Efforts on radioactivity after the Great East Japan Earthquake ~ Radioactivity Response Measures Taken by the Tokyo Waterworks ~

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Abstract: Radioactive iodine was detected in the purified water of the water purification plant in Tokyo due to the nuclear power plant accident in the wake of the Great East Japan Earthquake on March 2011. The Bureau of Waterworks, Tokyo Metropolitan Government immediately started investigation of the removal of radioactive materials. The survey revealed that radioactive iodine can be removed by a combination of powdered activated carbon and pre-chlorination. Consequently, the result enabled the water purification plants to deal with radioactive iodine under an optimum treatment condition, approximately one week after the detection. Radioactive cesium was almost removed by coagulation and sedimentation and it was not detected in purified water. Also, based on the high interest in radioactive materials by customers, the Bureau of Waterworks, Tokyo Metropolitan Government has been continuing daily measurements of radioactive materials and publication of results from immediately after the nuclear disaster to date in order to relieve their concerns.

Keywords: radioactive iodine; radioactive cesium; monitoring; the East Japan Great Earthquake

Introduction

A nuclear disaster occurred at the Fukushima Daiichi Nuclear Power Plant due to the Great East Japan Great Earthquake on March 11, 2011, and radioactive materials were released into the environment. On March 21, when the first rainfall occurred after the nuclear accident, the influence of radioactive materials on the water supply was concerned. The Bureau of Waterworks, Tokyo Metropolitan Government (hereinafter called, "Tokyo Waterworks") measured radioactive materials in the purified water of the Kanamachi Water Purification Plant (WPP), which was sampled on March 22. As a result, radioactive cesium (^{134}Cs, ^{137}Cs) was below the detection limit, however 210 Bq/kg of radioactive iodine (^{131}I) was detected and it exceeded the provisional regulation value in drinking water for infants (100 Bq/kg, cf. 300 Bq/kg for other than infants). For this reason, when the measurement result was found out on March 23, the Tokyo Waterworks issued a restriction against infant consumption of drinking in the distribution area of the Kanamachi and the Misato WPPs, both of which are in the same water system.

Since radioactive iodine was detected in purified water, it was necessary to effectively remove radioactive iodine in the water purification treatment. Although it had been reported that powdered activated carbon (PAC) was effective for the removal of radioactive iodine, a specific processing condition for effective removal in actual water purification treatment were not clear. Therefore, the Tokyo Waterworks immediately started the investigation concerning the removal of radioactive iodine and clarified the effectiveness of the combined use of PAC and pre-chlorination for removal of radioactive iodine, and an optimum treatment condition. The Tokyo Waterworks applied the optimum treatment condition for radioactive iodine which was clarified from the survey results, to actual WPPs promptly from the end of March 2011 to the beginning of June.

In addition, due to the detection of radioactive iodine, intake restrictions were applied to infants for a short period of time, the interest of residents in radioactive materials in tap water had increased significantly. Therefore, the Tokyo Waterworks started continuous daily monitoring of radioactive materials in tap water and prompt release of the results. Afterwards, even after radioactive materials were not detected in tap water, monitoring of radioactive materials and publication of the results has been continued. Thus, safety of tap water can be confirmed and residents can use the tap water without anxiety.

In this article, we will report the various response measures to radioactive materials taken by the Tokyo Waterworks, and introduce safety of tap water in Tokyo.

Start of monitoring of radioactive materials and detection of radioactive iodine

At the time of the earthquake disaster, the Tokyo Waterworks did not possess analytical instruments that could measure radionuclides separately. Therefore, in response to the occurrence of the nuclear disaster, measurement of total alpha and beta radioactivity was started from March 15. To this end, existing total alpha and beta radiation measuring instruments were used. In this measurement, raw and purified water of representative WPPs are measured. There were no results exceeding the guidance level (0.5 and 1 Bq/kg for total alpha and beta radioactivity, respectively) of the World Health Organization (WHO) drinking water quality guidelines (3rd Edition) and the results were the same level as the past measurement.

