Removal of radioactive iodine and cesium species from rainwater by a pot-type water purifier, and chemical and physical analysis of radioactive components in rainwater

Hirokazu Miyoshi

Radioisotope Research Center, the University of Tokushima, 3-18-15 Kuromoto-cho, Tokushima 770-8503, Japan

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A pot-type water purifier captured 98% of the nonradioactive iodine and iodide when an ion-exchange resin and activated carbon were used. Approximately 93.8% of $^{131}$I, 74 to 84.5% of $^{134}$Cs, and 75 to 81.1% of $^{137}$Cs in rainwater were captured after six repeated purifications using the purifier. The remaining 6.2% of $^{131}$I was not removed by isotope exchange with nonradioactive I or by reduction to I using Na$_2$S$_2$O$_4$. Approximately 19% of $^{131}$I, 40% of $^{134}$Cs, and 33% of $^{137}$Cs were adsorbed on the silica gel. Analysis of the removal rates of 73% for the mixed ion exchange resin, 59% for the silver ions, and 18% for the silver nanoparticle-bound clay indicated that the content of $^{131}$I adsorbed particles components was 23%. The concentration ratio of radioactive components in rainwater decreased from 5.9 to 1.4 upon ultrafiltration with a YM-1 membrane filter, indicating that a soft aggregation of the aerosol occurred. The IP and γ-ray spectrum analysis of the filter produced a cottonlike image of $^{131}$I. Analysis of the TEM image and its electron diffraction pattern indicated the aggregation of a small fraction of the SiO$_2$ aerosol. The coagulation of radioactive components in rainwater using I-adsorbed SiO$_2$ nanoparticles may be due to the exchange of the $^{131}$I isotope with I in I-SiO$_2$ and the adsorption of $^{134}$Cs$^+$ and $^{137}$Cs$^+$ on the surface of SiO$_2$ or the precipitation of the $^{131}$I-adsorbed aerosol and $^{134}$Cs$^+$- and $^{137}$Cs$^+$-adsorbed aerosols.

Key words: removal, radioactive iodine, radioactive cesium, rainwater, analysis

1. Introduction

The Fukushima Daiichi nuclear plant accident caused widespread contamination with the release of radioactive materials into the environment [1, 2]. Moreover, tap water was considerably contaminated by radioactive iodine from March to April [3]. A simple and cheap method of radioactive iodine removal was required at that time. Usually, iodine is captured by activated carbon [4] and iodide is captured by an ion exchange resin. Although we attempted to remove radioactive iodine and iodide using a pot-type water purifier, it was not easy to obtain sufficient amount of contaminated tap water for use in the removal experiments. Therefore, rainwater samples containing radioactive materials from Fukushima prefecture and the Tokyo metropolitan area were used in this study. The radioactive materials in rainwater include several chemical species such as iodine, iodide, cesium ion, and their adsorbed particles such as aerosol [2].

In this study, nonradioactive iodine solution was tested using a pot-type water purifier, and the capture of radioactive iodine in rainwater was investigated using the pot-type water purifier. The removal rate was determined from repeated purifications. The removal of the remaining radioactive iodine by isotope exchange with nonradioactive I, reduction to I using Na$_2$S$_2$O$_4$, and adsorption by silica gel was explored. Furthermore, each species in rainwater was evaluated by separating it to clarify the remaining components as follows. The selective capture of iodide, iodine, and cesium ions was carried out using an ion exchange resin, silver ions, and clay, respectively. The concentration of radioactive iodine in rainwater was determined by ultrafiltration using a YM-1 membrane filter, was carried out,
2. Materials and Methods

2.1. Materials

A 0.01 M iodine solution, sodium dithionite (Na₂S₂O₄), silica gel (Wakoel killer C-200, 75–150 µm particle size), and silica gel (medium granular) were purchased from Wako Pure Chemicals Co., Ltd. The iodine solution was diluted with distilled water and used without further purification. Rainwater samples obtained from Sasakino, Fukushima City on April 1, 2011 and from the Tokyo metropolitan area on March 22, 2011 were used. A pot-type water purifier, Brita Navelia (Brita Japan Co., Ltd.), was used. The filter cartridge used was the “CLASSIC” type. The water purification involved a 4-step filtration: intensive prefiltration with a fine mesh, ion exchange filtration, activated carbon filtration, and intensive final filtration with a special fine-mesh filter that retains the particle mixture [5]. Before using the pot-type water purifier, pretreatment using a tap water was performed in accordance with the manufacturer’s instructions.

