicated by a red square in Fig. 5. This is due to possible deeper penetration of r-Cs in degraded concrete as compared to sound concrete.

WJ abrasion generates concrete sludge. The solid parts were eliminated by flocculation with poly-aluminum chloride. The radiation dose rate of the separated water was at the level of the background radiation dose rate. This suggests that WJ is a suitable decontamination method not generating contaminated waste water.

5.3 Dissolution behaviors of r-Cs from contaminated concrete
From the water after WJ application, r-Cs was not detected. However, the in-field measurement detection limit is relatively high and there is a possibility of a minor amount of dissolution of r-Cs. Therefore, in-laboratory dissolution experiments were conducted on the same samples to investigate the dissolution behaviors of r-Cs...
of r-Cs from the concrete.

The original concrete had an r-Cs concentration of 6930 Bq/kg. The water filtered out from the crushed concrete after the dissolution test had an r-Cs concentration of less than 2 Bq/L. No dissolution of r-Cs was detected by water from the concrete. In the case of the KCl solution, 34.3% of r-Cs leached out. With the extension of the shaking time to 7 days, the leached amount increased to 42.4%. After the 7 days shaking period, the solution was replaced with a new 1 M KCl solution, and the mixture was shaken for 1 day; this process was conducted three times in total. The leaching ratio increased from 42.4% to 45.8%, 46.9%, and 47.6% with repeated dissolution processes.

The r-Cs in concrete obtained in this study is water insoluble. Evidently, the surfaces of the concrete samples used in this study have been exposed to rainfall and the soluble r-Cs has already washed out. However, it can be partially replaced by concentrated K ions. In general, cement contains approximately 0.1 to 1 ppm stable Cs and 0.1% K. If these alkalis remain in the concrete, r-Cs coexists in the concrete at a high concentration on the order of less than 2 Bq/L. No dissolution of r-Cs was detected by water from the concrete. In the case of the KCl solution, 34.3% of r-Cs leached out. With the extension of the shaking time to 7 days, the leached amount increased to 42.4%. After the 7 days shaking period, the solution was replaced with a new 1 M KCl solution, and the mixture was shaken for 1 day; this process was conducted three times in total. The leaching ratio increased from 42.4% to 45.8%, 46.9%, and 47.6% with repeated dissolution processes.

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However, the measurement results are inconsistent with these characteristics. During their exposure to the environment over their lifetime (tens of years), the original alkalis from the surface of the road concrete might have been washed out by rainfall, and an extremely small amount of r-Cs was supplied by rainfall after the accident. This r-Cs might be strongly adsorbed by C–S–H with a low C/S ratio after Ca dissolution by rainfall, carbonation products after C–S–H, or clay minerals originally contained in the aggregates. At this point in time, the contribution to adsorption by the clay is not clear. However, the adsorption of r-Cs is strong in water with gradual exchange with K ions occurring at a high concentration (1 M). Cs dissolution from the cement hydrate is rapid, and this dissolution behavior suggests that the clay primarily adsorbs the Cs. Additionally, looking at the K concentration in the concrete, aggregate has an ability to absorb alkalis and after the occurrence of the alkali-silica reaction, additional alkalis are absorbed, thus resulting in a significantly lower K concentration than expected in simple cement paste. In general, K concentration in the pore solution of concrete is expected to be on the order of 0.1 M; however, after some interaction between concrete components and alkalis, the actual K concentration can be significantly lowered. Pore solution analysis is required for further elucidation.

5.4 Basic r-Cs penetration profiles by radiography

The basic r-Cs contamination characteristics will be first introduced, and then the detailed distribution will be presented. The contamination characteristics of concrete wall and gutter cover samples taken from the village of Iitate are summarized in Table 3 and their penetration behaviors are shown in Fig. 8 as radiographs. For the concrete walls, as compared to the side surfaces, the upper surfaces showed higher contamination and less neutralization depths. The r-Cs released from the reactor is believed to have been transported as atmospheric plumes and thus may have leached into the ground surface through rainfall. This matches with the observation that the reverse sides of the concrete gutter cover plates were not contaminated. There are no clear correlations between the neutralization depth and the contamination level.