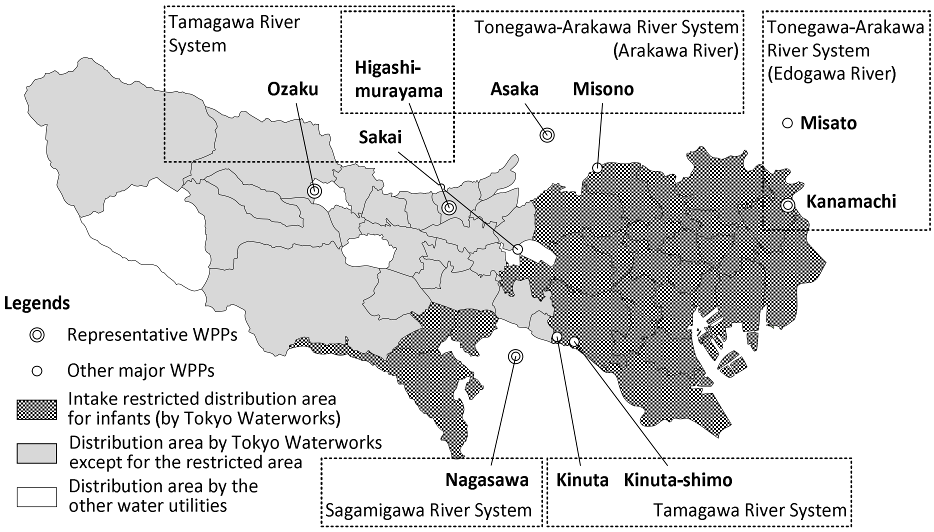

Figure 1 Water distribution area of the Bureau of Waterworks, Tokyo Metropolitan Government, and representative water purification plants of each water system and intake restriction area of infants.

Afterwards, in consideration of the influence of the first rainfall (March 21) after the occurrence of the nuclear disaster, from March 22, by commissioning to other research institutes, measurement of radioactive iodine and cesium of purified water of Kanamachi, Asaka and Ozaku WPPs started (see Figure 1). Figure 2 shows the results of purified water from the start of measurement to April 8. Radioactive iodine in the purified water, which was collected on March 22 at Kanamachi WPP, was 210 Bq/kg and it exceeded the provisional regulation value for infants (100 Bq/kg). Therefore, the Tokyo Waterworks immediately announced this result and a restriction of the intake of infants to areas where purified water of the Kanamachi WPP was distributed. In addition, as Tokyo Metropolitan Government, 240,000 bottles of bottled water (500mL) were reserved for infants and distributed at the ward offices and others in the intake restricted area. Meanwhile, the Tokyo Waterworks decided to announce the measurement results of purified water on the day of measurement for the time being.

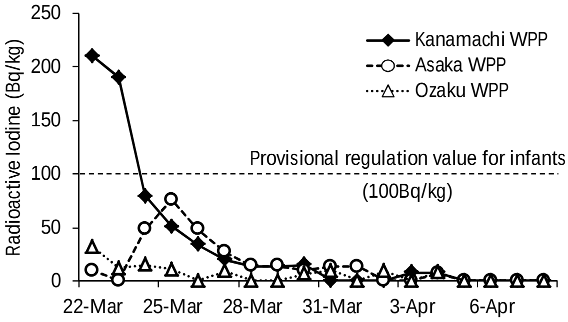

Figure 2 Radioactive iodine (^{131}I) in purified water of representative WPPs immediately after the nuclear disaster (2011)

On March 24, the Tokyo Waterworks confirmed that radioactive iodine in the purified water of Kanamachi WPP had fallen below the provisional regulation value for infant and lifted the intake restriction. In addition, because other WPPs were located farther from the Fukushima Nuclear Plant than Kanamachi, radioactive iodine in their purified water did not exceed the provisional regulation value. The highest detected value of radioactive iodine in purified water in the other WPPs was 76 Bq/kg on March 25 at the Asaka WPP, which intakes raw water from the Arakawa River, and approximately 27 km far from Kanamachi.