2.2. Removal of nonradioactive iodine and iodide in an iodine solution and radioactive species in rainwater using a pot-type water purifier

A 0.01 mM nonradioactive iodine solution was prepared and a 100 ml portion was poured into the pot-type water purifier. The absorption spectrum of the filtrate was determined using a UV-vis spectrometer (Shimadzu UV-1700). These procedures were repeated five times using the same solution for the same water purifier. The removal rate was determined on the basis of the area of the absorption spectrum of the 0.01 mM solution. The obtained results were discussed to clarify the components of the radioactive species that were removed and not removed using the water purifier.

2.3. Selective capture of iodide, iodine, cesium ion, and particles such as aerosol using an ion exchange resin, silver ions, clay, and ultrafiltration

Rainwater (2011.4.1) from Sasakino, Fukushima City was used for adsorption and rainwater (2011.3.22) from the Tokyo metropolitan area was used for ultrafiltration. Ag-nanoparticle (Np)-bound clay (silver Nps/clay) [6], mixed ion exchange resin (Sigma, Amberlite MB-150), and Ag⁺-bound Zr₆(PO₄)₃ [7] were used for adsorption, and their adsorbents concentrations were sufficient for the amount of radioactive materials of the rainwater from a calculation of the iodine and iodide contents.

100 ml of rainwater (obtained from the Tokyo metropolitan area on March 22, 2011) was purified subsequently using three kinds of membrane filters (1.2, 0.45, and 0.2 µm pore sizes, Sartorius Co., Ltd.) for filtration and one (Amicon YM-100, 10 nm pore size, Millipore Co., Ltd.) for ultrafiltration. A 3 ml aliquot of each filtrate was obtained and its count rate was measured using an auto gamma counter (ARC-380, Aloka Co., Ltd.).

A 10 ml aliquot of the purified rainwater was used for further ultrafiltration using a membrane filter having a 2.6 nm pore size (Amicon YM-1, Millipore Co., Ltd.). After 3 ml of the filtrate was discarded, another 3 ml of the filtrate was obtained, and its count rate was measured using the gamma counter.
One gram of the mixed resin was added to 10 ml of the purified rainwater and was stirred ten times for 167 min for adsorption. Then, a 3 ml aliquot of the solution was obtained and its count rate was measured using the gamma counter.

Ag-Np-bound clay powder (0.1 g) was added to 10 ml of the purified rainwater and stirred ten times for 2 h 47 min. After the suspension was filtered using the YM-100 membrane filter, 3 ml of the filtrate was discarded, and another 3 ml of the filtrate was obtained; then, its count rate was measured using the gamma counter.

Ag⁺-bound Zr₂(PO₄)₃ powder (0.01 g) was added to 10 ml of the purified rainwater and stirred ten times for 2 h 47 min. After the suspension was filtered using the YM-100 membrane filter, 3 ml of the filtrate was discarded, and another 3 ml of the filtrate was obtained; then, its count rate was measured using the gamma counter.

2.4. Separation of radioactive materials in rainwater by fractionation using ultrafiltration and coagulation with silica Nps

After 490 ml of rainwater (obtained from the Tokyo metropolitan area on March 22, 2011) was filtered using the YM-100 membrane filter, a subsequent filtration using the YM-1 membrane filter was carried out. Finally, a 5 ml aliquot of the radioactive-particle-concentrated rainwater was obtained. The YM-1 filter paper was attached to an imaging plate (IP) (Fuji MS2040) for about 6 months, and the image was observed using Fluoanalyzer 9000 (Fujifilm Co., Ltd.). The γ-ray spectrum was determined for 281890 s using the Ge semiconductor detector. The count rates of the rainwater and filtrate were measured using the gamma counter. The rainwater was dropped on a Cu grid and dried. Transmission electron microscopy (TEM) observations were performed using Hitachi H-800.

A 0.1 ml aliquot of silica NPs (LUDOX, HS-30, 7 nm size) was dispersed in 10 ml of distilled water, and 0.01 M nonradioactive iodine solution was added at 1 ml each time until it became light yellow while stirring with a magnetic stirrer for 2 days. Then, the light yellow suspension was filtered using the YM-10 membrane filter, which was washed with distilled water until its color did not change. Finally, 1 ml of nonradioactive-iodine-adsorbed silica Nps (I-SiO₂ Nps) was obtained. This was added to 100 ml of rainwater (obtained from Sasakino, Fukushima City on April 1, 2011) and stirred for 2 days with a magnetic stirrer. Then, it was ultrafiltrated using a YM-10 membrane filter and 3.5 ml of the rainwater was obtained. The count rates of the rainwater and the filtrate were determined using the gamma counter, and ¹³¹I, ¹³⁴Cs, and ¹³⁷Cs were assigned using the Ge semiconductor detector.