Figure 8 shows that the penetration depths were limited in most cases. The cover plate sample I-6, from the village of Iitate (Fig. 2), was found to have deeper r-Cs penetration (Fig. 2). Figure 9 shows an enlarged image of this sample. As shown in Fig. 9, the surface of I-6 was relatively rough and r-Cs penetration into the concrete occurred in the region of neutralization. While it is difficult to quantitatively specify, the penetration depth seems to be a few millimeters.

Hereafter, the r-Cs distribution is described in more detail with quantitative mapping images. Typical examples of r-Cs contamination in the town of Okuma are shown in Fig. 10. Figures 10 (a), (b), and (c) show examples of r-Cs penetration on the order of several millimeters. In the left column, the top image is the cut surface of the concrete, the bottom image is the r-Cs distribution found by autoradiograph, and the middle image is the r-Cs distribution overlain on the cut surface. The right side of the figure presents 3D images of the r-Cs concentration distribution maps; in these maps, the z-axis corresponds to the r-Cs concentration on a linear
Table 3 Contamination characteristics of samples from the village of Iitate. Cores 1, 2 and 3 are from concrete walls. “u” and “s” represent cores from upper and side surfaces, respectively. Cores 4, 5 and 6 are from concrete gutter cover plates.

<table>
<thead>
<tr>
<th>Core</th>
<th>r-Cs conc. Bq/cm²/Ge</th>
<th>Surface dose ratio kcpm/GM</th>
<th>Neutralization depth μSv/h/NaI mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>I-1u</td>
<td>14.0</td>
<td>3.7</td>
<td>0.41</td>
</tr>
<tr>
<td>I-1s</td>
<td>5.0</td>
<td>1.5</td>
<td>0.25</td>
</tr>
<tr>
<td>I-2u</td>
<td>34.4</td>
<td>10</td>
<td>0.</td>
</tr>
<tr>
<td>I-2s</td>
<td>0.2</td>
<td>0.3</td>
<td>0.21</td>
</tr>
<tr>
<td>I-3u</td>
<td>36.4</td>
<td>10</td>
<td>0.72</td>
</tr>
<tr>
<td>I-3s</td>
<td>5.9</td>
<td>3</td>
<td>0.33</td>
</tr>
<tr>
<td>I-4</td>
<td>21.4</td>
<td>7</td>
<td>0.7</td>
</tr>
<tr>
<td>I-5</td>
<td>14.6</td>
<td>5</td>
<td>0.71</td>
</tr>
<tr>
<td>I-6</td>
<td>33.3</td>
<td>6</td>
<td>1.05</td>
</tr>
</tbody>
</table>

For sample O-3 [Fig. 10 (a)], the majority of the r-Cs remained at the surface; however, along a small crack of several millimeters, r-Cs penetration can be observed. For sample O-6 and O-8 [Figs. 10 (b) and (c)], the r-Cs penetrated through the cement paste while avoiding the aggregate, because the aggregate parts remained black, thus suggesting no presence of r-Cs. Even in these cases, the r-Cs concentration on the surface of the concrete is not homogeneous and it exhibits spots of high concentration. The reason for the appearance of these hot spots is not clear although it may be attributed to the presence of clay minerals.

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![Fig. 9 Enlarged images of the I-6 sample (2 cm × 5 cm) of a road gutter cover (with a relatively degraded surface) from the village of Iitate for determining the neutralization depth (left, no purple color region), and the corresponding radiographs (right).](image)

![Fig. 8 Concrete plate samples from the village of Iitate with plastic bars (a cross section of 2 cm²) for shielding (upper) and their radiographs (lower). The upper rims are the concrete surfaces exposed to the environment. Brighter colors indicate denser contamination.](image)
(a) Sample O-3 of a concrete column from the town of Okuma. A crack of several millimeters depth can be observed.

(b) Sample O-6 of a concrete column from Okuma. R-Cs penetrated through the cement layers to a maximum depth of 10 mm, avoiding the aggregate.

