Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.923205

54 (2015) 3494–3501
June

Taylor & Francis
Taylor & Francis Group

Removal of radioactive iodine and cesium in water purification

S. Kitada[a,*], T. Oikawa[b], S. Watanabe[b], K. Nagai[b], Y. Kobayashi[b], M. Matsuki[b], K. Tsuchiya[b], K. Nakamura[b], M. Sasaki[b], Y. Shinoda[b], T. Iwamoto[c]

[a]*Murayama-Yamaguchi Reservoir Management Office, Bureau of Waterworks, Tokyo Metropolitan Government, 2-5-2, Syohrakuji, Tokorozawa, Saitama, Japan, email: kitada-shingo@waterworks.metro.tokyo.jp*

[b]*Water Quality Management Center, Bureau of Waterworks, Tokyo Metropolitan Government, 2-7-1, Hongo, Bunkyo, Tokyo, Japan, emails: oikata-tomo@waterworks.metro.tokyo.jp (T. Oikawa), watanabe-soichi@waterworks.metro.tokyo.jp (S. Watanabe), nagai-kouichi@waterworks.metro.tokyo.jp (K. Nagai), kobayashi-yasuhiro@waterworks.metro.tokyo.jp (Y. Kobayashi), matsuki-masahiro@waterworks.metro.tokyo.jp (M. Matsuki), tsuchiya-kaori@waterworks.metro.tokyo.jp (K. Tsuchiya), nakamura-kayo@waterworks.metro.tokyo.jp (K. Nakamura), sasaki-makoto@waterworks.metro.tokyo.jp (M. Sasaki), shinoda-yutaka@waterworks.metro.tokyo.jp (Y. Shinoda)*

[c]*Purification Division, Bureau of Waterworks, Tokyo Metropolitan Government, 2-8-1, Nishi-Shinjuku, Shinjuku, Tokyo, Japan, email: iwamoto-tomoe@waterworks.metro.tokyo.jp*

Received 15 January 2014; Accepted 14 March 2014

ABSTRACT

Radioactive iodine, one of the radionuclides released in the nuclear power plant accident on 11 March 2011, was detected in purified water at water purification plants (WPPs). However, information about removal of radioactive materials in actual water purification process was limited. Therefore, we investigated the removal of radioactive materials (iodine and cesium) immediately after the detection. It is found that non-radioactive iodine in water could be removed by the combined use of pre-chlorination and powdered activated carbon (PAC) treatment. The same result was also obtained in terms of radioactive iodine. Removal of non-radioactive iodine in WPPs was also investigated. Approximately, 60% of iodine was removed by combination of pre-chlorination (0.5–1.0 mg/L) and PAC (15–30 mg/L) in coagulation and sedimentation processes. In water purification process, cesium was mostly removed by coagulation and sedimentation; hence, radioactive cesium was not detected in purified water. It was confirmed that a thorough turbidity control is essential for the prevention of radioactive cesium contamination of purified water. Meanwhile, radioactive iodine in purified water has not been detected since 5 April 2011 and radioactive cesium since 22 March 2011, when the measurement was commenced. Moreover, dehydrated sludge including radioactive cesium has been treated in accordance with Japanese laws and regulations.

Keywords: Coagulation and sedimentation process; Pre-chlorination; Powdered activated carbon; Radioactive iodine; Radioactive cesium

---

*Corresponding author.

