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# Removal of radioactive iodine and cesium in water purification processes after an explosion at a nuclear power plant due to the Great East Japan Earthquake

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## ABSTRACT

The presence of radionuclides at five water purification plants was investigated after an explosion at a nuclear power plant hit by the Great East Japan Earthquake on 11 March 2011. Radioactive iodine ( $^{131}\text{I}$ ) and cesium ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) were detected in raw water in Fukushima and neighboring prefectures.  $^{131}\text{I}$  was not removed by coagulation–flocculation–sedimentation.  $^{131}\text{I}$  was removed by granular activated carbon (GAC) and powdered activated carbon (PAC) at a level of about 30%–40%, although  $^{131}\text{I}$  was not removed in some cases. This was also confirmed by laboratory-scale experiments using PAC. The removal percentages of  $^{131}\text{I}$  in river and pond waters by 25 mg dry/L of PAC increased from 36% to 59% and from 41% to 48%, respectively, with chlorine dosing before PAC.  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were effectively removed by coagulation at both a water purification plant and in laboratory-scale experiments when turbidity was relatively high. In contrast,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in pond water with low turbidity were not removed by coagulation. This was because  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in river water were present mainly in particulate form, while in pond water they were present mainly as cesium ions ( $^{134}\text{Cs}^+$  and  $^{137}\text{Cs}^+$ ). However, the removal of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in pond water by coagulation increased markedly when  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were mixed with sediment 24 h before coagulation.

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## 1. Introduction

The Great East Japan Earthquake occurred on 11 March 2011, and caused an explosion at the Tokyo Electric Power Company (TEPCO) Fukushima Daiichi Nuclear Power Plant, which resulted in the release of large amounts of radionuclides into the environment (Water Supply Division (WSD), Health Service Bureau (HSB), Ministry of Health, Labour, and Welfare (MHLW), 2011). Radioactive iodine ( $^{131}\text{I}$ ) and cesium ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) were detected in drinking water after the explosion (WSD, HSB, MHLW, 2011; Ikemoto and Magara, 2011).

Several groups have investigated the occurrence of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  at water purification plants (Goossens et al., 1989; Esumi et al., 1986; Gäfvert et al., 2002; Kamata et al., 1973) although data after nuclear accidents were limited. It was considered that radionuclides existed in particulate and dissolved forms in environmental water considering their forms in the air released after nuclear power plant accidents (Noguchi and Murata, 1988). They may also exist in colloidal form. Dissolved  $^{131}\text{I}$  is separated into various forms, such as radioactive iodide ion ( $^{131}\text{I}^-$ ), iodine molecule ( $^{131}\text{I}_2$ ), hypoiodous acid ( $\text{HO}^{131}\text{I}$ ), iodate ion ( $^{131}\text{IO}_3^-$ ), and organo- $^{131}\text{I}$ . The

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proportions of the forms of  $^{131}\text{I}$  were considered to be different among environmental waters and to change due to reactions in the environmental water. For example,  $^{131}\text{I}^-$  slowly transforms into  $^{131}\text{IO}_3^-$  in environmental water under the action of sunlight (Lettinga, 1972).  $^{131}\text{I}_2$  and  $\text{HO}^{131}\text{I}$  react with natural organic matter (NOM) in the environmental water. Thus, it was assumed that  $^{131}\text{I}$  removal at water purification plants would depend on the environmental water. Information regarding the occurrence of radionuclides at water purification plants is valuable. However, their levels in the environmental water are usually very low, and thus such data are limited.

There have also been experimental investigations of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  removal by water purification processes, such as coagulation and powdered activated carbon (PAC) (Morton and Straub, 1955; Lettinga, 1972; Brown et al., 2008a, b). Brown et al. (2008a and 2008b) evaluated the treatability of radionuclides by each unit process applied to water purification plants based on data obtained from the literature. These data are useful, but in many cases the experiments were performed using only certain forms of radionuclide; e.g.,  $^{131}\text{I}^-$  and radioactive cesium ion ( $^{134}\text{Cs}^+$  and  $^{137}\text{Cs}^+$ ). As described above, radionuclides seem to exist in particulate and dissolved forms in environmental water. Thus, the removal of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  from environmental water may occasionally be different from those of  $^{131}\text{I}^-$ ,  $^{134}\text{Cs}^+$ , and  $^{137}\text{Cs}^+$ , respectively. It was reported that  $^{131}\text{I}^-$  removal by PAC was low (Lettinga, 1972); however, some other forms of  $^{131}\text{I}$  (e.g., organic  $^{131}\text{I}$ ) were removed by PAC (Summers et al., 1988). In addition, it was reported that ionic forms of  $^{137}\text{Cs}$ -radioactive barium ( $^{137}\text{Ba}$ ) were not removed by coagulation (Morton and Straub, 1955). However,  $\text{Cs}^+$  is known to be sorbed by soils and minerals (Cornell, 1993; Staunton, 1994). Thus, it was considered that particles of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  could be removed by coagulation. In fact, it was reported that removal of ionic forms of  $^{137}\text{Cs}$ - $^{137}\text{Ba}$  by coagulation was increased in the presence of clay (Morton and Straub, 1955). As in the case regarding the occurrence of radionuclides at water purification plants, only limited experimental data obtained using environmental water containing radionuclides are available.