Survey on removal of radioactive materials

Radioactive iodine With regard to radioactive iodine, it was reported that the high removal rate was obtained by PAC (Lettinga, 1972; Summers et al. , 1988; Honma et al. , 1988; Smith et al., 2001) and the removal rate was increased by a combined use of chlorination (Lettinga, 1972; Summers et al. , 1988). Following these findings, the Ministry of Health, Labor and Welfare (MHLW) announced notice to water utilities to take measures against radioactive iodine by PAC on March 19. However, it was also reported that if the radioactive iodine present form was IO_3^-, the removal rate with PAC was low (Lettinga, 1972). Therefore, at the time when radioactive iodine was detected, it was not clear how to set the chlorination dose and the PAC injection rate in the actual treatment to remove radioactive iodine effectively.

In response to the detection of radioactive iodine in purified water, the Tokyo Waterworks investigated the removal of radioactive iodine promptly. In the survey, table experiments (jar test) were carried out on removal by treatment with PAC alone, by combined use of PAC and pre-chlorination (chlorine injection ahead of coagulation). At that time, it was difficult to measure radioactive iodine for experiments, thus the Tokyo Waterworks used potassium iodide (KI) or potassium iodate (KIO_3) containing non-radioactive iodine which was the same chemical property as radioactive iodine. Experiments were conducted by sample water, which was prepared by addition of KI or KIO_3 to the raw water of the Misato WPP which intakes raw water from the same Edogawa River as Kanamachi. In addition, to verify the obtained results, an additional experiment was carried out by using adjusted water obtained from mixing rainwater containing radioactive iodine (^{131}I) into the raw water of Misato WPP. Furthermore, the effectiveness of measures to remove radioactive iodine at the WPPs was verified by measurement of nonradioactive iodine in the raw and process water. Kitada et al. (2015) reported details of these experimental methods.

When the coagulant was added only after contacting PAC (40 mg/L) for 30 minutes using raw water to which non-radioactive I^- or IO_3^- was added, the removal rates of I^- and IO_3^- were low (both approximately 6%). Then, with respect to the removal in the combined use of PAC and pre-chlorination, the removal rate of I^- at the chlorine dose of 0 to 3 mg/L is shown in Figure 3 (left). Even though I^- could not be sufficiently removed when the chlorine dose was 2 mg/L or more, when the chlorine dose was 0.5 or 1.0 mg/L, approximately 20 to 30% of iodine could be removed. Based on these results, experiments were carried out on the influence of the PAC injection rate with the chlorine dose of 0.5 mg/L. Figure 3 (right) shows both the result of adding nonradioactive iodine and the verification result using adjusted raw water which contains radioactive iodine. It was found that 40 to 60% of I^- could be removed by combined PAC (at 20 to 30 mg/L) and pre-chlorination (at 0.5 mg/L). In addition, nearly the same removal was obtained for radioactive iodine. Consequently, to effectively remove radioactive iodine in water purification treatment, it was found that PAC injection should be combined with pre-chlorination at a dose of 0.5 to 1.0 mg/L.