Presented at the 5th IWA-ASPIRE Conference, 8–12 September 2013, Daejeon, Korea

1. Introduction

The Great East Japan Earthquake on 11 March 2011 damaged the Fukushima Daiichi nuclear power plant, causing the release of large amounts of radioactive materials such as iodine, cesium and others. They occurred mainly on 15 March, 16 March, and 21 March [1,2]. On 21 March, there were concerns that the radioactive materials concentration in raw water would increase due to the first rainfall after the nuclear power plant accident. Based on the reported cases where powdered activated carbon (PAC) was used to remove radioactive iodine, on 19 March 2011, the Ministry of Health, Labour and Welfare (MHLW) issued a notification to public water suppliers to inject PAC for the removal of radioactive iodine. On 22 March, Tokyo Waterworks measured radioactivity in purified water at Kanamachi Purification Plant. From the reports, we found that the value of radioactive cesium (^{134}Cs, ^{137}Cs) was reduced below detection limits, but that of radioactive iodine (^{131}I) was 210 Bq/kg, which exceeded the provisional regulation value for radioactivity in drinking water for infants (100 Bq/kg, cf. 300 Bq/kg for other than infants). Thus, Tokyo Waterworks quickly issued a restriction on infant consumption of drinking water in the distribution areas of Kanamachi and Misato Purification Plants, both of which are located in the same water system. However, information about removal of radioactive materials in actual water purification process was limited. Therefore, we launched a study on the removal of radioactive materials through the water purification process.

In the case of radioactive iodine, there have been reported cases in which PAC treatment was used to remove radioactive iodine [3–6]. Specifically, it has been reported that the removal ratio of PAC treatment improved after the addition of chlorine [3,4]. The form of radioactive iodine in water is an important factor relating to its effective removal through the water purification process. Iodine is present mainly as iodide ion (I^-) and iodate ion (IO_3^-) in water [7], and most of the iodine in tap water exists as IO_3^- due to the oxidation of I^- by chlorination [8]. In addition, it has been reported that PAC treatment was insufficient for removing IO_3^- [3]. Therefore, we speculated that although PAC is effective in removing radioactive iodine from water, its removal ratio might significantly depend on the form of iodine. Furthermore, immediately after the nuclear accident, there was no clear information about what conditions should be applied to the oxidation processes, such as chlorination, in order to remove radioactive iodine. Therefore, we conducted experiments relating to the removal of radioactive iodine by PAC treatment with pre-chlorination process.

Radioactive cesium has higher solid–liquid distribution coefficient (K_d), the adsorption index of radioactive materials of soil, than radioactive iodine [9]. K_d of radioactive cesium is 1,200 on average, whereas that of radioactive iodine is 6.9. Therefore, in the environment, radioactive cesium exists mainly in the adsorbed form in the soil, and is assumed to flow into water purification plants (WPPs) with suspended particles. According to the measurement of Tokyo Waterworks, radioactive cesium had not been detected in purified water, but occasionally in raw water, while highly concentrated radioactive cesium, exceeding 10,000 Bq/kg, was detected in the dehydrated sludge of the plant right after the nuclear accidents. From these facts, we assumed that radioactive cesium in raw water was removed by the water purification process and concentrated to dehydrated sludge. Nevertheless, information concerning the removal of radioactive cesium in WPPs was insufficient.

Therefore, with due consideration given to the forms of iodine, we conducted experiments relating to the removal of radioactive iodine by PAC treatment along with pre-chlorination process and investigated the optimal conditions of pre-chlorination. Furthermore, we verified the validity of these conditions at actual WPPs. Moreover, we investigated the removal of radioactive cesium by means of water purification treatment and confirmed the importance of turbidity control.

2. Materials and methods

2.1. Removal of radioactive iodine

2.1.1. Experiments of non-radioactive iodine removal

As iodine is present mainly as I^- or IO_3^- in water, jar tests were conducted to assess the effectiveness of PAC treatment alone and the combined use of pre-chlorination and PAC. Since the concentration of I^- in raw water was too low to measure with ion chromatograph (IC), raw water of Misato Purification Plant was used for jar tests, with the addition of approximately 1 mg/L potassium iodide (KI) or potassium iodate (KIO_3). The conditions of raw water used in the jar tests are shown in Table 1. PAC, sodium hypochlorite (NaOCl) and coagulant (poly-aluminium chloride) used were the same as at the Misato Purification Plant. In the jar tests, NaOCl was added 30 min after PAC injection, and then poly-aluminium chloride (30 mg/L) was added after a lapse of 30 s, and the samples underwent rapid agitation(120 rpm, 2 min.), slow agitation (55 rpm, 10 min.) and clarification (10 min.). The clarified water was filtered with the 0.45 μm