The procedures in the above method are shown in Scheme 1 and are labeled 2-2, 2-3, and 2-4.
3. Results and Discussion

3.1. Capture of nonradioactive and radioactive iodine using a pot-type water purifier with ion exchange resin and activated carbon

Figure 1 shows absorption spectra of the iodine solution. As shown in Fig. 1, there were three peaks at 230, 290, and 350 nm, indicating that the 0.1 mM iodine solution was composed of I⁻ and I₃⁻ [8]. The absorptions of I₂ are at 460 and 270 nm [8]. In Fig. 1, the absorption at 460 nm was weak because of the low concentration. After the solution was poured into the pot-type water purifier, the absorbance decreased, indicating that iodide and iodine were captured by the water purifier. Five purifications were carried out and the average removal rate was 98% (n = 5). One L of 0.1 mM iodide solution contains 12.7 mg of iodine per 1 L and the calculated radioactivity of 12.7 mg as ¹³¹I corresponds to 58 GBq/L. Therefore, this indicates that until 56 GBq/L, the water purifier can remove the radioactive iodine from the contaminated water. This value was sufficient to remove ¹³¹I from the rainwater, if the chemical form of the ¹³¹I in the rainwater are iodine and iodide. This calculation is based on the experimental results for 0.1 mM nonradioactive iodine, not a maximum value. Actually, other nonradioactive materials were also adsorbed, and the maximum value of ¹³¹I may have been lower than the calculated value.

Figure 2 shows a γ-ray spectrum of the rainwater from Fukushima prefecture determined on April 9, 2011. As shown in Fig. 2, a large peak at 0.364 MeV and some subpeaks were detected and were assigned as follows: ¹²⁹Te at 0.459 MeV (7.7%); ¹³¹I at 0.081 MeV (2.6%), 0.284 MeV (6.1%), 0.364 MeV (81.7%), 0.637 MeV (7.2%), and 0.721 MeV (1.8%); ¹³⁴Cs at 0.569 MeV (15.4%), 0.605 MeV (97.6%), and 0.795 MeV (85.5%); ¹³⁷Cs at 0.661 MeV (85.1% ¹³⁷mBa); ¹²⁹mTe at 0.696 MeV (3.1%). Percentages indicate emission rates of γ-rays (%).

Figure 3 shows the average removal rates (n = 2) of ¹³¹I, ¹³⁴Cs, and ¹³⁷Cs based on consecutive purifications using the water purifier. The average removal rates increased from 81.2 to 93.8 % for ¹³¹I, 74 to 84.5% for ¹³⁴Cs, and 75 to 81.1% for ¹³⁷Cs after six purifications, indicating that the removal rate depended on the contact area and time. Kamei et al. reported that the adsorbed substances may detach when saturation is reached or over time owing to the physical adsorptive capacity of activated carbon, i.e., van der Waals forces [9]. Approximately 6.2% for ¹³¹I, 15.5% for ¹³⁴Cs, and 18.9% for ¹³⁷Cs remained, and the unremoved species may have different chemical forms such as particles, i.e., ¹³¹I-, ¹³⁴Cs-, and ¹³⁷Cs-adsorbed particles like aerosols. First, the removal of ¹³¹I was examined using isotope exchange with nonradioactive I⁻, by reduction to I⁻ using Na₂S₂O₄, and adsorption onto silica gel.
and silica column water are estimated to be ions such as different chemical forms of the radioactive materials. moved and about 6.2% remained. These indicated that there were some components that remained in the rainwater. For 3.2. Selective adsorption of radioactive iodine and cesium in rainwater using ion exchange resin, silver ions, and silver Nps Some experiments with the water purifier clarified that there were some components that remained in the rainwater. For example, approximately 93.8% of radioactive iodine was removed and about 6.2% remained. These indicated that there are different chemical forms of the radioactive materials. The chemical forms of the radioactive materials in the rainwater are estimated to be ions such as $^{131}$I, $^{134}$Cs, and $^{137}$Cs, molecules such as $^{131}$I$_2$, and particles such as aerosol-adsorbed $^{131}$I, $^{134}$Cs, and $^{137}$Cs. Each radioactive material requires an appropriate material to capture it. A mixed ion exchange resin can capture both ions of iodide such as $^{131}$I and cesium ions such as $^{134}$Cs and $^{137}$Cs. Silver ions can capture only iodide such as $^{131}$I. A silver Nps/clay can capture cesium ions such as $^{134}$Cs$^+$ and $^{137}$Cs$^+$ and iodine such as $^{131}$I$_2$.[10]. Hoskins and Karanfil have reported on the removal and sequestration of iodide using silver-impregnated activated carbon.[11]. Table 3 shows the total removal rates of $^{131}$I, $^{134}$Cs, and $^{137}$Cs using ion exchange resin, silver ions, and silver nanoparticles. Removal rates of 73% for the mixed ion exchange resin, 59% for the silver ions, and 18% for the silver Nps/clay were obtained, as shown in Table 3. From their results, some equations were derived as follows.