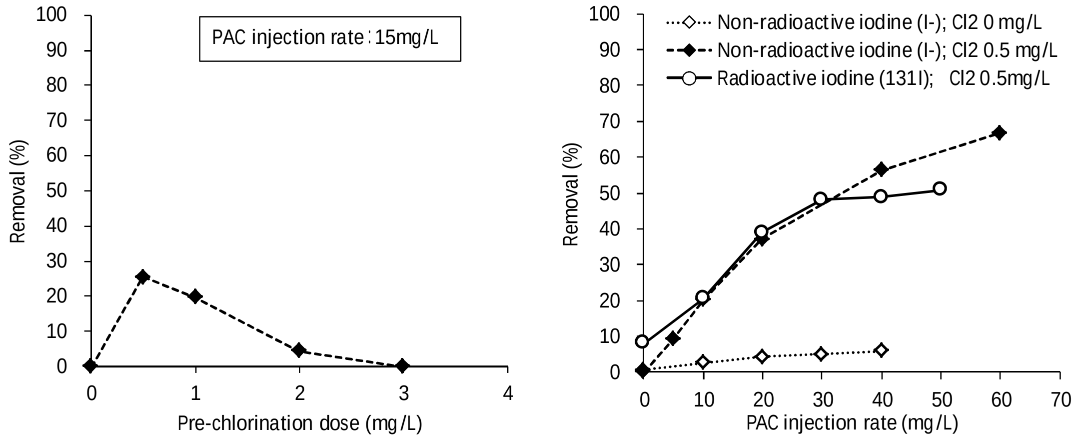

Figure 3 (Left) Relationship between pre-chlorination dose and removal of nonradioactive iodine (iodide ion: I^-) with PAC/pre-chlorination combination, (right) Relationship between PAC dose and removal of nonradioactive iodine (I^-) and radioactive iodine (^{131}I) with PAC/pre-chlorine combination

Figure 4 shows the removal of nonradioactive iodine in settled and purified water by combination of PAC and pre-chlorination in actual WPPs. It was confirmed that the combined use of PAC and pre-chlorination (dose: 1.0 mg/L) was effective as in the table experiment.

Radioactive cesium Radioactive cesium was not detected in purified water and was hardly detected from raw water. However, as described later, it was detected at a high concentration in the dehydrated sludge of the WPPs. It was reported that radioactive cesium was highly adsorptive to soil (IAEA 2010), and it seemed that it existed mainly adsorbed to the soil in the river water. For this reason, the MHLW announced that for radioactive cesium, a

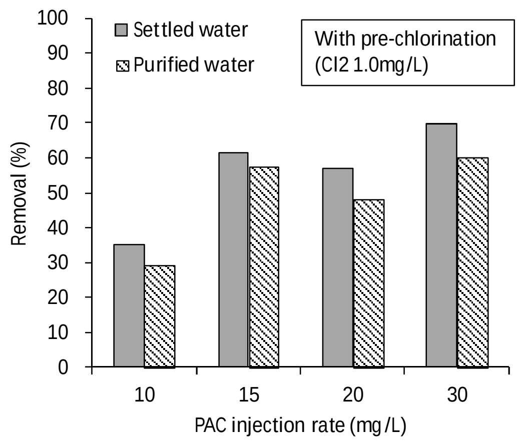

Figure 4 Relationship between pre-chlorination dose and removal rate of non-radioactive iodine (total iodine) in settled and purified water

high removal rate can be obtained by removal of turbidity in water purification treatment, however at the time, the information on removal of radioactive cesium was insufficient. Therefore, the behaviour of radioactive cesium in water purification treatment was estimated by measuring non-radioactive cesium which has the same chemical property. In addition, the removal of radioactive cesium by coagulation and sedimentation was verified, using turbid water in actual WPPs by measuring radioactive cesium. Kitada et al. (2015) reported details of the experiment.

Figure 5 shows the concentration of nonradioactive cesium in coagulation and sedimentation at the actual WPPs. It was found that the cesium concentration was significantly lower in the settled water than in the raw water, and most of the cesium in the raw water was removed by coagulation and sedimentation. Therefore, it was considered that most of the cesium in raw water is adsorbed to suspended particle. In addition, the verification result showed that the removal rate of radioactive cesium by coagulation and sedimentation at high turbidity was 93%. Consequently, it was confirmed that radioactive cesium can be effectively removed by coagulation and sedimentation. Radioactive cesium has never been detected in purified water, so it was thought that nearly all radioactive cesium could be removed by thoroughly removing turbidity even at high turbidity.