Table 1
Conditions of raw water used in the jar tests

	Water temperature (℃)	turbidity (Degree)	pH (–)	$KMnO_4$ consumption (mg/L)	TOC* (mg/L)
Mean value	9.5	6.9	7.5	5.6	1.2
range	(8.2–10.6)	(5.5–11)	(7.4–7.5)	(4.5–7.9)	(1.2)

*TOC: n=1.

pore-sized membrane filter, and then the filtered water was used as a sample to measure I^- and IO_3^- with IC (IC-2000, Dionex) with IonPac AS-19 column using 15–50 mM (gradient) KOH solution as a mobile phase. ECD detector was used for the detection of I^-. IO_3^- was detected by UV-detection (268 nm) with post-column derivatization method using reagents of 1.2 mM $NaNO_2$ and 1.0 M H_2SO_4/1.5 M KBr.

2.1.2. Experiments of radioactive iodine removal

In order to verify the results of the experimental jar tests using raw water with the addition of non-radioactive iodine, the removal of radioactive iodine was examined. Jar tests using raw water at the Misato Purification Plant, with the addition of the rainwater containing radioactive iodine (^{131}I), were conducted and the concentrations of radioactive iodine contained in the clarified water were measured with germanium semiconductor detector (GEM-25,185-P, ORTEC). Treatment chemicals and conditions for the jar tests were the same as those of the jar tests using raw water with the addition of non-radioactive iodine.

2.1.3. Verification of iodine removal in WPPs

To verify the validity of the obtained results from our experimental jar tests relating to radioactive iodine removal, measurements using raw and/or process water at actual WPPs were carried out. Since the stable procurement of PAC was difficult after the earthquake, PAC dose at each WPPs was determined in accordance with its availability. Therefore, the actual PAC dose on the verification was set as 10–30 mg/L. ICP-MS was used in order to measure low concentrations of iodine comparable to the concentration levels in river water. The ratio of total iodine to ^{127}I was measured with ICP-MS (Agilent, 7500cx) after adding dilute nitric acid (1% v/v) in samples. In pre-treatment, samples were not heated in order to prevent the separation of molecular iodine (I_2) under acidic conditions. Determination limit was 0.13 µg/L with a coefficient of variation (CV) of be 10%. IO_3^- was measured by IC (ICS-3,000, Dionex) with IonPac AS11 column using 2 mM KOH solution as an eluent and flow ratio was 1.0 mL/min. IO_3^- was detected by UV-detection (268 nm) with post-column derivatization method using reagents of 1.2 mM $NaNO_2$ and 1.0 M H_2SO_4/1.5 M KBr. Determination limit was 0.5 µg/L with a CV of 10%.

2.2. Behavior of radioactive cesium

2.2.1. Cesium removal ratio on water purification process

The removal ratio of radioactive cesium in the water purification processes was estimated by measuring non-radioactive cesium (^{133}Cs), which is the isotope of radioactive cesium (^{134}Cs and ^{137}Cs), using ICP-MS. The total cesium concentration was measured with ICP-MS after adding dilute nitric acid (1% v/v). The dissolved cesium concentration was measured by using filtrated samples with membrane filter (Mixed Cellulose Ester, 1.0 µm, Advantec). The concentration of particles that adsorbed cesium was derived by subtracting dissolved cesium from the total cesium. Also, the removability of radioactive cesium by coagulation and sedimentation was verified by means of measuring radioactive cesium with germanium semiconductor detectors, using high-turbidity raw waters of actual WPPs. The measurement of radioactive cesium was conducted for 100 times longer than usual. The average turbidity values of raw and clarified water used for the measurement were 240 and 0.6 degrees, respectively.