$$73\% = A + B, 27\% = C + D \quad (1)$$
$$59\% = A, 41\% = B + C + D \quad (2)$$
$$18\% = C + B, 82\% = A + D \quad (3)$$

Here, A represents $^{131}$I, B represents $^{134}$Cs$^+$ and $^{137}$Cs$^+$, C represents I$_{-}$, and D represents particles. The above equations yielded the following results: $A(131I) = 59\%$, $B(134Cs, 137Cs) = 14\%$, $C(I_2) = 4\%$, and $D$ (particles) = 23%. Then, the rate of $^{131}$I was 86% (59% + 4% + 23%) and that of $^{134}$Cs + $^{137}$Cs was 14%. Ohta et al. reported that about 30% of $^{131}$I was not retained by the resin and a portion of $^{131}$I became bound to organic matter from the soil.[12]. This almost agreed with the result for D (particle) above.

3.3. Separation of radioactive materials in rainwater by fractionation using ultrafiltration and coagulation with silica Nps

Table 4 shows total removal rates $^{131}$I, $^{134}$Cs, and $^{137}$Cs using filtration and ultrafiltration. As shown in Table 3, in the case of the YM-1 filter (2.6 nm pore size), the removal rates of $^{131}$I, $^{134}$Cs, and $^{137}$Cs became maximum, indicating that there are some granular substances larger than 2.6 nm. If it was particles, they will become concentrated using the YM-1 filter.

Table 3 Removal rates of total $^{131}$I, $^{134}$Cs, and $^{137}$Cs using ion exchange resin, silver ions, and silver Nps for rainwater sampling at Tokyo Metropolitan Area.

| Method            | Material                        | Removal rate (%) of ($^{131}$I + $^{134}$Cs + $^{137}$Cs) |
|-------------------|---------------------------------|----------------------------------------------------------|
| Ion Exchange Resin| Mixed Resin                     | 73                                                       |
| Silver Ions       | Ag/Zr$_2$ (PO$_4$)$_3$          | 59                                                       |
| Silver Nanoparticles (Nps) | Ag Nps/Clay                    | 18                                                       |

Use removal rates (%) of total $^{131}$I, $^{134}$Cs, and $^{137}$Cs: (Net count rate of rainwater before each treatment minus net count rate of rainwater after each treatment was divided by net count rate of rainwater before each treatment.)
Table 4  Rates of total $^{131}$I, $^{134}$Cs, and $^{137}$Cs removal using filtration and ultrafiltration for rainwater sampling at Tokyo Metropolitan Area.

| Method                        | Filter Material (Pore Size)   | Removal rate (%) of $(^{131}$I + $^{134}$Cs + $^{137}$Cs) |
|-------------------------------|-------------------------------|-------------------------------------------------------------|
| Filtration                    | Membrane Filter (1.2 μm)      | 21                                                          |
|                               | Membrane Filter (0.45 μm)     | -27                                                         |
|                               | Membrane Filter (0.2 μm)      | 5                                                           |
| Ultrafiltration               | YM-100 Filter (10 nm)         | -11                                                         |
|                               | YM-1 Filter (2.6 nm)          | 132                                                         |

Removal rates (%) of total $^{131}$I, $^{134}$Cs, and $^{137}$Cs: (Net count rate of rainwater minus net count rate of each treated rainwater) was divided by net count rate of rainwater.

Table 5 shows the concentration of radioactive components obtained by ultrafiltration. First, the concentration ratio was 5.9, then, the concentration ratio decreased to 1.4, indicating that there was soft aggregation of the aerosol.

Figure 4 shows an IP image of the YM-1 filter after the above-mentioned ultrafiltration. As shown in Fig. 4, the image is like cotton distributed around the filter surface.