(c) Sample O-8 of a concrete column from Okuma. Several millimeters of penetration of r-Cs is observed.

(d) Sample O-9 of a road-side gutter cover (not degraded) from Okuma. There is no significant penetration of r-Cs.

Fig. 10 Examples of r-Cs contamination profiles.

*Left upper:* Cross section of measured samples.  
*Left middle and lower:* Radiographs.  
*Right:* Quantitative mapping of Cs-137. x- and y-axes are the positions in mm, whereas the z-axis is the Cs-137 concentration (Bq/kg) on a linear scale. The legend is the Cs-137 concentration (Bq/kg) on a logarithmic scale.
The r-Cs concentration level is expected to depend on the sample location in terms of the environmental contamination level. Sample O-9 from the town of Okuma was collected from the most highly contaminated region, with its peak concentration approximately 10 MBq/kg as shown in Fig. 10 (d), which is one order of magnitude higher than that of other samples. However, the contamination depth of r-Cs for this sample is limited. Beyond several millimeters, a green region is still observed, but the concentration level is three orders of magnitude lower; this region may be attributed to the effects of secondary X-rays from the high concentration of r-Cs on the surface.

On the contrary, in the case of a porous tuff block, r-Cs penetration to up to 10 mm is observed as shown in Fig. 11. This deeper penetration suggests that the water suction of porous materials is an important factor in determining the contamination depth. Prior to the r-Cs plume from the reactor, if the concrete was pre-wetted by the tsunami or non-contaminated rainfall, the contamination depth may be limited. However, if the block was dry, significant water suction is expected and deeper penetration would be expected. Even in this process of rapid water suction, the adsorption reaction of r-Cs by clay seems comparable because the concentration gradient is observed.

5.5 Mechanism of r-Cs penetration

Based on data supplied by the Japan Meteorological Agency, before the day of contamination, from March 8th to the morning of March 14th, 2011, there was no rain in the region, the sky was clear, and a western wind was dominant. Therefore, the concrete was expected to be dry. The 2011 Tohoku earthquake occurred on March 11th, with the leakage of r-Cs starting on March 12th. The atmospheric movement of r-Cs has been simulated (Kitaayama et al. 2018). Just after the accident, the majority of r-Cs drained to the Pacific Ocean due to the western wind. As a result of the earthquake and the following NPS accident, data from weather stations near the Fukushima Daiichi NPS is not available for the time period immediately after the earthquake. According to the data at Onahama, which is 60 km south of the Fukushima Daiichi NPS and similarly faces the Pacific Ocean, from the afternoon of March 14th, the wind direction changed over time. Judging from the data of nearby sites, from late afternoon of March 15th to the morning of March 16th, the area under consideration received rainfall. Before rainfall, the r-Cs plume seems to have been spread in that region. After this first contamination, from March 17th to 19th, the western wind became strong with no rainfall and a clear sky observed. On March 20th, the direction of the wind changed again, and rain recommenced, continuing until March 23rd. During this period, a second contamination occurred.

Water suction is expected to occur much faster than diffusion. It is not clear whether r-Cs trapped from rain by some minerals such as clay is faster than by water suction. In the case of concrete composed of pure materials that cannot adsorb Cs, r-Cs may have penetrated further, passing through the cement paste (Nagoya University 2018). From the r-Cs contamination profiles, the r-Cs was quickly trapped at the surface of the concrete and did not penetrate further for several years, even by further weathering and rainfall. This behavior is different from cement paste but similar to clay. For this trapping, water may be required to have been retained for a longer period of time by impermeable concrete. In contrast, if the concrete is porous and relatively permeable and contains clay, water would move quickly enough without adsorption by clay as can be estimated from Fig. 11. These are, nevertheless, only suppositions and require direct evidence and detailed experiments, as planned in a forthcoming project (Maruyama 2018; Nagoya University 2018).