2.2.2. Estimating radioactive cesium concentration in raw water based on the dehydrated sludge

According to Tokyo Waterworks measurements, highly concentrated radioactive cesium had been detected in dehydrated sludge, while it was detected occasionally in raw water at low levels. Therefore, it was assumed that certain amount of radioactive cesium existed in raw water, but the concentration of it was under the detection limit (approximately 1–2 Bq/kg). To grasp the actual concentration of radioactive cesium in raw water, we estimated it from measurement results of dehydrated sludge. The amount of suspended substances in raw water was calculated by deducting the amount of poly-aluminium chloride and PAC that were injected in the water purification process from the amount of solids in dehydrated sludge. The concentration of radioactive cesium was calculated with the assumption that all radioactive cesium in dehydrated sludge exists in suspended substances in raw water. In the calculation, mean values for the turbidity of raw water and dose of chemicals between 7 and 14 d before the sampling of dehydrated sludge were used, because it takes about 7 d from water intake to the dehydration of sludge. Furthermore, the average amount of dehydrated sludge for 7 d before the sampling day was used.

3. Results and discussion

3.1. Removal of radioactive iodine

3.1.1. Experiment of non-radioactive iodine removal

Fig. 1 shows the removal ratio of I^- and IO_3^-. After 30 min of contact time of PAC, poly-aluminium chloride was added into the raw water with the prior addition of I^- and IO_3^-. The removal efficiency for I^- and IO_3^- was approximately 6% with a 40-mg/L dose of PAC. Consequently, it was found that I^- and IO_3^- were unable to be removed sufficiently by coagulation and sedimentation processes with the addition of PAC alone.

Then, the jar tests were conducted to assess the efficacy of the combined use of PAC treatment and pre-chlorination. PAC dose was set as 15 mg/L in order to evaluate whether the combined use was able to remove I^- on the actual dose at the Misato PP. Fig. 2 shows the removal ratio of I^- at the jar test using raw water with the addition of I^- under the conditions of 0–3 mg/L pre-chlorination and 15 mg/L PAC. When chlorine was added at a dose of 0.5 or 1.0 mg/L, I^- was removed to some extent (nearly 20–30%). Therefore, it was found that the combined use of PAC and pre-chlorination was able to remove I^- in the actual treatment condition at WPPs.

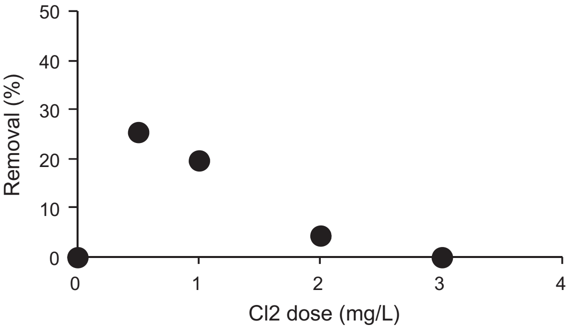

Fig. 2. Relation between dose of chlorine and removal of I^- with PAC treatment.

3.1.2. Experiments of radioactive iodine removal

The removal capability of the combined use was investigated. Fig. 3 shows the removal ratio of I^- in the jar test using raw water and raw water with the addition of the rainwater containing radioactive iodine (57 Bq/kg) under the conditions of 0.5 mg/L pre-chlorination and 0–50 mg/L PAC treatment. Removal of 40–60% of I^- with the combined use of pre-chlorination (0.5 mg/L) and PAC was confirmed, when 20–30 mg/L of PAC was added. Based on these results, the combined use of 0.5–1.0 mg/L chlorine and PAC was confirmed to be effective for removing radioactive iodine.