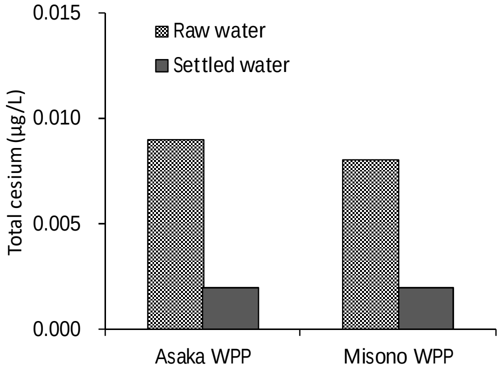

Figure 5 Nonradioactive cesium (total cesium) concentration of raw water and settled water

Response to radioactive materials in water purification plants

The Tokyo Waterworks had strengthened the injection of PAC since March 21, 2011 when there was the first rainfall after the nuclear disaster. Since radioactive iodine was detected in the purified water of the Kanamachi WPP which was collected on March 22, further injection reinforcement was done from March 23. When the above-mentioned survey results became clear since March 28, combined use of PAC and pre-chlorination was started at the Kanamachi and the Misato WPPs. For the other major WPPs, the Tokyo Waterworks started using a combination of PAC and pre-chlorination from March 30. Combined use of PAC and pre-chlorination was carried out at all WPPs during rainfall until June 8, 2011, because there was concern about increase of radioactive iodine. As a result, radioactive iodine in purified water has not been detected since April 5, 2011.

In addition, the results of this experiment promptly provided to the MHLW, Japan Water Supply Association, related water utilities, and others. It was adopted as a radioactive iodine countermeasure of the MHLW which published in June 2011.

Monitoring of radioactive materials and publication of results

Since it was widely reported that radioactive iodine was detected and exceeded the provisional regulation value for infant in tap water, the inquiries from the citizens to the department in charge have drastically increased in accordance with Figure 6. Afterwards, radioactive iodine in purified water gradually declined, and had fallen below detection limit in either Kanamachi or Asaka WPPs in April 4. However, the attention from the citizens was high and the inquiries were continued. With regard to raw water, it was not detected after detection at the Kanamachi WPP on April 13. As a result, the detection of radioactive iodine in raw or tap water settled in approximately three weeks from the disaster, and safety of tap water was confirmed. Regarding radioactive cesium, it was sometimes detected (up to 4 Bq/kg) in raw water until September 4, 2011, however it has never detected in purified water at all WPPs.

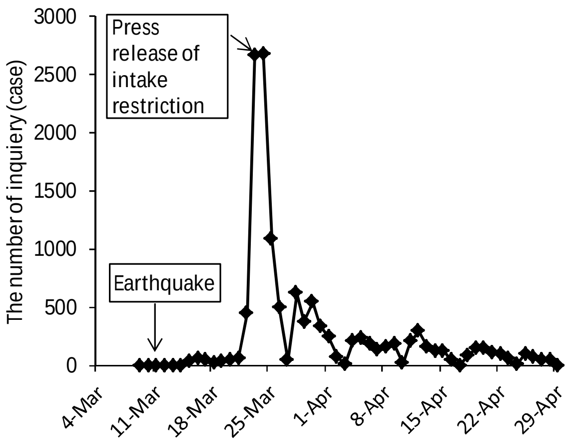

Figure 6 The number of inquiries to radioactive substances to customer centers after nuclear disaster (2011)

Even after radioactive iodine was no longer detected, anxiety of radioactive materials in tap water was not solved easily, and inquiries were continued. For this reason, to confirm the safety of tap water and relieve citizen's concerns, the Tokyo Waterworks acquired radioactive material inspection devices (germanium semiconductor detector) and constructed a monitoring system. The Tokyo Waterworks has been conducting daily inspections on purified water at representative WPPs and the results are publicized on the website on the following day in principle (see Table 1). Regarding the other major WPPs and ones that uses groundwater as the raw water, the Tokyo Waterworks regularly checks the radioactive materials and publish the results.