In the present study, we investigated the removal of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  at water purification plants after the explosion at a nuclear power plant caused by the Great East Japan Earthquake. We also investigated  $^{131}\text{I}$  removal by PAC and removal of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  by coagulation taking their forms in environmental water into consideration.

## 2. Material and methods

### 2.1. Reagents and solutions

The reagents and solutions used are described in Supplementary Materials. Three types of PAC (PAC-1 to PAC-3) were used in the present study.

### 2.2. Sampling

Raw, process, and finished water samples at five water purification plants (WPP-1 to WPP-5) were collected in April 2011.

The details of these water purification plants and sampling procedures are described in Supplementary Materials. In cases where levels of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  levels in raw water were higher than 1 Bq/L, the data were used for analysis. Pond water was also collected from Iitate Village in Fukushima Prefecture on 7 May 2011. Water quality data of raw waters of WPP-1 to WPP-5 and pond water are listed in Table S1. Granular activated carbon (GAC) and sand from the rapid sand filter at WPP-1 were collected from April to September 2011. The GAC was that at depths of up to 5 cm from the surface. Moreover, sediments were collected from the Yodo and Ara Rivers, and were designated as sediments A and B, respectively.

### 2.3. Removal test of radioactive iodine and cesium

Removal of  $^{131}\text{I}$  in river and pond waters by PACs was investigated. To investigate the form of  $^{131}\text{I}$ , its removal by silver ion ( $\text{Ag}^+$ ) was also performed. Removal of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in river and pond waters by coagulation with polyaluminum chloride (PACl) was investigated. Removal of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in pond waters by PAC (PAC-1) and cation exchange resin was also investigated. The procedures of the removal tests of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  are described in Supplementary Materials.

### 2.4. Analytical methods

Gamma-emitting radionuclides ( $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$ ) in the samples were determined using high-purity germanium semiconductor detectors (GX2518; Canberra Co., Meriden, CT and EGPC20-190-R; EURYSIS Co., Cedex, France). In the case of occurrence of radionuclides at water purification plants, their levels were expressed as those on sampling days and time using their half-lives. In the radionuclide removal experiments, their levels on each experimental day were used. Other analytical methods are described in Supplementary Materials.

## 3. Results and discussion

### 3.1. Levels of radioactive iodine and cesium in drinking water after explosion at a nuclear power plant

After the explosion at a nuclear power plant, the levels of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  in drinking water have been investigated at Fukushima Prefecture and its 10 neighboring prefectures (Fig. S1) (Ikemoto and Magara, 2011). Figure S3 shows the results of monitoring of  $^{131}\text{I}$  in drinking water at Fukushima Prefecture and its 10 neighboring prefectures in March to May 2011 (WSD, HSB, MHLW, 2011; Asami and Akiba, 2011). During the monitoring, results indicating  $> 100$  Bq/kg of  $^{131}\text{I}$  in drinking water were only reported in March 2011.  $^{131}\text{I}$  in drinking water exceeded 300 Bq/kg and drinking water for the general public was restricted at 1 water utility in Iitate Village, Fukushima Prefecture (maximum, 965 Bq/kg).  $^{131}\text{I}$  in drinking water exceeded 100 Bq/kg and drinking water for infants was restricted at 20 water utilities in 5 prefectures (Fukushima, Ibaragi, Tochigi, and Chiba Prefectures, and Tokyo Metropolitan Government) (WSD, HSB, MHLW, 2011). Figure S4 shows the profiles of  $^{131}\text{I}$  in tap water in Iitate Village, Fukushima

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