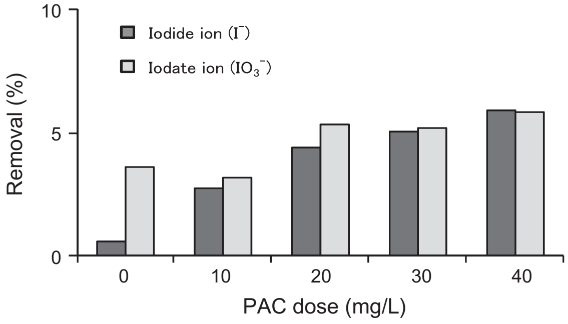

Fig. 1. Removal of I^- or IO_3^- using PAC alone.

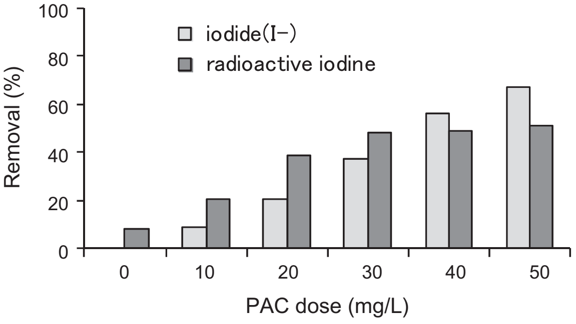

Fig. 3. Relation between dose of PAC and removal ratio of I^- and radioactive iodine under condition of pre-chlorination.

3.1.3. Verification of iodine removal on WPPs

Fig. 4 shows the removal ratio of total iodine using conventional treatment with 1.0 mg/L of chlorine and 10–30 mg/L dose of PAC. The concentration of iodine in raw water was nearly 10 µg/L, which was well above the limit of measurement with ICP-MS. It showed that the removal ratio of iodine was approximately 60% with the combined use of pre-chlorination (1.0 mg/L) and 15–30 mg/L dose of PAC, which was almost identical to the results obtained at the jar tests.

Fig. 5 shows the ratio of each form that iodine takes in clarified water in advanced water treatment stages (in which ozonation and biological activated carbon treatment are incorporated), which was measured with ICP-MS or IC. As shown in Fig. 5, ratio of IO_3^- in total iodine was low in raw or clarified water. On the other hand, the majority of iodine was oxidized to IO_3^- with ozonation, the proportion of IO_3^- in total iodine steeply increased. The removal ratio of total iodine was low with advanced water treatment. Biological activated carbon (BAC) treatment was ineffective for removing IO_3^-, and this treatment was partially effective for the elimination of the other forms of iodine. Based on the results, BAC treatment after ozonation was insufficient for removing radioactive iodine and therefore, the combined use of pre-chlorination and PAC was necessary for the effective removal of radioactive iodine even in advanced water treatment.

3.1.4. Measures to remove radioactive iodine on WPPs

From 21 March 2011, which was the day of the first rainfall after the nuclear power plant accident, the Tokyo Waterworks started PAC injection for the reduction of radioactive iodine. However, on 22 March, the radioactive iodine was detected in high concentrations in purified water coming from the Kanamachi Purification Plant. In view of these circumstances, the Tokyo Waterworks quickly stepped up their efforts to employ PAC injection on March 23.

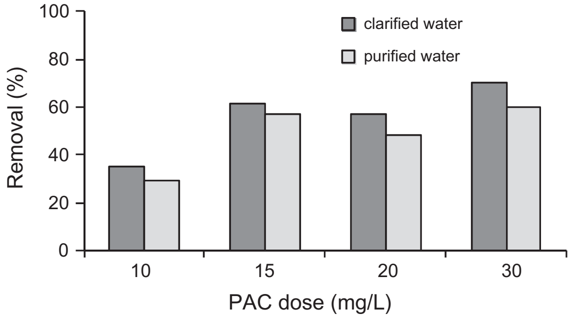

Fig. 4. Relation between PAC dose and removal ratio of total iodine on clarified/purified water of WPPs.