Table 1 Frequency of radioactive material inspection on purified water at the WPPs of the Tokyo Waterworks.

	FY 2011	FY 2012	From FY 2013
Representative WPPs in each water system (5 plants)	Daily	Daily	Daily
Other major WPPs (5 plants)	Weekly	Daily	Monthly
WPPs with raw water as surface water, shallow groundwater in Tama area	Monthly	Weekly	Monthly
WPPs with raw water as deep groundwater in Tama area	Monthly	Monthly	Once in three months

Monitoring and response to radioactive materials in dehydrated sludge

As radioactive iodine was detected in the purified water of the Kanamachi WPP collected on March 22, there was concern about the influence on the dehydrated sludge. Therefore, the Tokyo Waterworks started analyzing radioactive materials in the dehydrated sludge from March 28. As a result, radioactive iodine of 88,400 Bq/kg and radioactive cesium of 14,650 Bq/kg were detected in the sludge sampled in March 28. As this result became clear, the Tokyo Waterworks decided not to utilize the sludge. Afterwards, radioactive materials were measured every two weeks. In April 4, radioactive iodine in the sludge at the same WPP declined sharply to 11,200 Bq/kg, radioactive cesium 5,430 Bq/kg. In particular, since radioactive iodine has a short half-life of eight days, radioactive iodine in the sludge has decreased below 100 Bq/kg on May 17, and it has not been detected since June 15, 2011.

Meanwhile, radioactive cesium with a long half-life has continued to be detected after June 2011. Figure 7 shows the detection status of radioactive cesium contained in the dehydrated sludge at Kanamachi WPP. Radioactive cesium was detected at a high concentration of 14,650 Bq/kg on March 28, 2011, and then declined rapidly. Thereafter, it decreased below 1,000 Bq/kg since September, 2011. Currently it is mostly less than 100 Bq/kg.

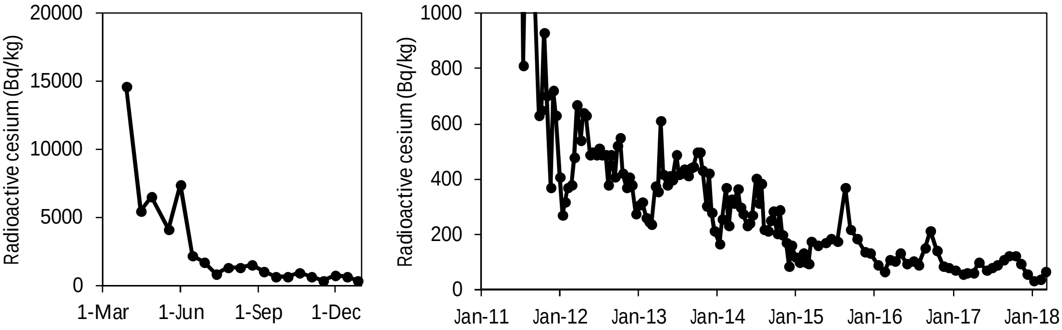

Figure 7 Trends of radioactive cesium concentration in the dehydrated sludge at Kanamachi WPP from March, 2011. (Left) From March, 2011 to December, 2011 (full scale: 20,000 Bq/kg); (Right) From March, 2011 to present (full scale: 1,000 Bq/kg).