![Fig. 11 Core from a tuff porous block (ø = 4 cm, left hand image is the surface). r-Cs penetration to 10 or 20 mm is observed.](image)

*Left:* Cross section of the measured sample.
*Middle:* Radiograph.
*Right:* Quantitative mapping of Cs-137. Figure details are the same as for Fig. 7.
5.6 R-Cs distribution on the surface of the concrete columns and effects of cracks

It is well known that ions in solution penetrate faster along cracks in concrete elements than in a sound matrix. R-Cs is expected to behave in the same way. In general, drying shrinkage of concrete leads to crack formation. The small crack-like feature shown in Fig. 10 (a) is an example of this type. After the 2011 Tohoku earthquake, cracks formed on many concrete structures due to mechanical deformation. One of the concrete samples taken for this study was cored at the position of a crack in a concrete structure destroyed by the 2011 tsunami.

Figure 12 shows the appearance of the column (left) and the concrete surface near the position of the sample taken (middle). The cracks seem to have been generated by bending stress during the accident as the cracks open from the right-hand side and propagate to the left-hand side, as shown in Fig. 12 (left). Figure 12 (right) shows the correlation between the radiation dose rate of γ-rays measured by the NaI scintillator and β-rays measured by the GM tube, as measured on-site with a lead shielding sleeve. At the position of the cracks, the β-ray dose rate is stronger than where there are no cracks and the γ-ray dose rate is stronger as compared to the β-ray rate, which indicates that a greater amount of R-Cs is contained both on the surface at the cracks and in deeper positions of the concrete.

Figure 13 (upper left) shows a collapsed column. The radiation distribution on the surface of the column was evaluated at an interval of approximately 5 cm as shown by green tape patches in Fig. 13 (upper right). In Fig. 13 (lower), the β-ray radiation dose rate measured by the GM tube in the field using a lead shielding sleeve is indicated with the major cracks. The crack width is limited as compared to that shown in Fig. 12, and the cracking mechanism is not clear. The cracks are suspected to result from the effects of both drying shrinkage and bending stress by tsunami. The contour of dose rate (cpm) was reproduced on the basis of the measurement data. As clearly shown, the radiation dose rate is higher along the cracks. At this scale, the R-Cs concentration seems relatively homogeneous yet still affected by cracks. Some R-Cs can be estimated to have penetrated the concrete and be concentrated at deeper positions in the cracks.

Figure 14 shows the correlation between radiation dose rates measured by the NaI scintillator and the GM tube for the column in Fig. 13. Clearly, there is one outlying data point, indicated as a small red dot in Fig. 13 (lower), which has a more than 10 times higher dose rate as measured by the GM tube or a 5 times higher dose rate as measured by the NaI scintillator compared with the other data points. A detailed observation at the site revealed that the radiation was generated from a 1 mm hole; however, the exact origin of the strong radiation could not be detected. Retrieving high-dose-rate materials such as those from the site and testing them in our facilities is prohibited. The existence of a tiny nanometer size “Cs ball” composed of fused silicate at extremely high dose rates in reactors has been reported (Satou et al. 2018). There may be such a substance in a small hole of entrapped air on the surface of the concrete column. The radiation dose contour shown in Fig. 13 (lower) was obtained by eliminating this high dose rate point as well as another high dose rate point near this point.

Figure 15 shows the correlation between radiation β- and γ-ray dose rates measured by the NaI scintillator and GM tube, respectively, after eliminating the two high dose rate data points. The results from the cracked positions showed a relatively higher dose rate as measured by the NaI scintillator than those as measured by the GM tube. As indicated in Fig. 12 (right), R-Cs is expected to have penetrated deeper into the concrete along the crack.

To verify how R-Cs penetrated along the crack, a concrete core (OC-1) was drilled at the crack position, indicated as a purple circle in Fig. 13 (lower). Figure 16 shows the appearance of the concrete core OC-1 and the R-Cs distribution on the surface of the core as detected by IP. Although a crack was included, the core was successfully extracted without obvious damage. The radia-
tion dose rate along the crack is higher than that at other positions, as expected from the large-scale evaluation shown in Fig. 13. The r-Cs distribution is not homogeneous even at areas without cracks, which indicates that some material may have preferentially adsorbed r-Cs.