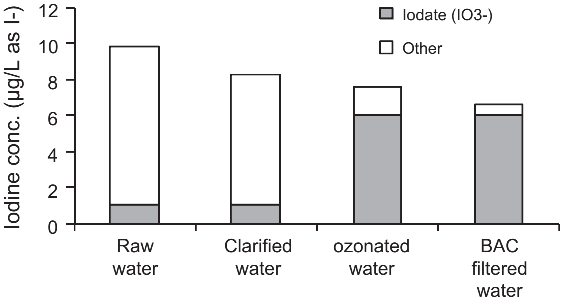

Fig. 5. Ratio of IO_3^- in total iodine in process water in advanced water treatment.

After it had been shown that the combined use of pre-chlorination and PAC demonstrated effective non-radioactive iodine removal at experimental jar tests, from 28 March, the Tokyo Waterworks started the combined use of pre-chlorination (0.5–1.0 mg/L dose of chlorine) and PAC at the Kanamachi and Misato Purification Plants, which treat the raw water of the same river. On 30 March 2011, it was determined that the combined use of pre-chlorination and PAC was to be applied at all major WPPs. Then, up to 8 June, the combined use of pre-chlorination and PAC was implemented at all major WPPs to reduce the possibility that radioactive iodine concentrations would increase due to rainfall. As a result, radioactive iodine has not been detected in purified water since 5 April 2011.

Furthermore, our results were quickly shared with the relevant bodies, including MHLW, Japan Water Works Association, and other public water utilities. Subsequent to public announcement of MHLW in June 2011, the combined treatment became a national guideline for the reduction of radioactive iodine.

3.2. Behavior of radioactive cesium

3.2.1. Verification of the removability of cesium in water purification process

Fig. 6 shows the total concentration of non-radioactive cesium in coagulation and sedimentation processes. The concentration of cesium decreased significantly in settled water compared to those in raw water, which means the cesium present in raw water had been removed to a great extent in coagulation and sedimentation processes. It was reported that cesium binded strongly with clay mineral [10]. Therefore, most of the cesium in raw water was speculated to be adsorbed to suspended particle.

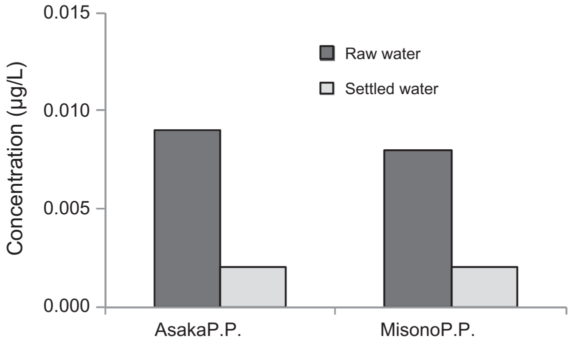

Fig. 6. Total cesium concentration by coagulation and sedimentation process.

In this regard, we measured radioactive cesium in raw and process waters with high turbidity at actual purification plants using germanium semiconductor detectors, and verified its removal ratio by coagulation and sedimentation. Fig. 7 shows the results. It was found that 93% of radioactive cesium in raw water was removed by coagulation and sedimentation. Consequently, it is supposed that all cesium can be removed by means of thoroughly removing suspended substances even when raw water is highly turbid, because radioactive cesium has not been detected in purified water so far.

3.2.2. Estimating radioactive cesium concentration in raw water based on those in dehydrated sludge

Fig. 8 shows the results of radioactive cesium concentration in dehydrated sludge in the Kanamachi Purification Plant from right after the accident to March 2013. In dehydrated sludge, the high radioactive cesium concentration of 14,650 Bq/kg was detected on 28 March 2011. However, it decreased drastically and went down to less than 1,000 Bq/kg since September 2011. As of March 2013, it was around 400 Bq/kg.

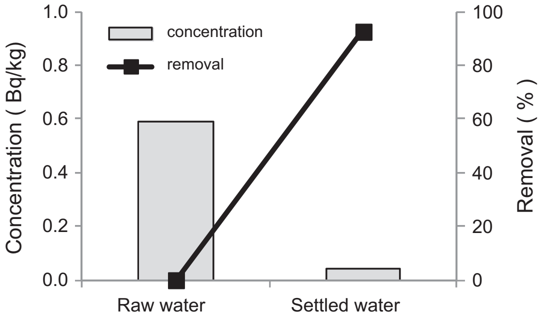

Fig. 7. Removal ratio of radioactive cesium at the time of high turbidity in water.