Regarding the handling of dehydrated sludge containing radioactive cesium, the MHLW announced a provisional guideline on June, 2011. MHLW was regarded sludge which contains less than 8,000 Bq/kg of radioactive cesium to be landfill, and sludge contains less than 100 Bq/kg to be reutilized to cement and others. Also, in March, 2013, a permanent guideline was published. Under this guideline, dehydrated sludge contains less than 400 Bq/kg of radioactive cesium become able to be utilized for horticultural soil and less than 200 Bq/kg for ground soil. Therefore, the Tokyo Waterworks restarted utilization of the sludge of the Ozaku WPP to granular modified soil because radioactive cesium was stably below 100 Bq/kg from October 17, 2012. Furthermore, utilization of the dehydrated sludge meeting the permanent guideline value was resumed from March 26, 2013.

Radioactive cesium in raw water has not been detected since June 2011, but the detection in the dehydrated sludge continued thereafter. Therefore, it seemed that there was a trace amount of radioactive cesium in the raw water and removed by the water purification treatment and concentrated in the dehydrated sludge. To confirm this speculation, concentration of radioactive cesium in raw water was estimated from measurement results of the sludge (Kitada et al. (2015) reported detailed estimation method). Table 2 shows estimation results of radioactive cesium concentration in raw water at Kanamachi WPP. The maximum value estimated in the raw water was 1.2 Bq/kg from the sludge sampled on March 28, 2011 immediately after the nuclear disaster. This value was approximately one tenth as compared with the guidance level (10 Bq/kg) of the WHO drinking water quality guideline and the regulated value (water quality control target value: 10

Table 2 Radioactive cesium concentration in dehydrated sludge and estimated concentration in raw water at Kanamachi WPP

Collection date (year)	Collection date (date)	Dehydrated sludge (Measured) (Bq/kg)	Raw water (Estimated) (Bq/kg)
2011	Mar 28	14,650	1.20
	Apr 14	5,430	0.55
	Apr 27	6,570	0.52
	May 17	4,100	0.36
	Jun 1	7,400	0.48
	:	:	:
2013	Mar 11, 14, 18, 21*	375	0.02
	Mar 25, 28, Apr 1, 4*	357	0.04

* A total of 4 samples are collected on each day of collection, and measurements are made on the mixed sample with the same weight.

Bq/kg) revised in April 2012. From this result, it was considered that the radioactive cesium in the raw water was concentrated on average about 10,000 times to dehydrated sludge. Therefore, it can be said that the decrease of the radioactive cesium in the sludge shown in Figure 7 indicates the decrease of radioactive cesium adsorbed to the sediment of the river.

Conclusion

In response to the nuclear disaster of the Fukushima Daiichi Nuclear Power Station occurred with the Great East Japan Earthquake on March 11, 2011, the Tokyo Waterworks started monitoring of radioactive materials in purified water from March 22. As a result, radioactive iodine exceeded the provisional regulation value for infant intake. Therefore, the Tokyo Waterworks investigated the effective removal method of radioactive iodine promptly and clarified that radioactive iodine can be effectively removed with PAC by combining with pre-chlorination at a dose of 0.5 to 1.0 mg/L. Based on this result, an optimal condition for removal of radioactive iodine was applied at WPPs around one week after the detection. This information was provided promptly to the national government and related organizations and the method contributed to the formulation of national radioactive material countermeasures. In addition, the Tokyo Waterworks investigated radioactive cesium and clarified that radioactive cesium can be drastically reduced by coagulation and sedimentation.

Due to high customer concern for radioactive materials in tap water, the Tokyo Waterworks has been measuring radioactive materials in purified water and publishing the results every day since the time of nuclear disaster occurrence. In purified water, radioactive iodine has not been detected since April 5, 2011 and radioactive cesium has not been detected since the beginning of the measurement.

Radioactive iodine in the dehydrated sludge was high immediately after the nuclear disaster. However, it has not been detected since June 15, 2011. Although radioactive cesium is detected even now, its value is greatly decreased, and is mostly less than 100 Bq/kg even in the Kanamachi WPP where initially detected at a high concentration exceeding 10,000 Bq/kg. In addition, the dehydrated sludge is landfilled or utilized according to the guideline prescribed by the government.

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