Figure 5 shows the $\gamma$-ray spectrum of the filter, which indicated that there were no peaks of $^{134}$Cs and $^{137}$Cs. That is, at that time put the filter on the IP $^{131}$I was the major radioisotope adsorbed on the filter. This supported the notion that $^{131}$I was the major radioisotope released at the early stage of the accident.

Figure 6 shows a TEM image of the 1.4-times concentrated rainwater; the aggregation of a small fraction of the aerosol can be seen in the rectangular image. From the electron diffraction pattern of the observed area, the d-values in Table 6 were observed, which assigned to silica (SiO$_2$) (JCPDS 11-695), suggesting the presence of silica components. This indicates that silica composed of aerosol in the air adsorbed the released radioisotope, and it could not be removed by the water purifier. This suggests that iodine molecules may adsorb on the silica Nps. Therefore, I-SiO$_2$ NPs were prepared by mixing the I$^3$- solution and silica Nps (7 nm) to obtain I-SiO$_2$ NPs. Then, the I-SiO$_2$ NPs were dispersed in rainwater and ultrafiltrated using the YM-10 membrane filter.

Table 7 shows the coagulation of radioactive components using I-SiO$_2$. The precipitate contained $^{131}$I, $^{134}$Cs, and $^{137}$Cs. Since the cesium ion has a positive charge, it is easy to adsorb on the surface of SiO$_2$, or cesium-adsorbed aerosol is easy to coagulate with SiO$_2$. Furthermore, the precipitate indicated a peak of $^{131}$I; this indicates that the $^{131}$I isotope exchanged with I of I-SiO$_2$, or $^{131}$I-adsorbed aerosol is also easy to coagulate with SiO$_2$. This supports the adsorption of $^{131}$I on the silica composed of aerosol.

Table 5  Concentration of radioactive components after ultrafiltration.

| Method                              | Sample                          | Net Count Rate/cpm | Ratio of Count Rate |
|-------------------------------------|---------------------------------|--------------------|---------------------|
|                                    | Type of Water                   | Place of Collection| Concentrated Solution| Filtrate          | Concentrated Solution/Filtrate |
| Concentration using ultrafiltration with YM-1 filter (1) | Rainwater (1) + Rainwater Tokyo Metropolitan Area | 323 55 5.9 | 220 156 1.4 |

Removal rates (%) of total $^{131}$I, $^{134}$Cs, and $^{137}$Cs: (Net count rate of rainwater minus net count rate of each treated rainwater) was divided by net count rate of rainwater.
4. Conclusions

Approximately 93.8% of $^{131}$I, 74 to 84.5% of $^{134}$Cs, and 75 to 81.1% of $^{137}$Cs in rainwater were captured after six repeated purifications using a pot-type water purifier. The remaining 6.2% of $^{131}$I was not removed by isotope exchange with nonradioactive I or by reduction to I using Na$_2$S$_2$O$_4$. Approximately 19% of $^{131}$I, 40% of $^{134}$Cs, and 33% of $^{137}$Cs were adsorbed on the silica gel. Analysis of the removal rates of 73% for the mixed ion-exchange resin, 59% for the silver ions, and 18% for the silver Nps/clay indicated that the content of $^{131}$I adsorbed particles components was 23%. The concentration ratio of radioactive components in rainwater decreased from 5.9 to 1.4 upon ultrafiltration with a YM-1 membrane filter, indicating that there was a soft aggregation of the aerosol occurred. The IP and $\gamma$-ray spectrum analysis of the filter produced a cottonlike image of $^{131}$I. Analysis of the TEM image and its electron diffraction pattern indicated the aggregation of a small fraction of the SiO$_2$ aerosol, suggesting that it may be a component that is not removed by the water purifier. The coagulation of radioactive components in rainwater using I-adsorbed SiO$_2$ nanoparticles may be due to the exchange of the $^{131}$I isotope with I in I-SiO$_2$ and the adsorption of $^{134}$Cs$^+$ and $^{137}$Cs$^+$ on the surface of SiO$_2$ or the precipitation of the $^{131}$I-adsorbed aerosol and $^{134}$Cs- and $^{137}$Cs-adsorbed aerosols. Finally, it was found that rainwater had $^{131}$I, $^{134}$Cs, and $^{137}$Cs components adsorbed on the SiO$_2$ aerosol, which could not be removed by the water purifier. To remove them, a coagulation method involving mixing with I-adsorbed SiO$_2$ nanoparticles was effective.

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