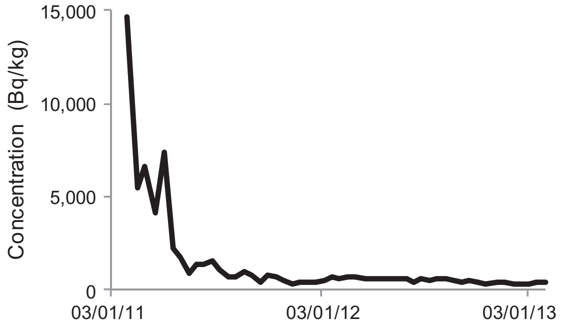

Fig. 8. Trend in radioactive cesium contamination in dehydrated sludge.

Table 2 shows the result of estimation for radioactive cesium concentration in raw water based on those in dehydrated sludge at the Kanamachi Purification Plant. While the concentration in dehydrated sludge right after the accident showed the highest, on 28 March 2011, the estimated value of radioactive cesium concentration in raw water was quite low: 1.2 Bq/kg. Even in the last estimation results (as of March 2013), the concentration of radioactive cesium in raw water is estimated about 0.02–0.04 Bq/kg, which is only 1/250 of the WHO guidance level for drinking water (10 Bq/kg) and the water quality management level in Japan (10 Bq/kg, which was revised in April 2012 by MHLW). Furthermore, it is less than 1/25 in comparison with the detection limit of 0.7–1.0 Bq/kg in current measurement conditions. The estimated concentration of radioactive cesium in raw water, converted into per 1 degree of turbidity, decreased from 0.171 Bq/kg/degree right after the accident to 0.003 Bq/kg/degree at present. So, it is suggested that the concentration of radioactive cesium in environment water is also decreasing.

In addition, MHLW proposed an interim policy in June 2011 for the handling of dehydrated sludge containing radioactive cesium. This allows landfill disposal of such sludge if its concentration is 8,000 Bq/kg or less, and its reuse in cement manufacture if the concentration is 100 Bq/kg or less. Also in March 2013, MHLW announced an additional policy that allows the reuse of dehydrated sludge for garden soil if its concentration is 400 Bq/kg or less, and for ground soil if the concentration is 200 Bq/kg or less. Currently, dehydrated sludge containing radioactive cesium is disposed as landfill in compliance with this standard; Ozaku Purification Plant, in which radioactive cesium has become almost undetectable, has resumed its sludge recycling and utilization since 17 October 2012.

Table 2
Estimated value of radioactive cesium concentration in raw water based on those in dehydrated sludge

$^{134,137}Cs$	Dehydrated sludge (measured value) (Bq/kg)	Raw water (estimate value) (Bq/kg)	Average raw water turbidity (degree)	per 1 degree turbidity (estimate value) (Bq/kg)
28 March 2011	14,650	1.20	7.0	0.171
14 Apr	5,430	0.55	5.7	0.096
27	6,570	0.52	6.3	0.083
17 May	4,100	0.36	7.2	0.050
Jun1	7,400	0.48	8.8	0.055
:	:	:	:	:
15 Mar 2013	375	0.02	9.5	0.002
29	357	0.04	13	0.003

4. Conclusions

While PAC treatment alone hardly removed I^- and IO_3^- in water, the combined use of pre-chlorination (0.5–1.0 mg/L) and PAC demonstrated the effective removal of I^-. Efficacy of the combined use of pre-chlorination and PAC also was confirmed by jar tests using raw water containing radioactive iodine. Furthermore, removal ratio of total iodine was approximately 60% on the condition of 0.5–1.0 mg/L pre-chlorination and 10–30 mg/L PAC at actual WPPs. In advanced water treatments, I^- was oxidized to IO_3^- by ozonation; therefore, iodine was hardly removed by following biological activated carbon treatment. Whereas, starting the removal of radioactive iodine a week after its detection under optimum conditions utilizing the experiment results, Tokyo Waterworks provided the relevant information promptly to the government and relevant agencies. This approach contributed to the formulation of government measures against radioactive contamination. Moreover, our results were quickly shared with the relevant bodies, including the national government and other public water utilities, and finally, the combined treatment became a national guideline for the reduction of radioactive substances.