Figure 17 shows processing of a concrete core sample taken from a collapsed column shown in Fig. 13. Figure 18 shows the appearance of the cross section of the core OC-1 (left), with the crack emphasized with a red line, the radiograph showing the penetration profile (middle).

Fig. 13 Radiation distribution on the surface of a collapsed column from a concrete structure destroyed by the 2011 tsunami from a region south of the Fukushima Daiichi NPS. The legend indicates the dose rate as obtained by the GM tube (cpm).

Upper left: Appearance of the structure.
Upper right: Appearance of the collapsed column.
Lower: Crack distribution and radiation contours based on the β-ray dose rate measured by the GM tube. Green rectangular tape patches indicate the measurement locations. The purple circle indicates the core sampling location. The red point indicates the position of a “hot spot.”

Fig. 14 Correlation between radiation dose rates as measured by the NaI scintillator and GM tube for the column shown in Fig. 10.

Fig. 15 Correlation between radiation dose rates by the NaI scintillator and GM tube for cracked and non-cracked locations for the column shown in Fig. 10.
and the quantitative r-Cs distribution along the crack (right). The crack passes through the core from its top to the bottom. Along the crack, r-Cs reaches to the bottom of the sample. However, the concentration gradient is extremely steep. The peak concentration is approximately 3.5 MBq/kg at a few millimeters below the surface. The concentration decreases to 100 kBq/kg at a depth of 10 mm depth, 10 kBq/kg at depth of 50 mm depth, and 1 kBq/kg at the end of the crack at a depth of 70 mm.

A core extracted from the damaged column (OC-2) shown in Fig. 12 is shown in Fig. 19. In this case, the peak concentration is 1 MBq/kg at a depth of 10 mm and spreads in a several millimeters range due to the appearance of several cracks. Then, the concentration decreases to 100 kBq/kg at a depth of 20 mm and 10 kBq/kg at a depth of 30 mm. At a position of 30 mm to the end of crack at 70 mm, the concentration gradually decreases from 10 kBq/kg to 5 kBq/kg. The decreasing trend of the r-Cs concentration in this case differs from that shown in Fig. 18. In the case of heavily damaged samples, contamination reaches relatively deeper with a greater extent and the r-Cs penetrates deeper. Besides, depending on the direction of the surface to the ground, the time in contact with rainwater may differ, which may provide further differences in the extent of contamination.
6 Conclusions

In this study, basic information relating to the characteristics of r-Cs penetration into concrete was obtained by collecting concrete samples from several actual structures in the Fukushima prefecture and analyzing them by several methods.

1. The penetration depth of r-Cs was estimated by simple measurements using the GM tube and NaI scintillator survey meters. In many cases, r-Cs remained at the surface; however, deeper penetrations were estimated for several samples.
2. Water jet could be applied as a decontamination tool as no r-Cs dissolution was detected in the used water.
3. From the contaminated concrete samples in the field, r-Cs was found to be water insoluble, yet in KCl solution (1 M), approximately half of the r-Cs was soluble.
4. No direct effect of neutralization on r-Cs penetration was observed.
5. By quantitative mapping of r-Cs based on autoradiography using imaging plates, the distribution characteristics of r-Cs in the concrete samples were discussed.
6. The majority of r-Cs remained near the concrete surface.
7. Depending on the degradation level of the concrete, r-Cs penetration of a few millimeters was observed in some samples. In these cases, the r-Cs penetrated into the concrete through the cement paste.
8. The r-Cs distribution on the concrete surface was not homogeneous, with hot spots, which could be attributed to the presence of strong adsorbing materials for Cs such as clay minerals.
9. Higher dose rates of r-Cs were detected along cracks in the concrete.
10. From the analysis of the cross section of the cored samples, r-Cs penetration along cracks was more than 10 cm.
11. The r-Cs distribution differed depending on the nature of the crack. Larger cracks resulted in deeper penetration and higher contamination. However, the majority of the r-Cs penetrated only a few centimeters.

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References