Radioactive cesium in raw water exists mainly in the particle-adsorbed state. Therefore, it can be removed to a great extent by coagulation and sedimentation processes. However, there is a risk that concentration of radioactive cesium in raw water increases in conditions such as the case of nuclear accident and high turbidity of raw water just after the accident. In order to prevent the leak of radioactive cesium into purified water, strict water quality management in the aspect of turbidity is required in water purification processes. We estimated the radioactive cesium concentration in raw water based on those in dehydrated sludge. The estimation showed that maximum concentration of radioactive cesium in raw water was 1.2 Bq/kg in March 2011. In March 2013, it was calculated to be about 0.02–0.04 Bq/kg, which is greatly less than WHO guidance level for drinking water and the water quality management level in Japan (10 Bq/kg).

In addition, radioactive iodine has not been detected in purified water since 5 April 2011, neither radioactive cesium since 22 March 2011, when the measurement was started. Also, dehydrated sludge containing radioactive cesium has been treated in accordance with Japanese laws and regulations.

References

[1] Y. Nagakawa, T. Suzuki, Y. Kinjo, N. Miyazaki, M. Sekiguchi, N. Sakurai, H. Ise, Measurement of the ambient radioactivity and estimation of human radiation exposure dose in Tokyo with regard to the radioactive substance leakage due to the Fukushima Daiichi nuclear power plant accident, Radioisotopes 60 (2012) 467–472 (in Japanese).

[2] K. Nagaoka, S. Sato, S. Araki, Y. Ohta, Y. Ikeuchi, Changes of radionuclides in the environment in Chiba, Japan, after the Fukushima nuclear power plant accident, Health Phys. 102(4) (2012) 437–442.

[3] G. Lettinga, Radioactivity and water supplies, PhD. thesis, Delft University of Technology, Delft, 1972.

[4] R.S. Summers, F. Fuchs, H. Sontheimer, The fate and removal of radioactive iodine in the aquatic environment, *Aquatic Humic Substance*, ACS Publications, 1988, pp. 623–636.

[5] S. Honma, T. Takahashi, K. Yamagaki, Radioactive contamination of water supply in Niigata City by Chernobyl Reactor accident and its removal experiments using powder activated carbon, J. Jpn. Water Works Assoc. 57(1) (1988) 24–29 (in Japanese).

[6] J. Smith, O.V. Voitsekhovitch, L. Håkanson, J. Hilton, A critical review of measures to reduce radioactive doses from drinking water and consumption of freshwater foodstuffs, J. Environ. Radioact. 56 (2001) 11–32.

[7] H. Takagi, I. Iijima, K. Iwashima, Special Articles: Environmental Sciences and Analytical Chemistry. Determination of iodine with chemical forms in rain water by fractional sampling/NAA, Bunseki Kagaku 43 (1994) 905–909 (in Japanese).

[8] K. Kametani, T. Matsumura, M. Naito, Separation of iodide and iodate by anion exchange resin and determination of their ions in surface water, Bunseki Kagaku 41 (1992) 905–909 (in Japanese).

[9] IAEA, Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environment, (2010) Technical Reports Series No. 472.

[10] H. Tsukada, A. Takeda, S. Hisamatsu, J. Inaba, Concentration and specific activity of fallout ^{137}Cs in extracted and particle‐size fractions of cultivated soils, J. Environ. Radioact. 99 (2008) 875–881.