Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2017.20934

91 (2017) 192–197
October

Demonstration testing of a system for the high-speed monitoring of the radioactive concentration of wastewater in situ

Hisatomo Fukui[a,*], Hiroshi Oota[a], Hiroyuki Hirano[b], Toshihisa Hatano[c], Hideyuki Saito[d]

[a]*Kajima Corporation, 5-11 Akasaka 6-Chome, Minato-ku, Tokyo 107-8348, Japan, Tel. +81-3-5544-0808; email: hisatomo@kajima.com (H. Fukui), Tel. +81-3-5544-0866; email: hiroshi-oota@kajima.com (H. Oota)*
[b]*Kajima Technical Research Institute, 19-1 Tobitakyu 2-Chome, Chofu-shi, Tokyo 182-0036, Japan, Tel. +81-42-489-6257; email: hiranohi@kajima.com*
[c]*National Institutes for Quantum and Radiological Science and Technology, 1233 Watanukimachi, Takasaki-shi, Gunma 370-1292, Japan, Tel. +81-27-346-9204; email: hatano.toshihisa@qst.go.jp*
[d]*Nikkin Flux Inc., 1651 Kamekubo, Fujimino-shi, Saitama 356-0051, Japan, Tel. +81-49-262-1234; email: hideyuki_saito@nikkin-flux.co.jp*

Received 14 December 2016; Accepted 16 May 2017

ABSTRACT

While the NaI(Tl) scintillation detector lacks a high energy resolution, it has the merit of high sensitivity. The NaI(Tl) scintillation detector is considered suitable for the high-speed measurement of cesium-134 and cesium-137 together. We developed "Cesimoni-water" as a system for the in situ monitoring of the radioactive concentration of wastewater by combining an NaI(Tl) scintillation detector with a water shield technique for shielding out external natural background radiation. In this paper, we describe the Cesimoni-water system and demonstration tests carried out with the system using simulated wastewater.

Keywords: Radioactive concentration; Radiocesium; In situ monitoring; NaI(Tl) scintillator

1. Introduction

The radioactive fallout from the Fukushima Daiichi nuclear disaster caused by the Great East Japan Earthquake was scattered onto fields over a wide region of northern Japan. Several studies have reported inventory analyses of the released radionuclides [1–9]. Radioactive decontamination work is now underway in Fukushima and interim storage facilities to store the contaminated soil, leaves, and debris are currently being planned. The wastewater discharged from the radioactive decontamination work and interim storage facilities is likely to contain radioactive materials. Wastewater containing radiocesium is generally treated using some sort of radiocesium-removal system in addition to a conventional wastewater treatment process such as a coagulating sedimentation system. After the wastewater is treated, it is transferred to an effluent tank for temporary storage and analyzing. Approximately 2 L of the water is collected from the effluent tank and analyzed by a germanium semiconductor detector. The treated water in the tank can be discharged to a public water area if the analytical results satisfy the regulations [10]. The germanium semiconductor detector has a high energy resolution but lacks high sensitivity and the required apparatus unsuitable for in situ monitoring, such as equipment for liquid nitrogen cooling and lead shield [11,12]. It also takes considerable time to perform its measurements, which makes it unsuitable for the high-speed monitoring of large amounts of wastewater. Meanwhile, the NaI(Tl) scintillation detector lacks a high energy resolution but has the merit of high sensitivity. The NaI(Tl) scintillation detector is considered suitable for the high-speed monitoring

* Corresponding author.

Presented at the 13th IWA Specialized Conference on Small Water and Wastewater Systems & 5th IWA Specialized Conference on Resources-Oriented Sanitation, 14–16 September, 2016, Athens, Greece.

of cesium-134 and cesium-137 together, as the absorption spectra of cesium-134 and cesium-137 overlap [13–17].

We developed "Cesimoni-water," a system for the monitoring of the radioactive concentration of wastewater in situ using an NaI(Tl) scintillation detector in combination with a water shield to shield out external natural background radiation. We describe the Cesimoni-water system and demonstration tests we performed on it using simulated wastewater to assess its feasibility for the measurement of radioactivity in situ.

2. Materials and methods

2.1. Outline of the Cesimoni-water system

2.1.1. Tank configuration and stirring system

The Cesimoni-water system (Fig. 1) consists of three components: a water tank, a stirring system for analysis water, and a gamma ray detection system.

Several studies have identified relatively higher concentrations of radiocesium in the suspended solids in wastewater [18–20]. The accurate measurement of the radioactive concentration, therefore, requires that the wastewater containing the suspended solids be sufficiently stirred. Figs. 2 and 3, respectively, show a top view and cross-section view of the Cesimoni-water system. The water sample to be analyzed is poured into a cylindrical tank with a capacity of 3.5 m^3 (Cabin 1) encased within a larger cubic tank (Cabin 2). The cylindrical tank is equipped with a small inner cylinder, a gamma ray detector at the center, and two submersible stirring pumps. The top pump with an electric capacity of 3.0 kW (Model U-244KA, Sakuragawa Pump Mfg. Co., Ltd., Japan) and bottom pump of 2.2 kW (Model U-233KA, Sakuragawa Pump Mfg. Co., Ltd., Japan) are, respectively, set below the surface of the water in the cylindrical tank and on the bottom of the tank. Both pumps discharge the water into the space between the outer and inner cylinders, creating a swirl flow that efficiently circulates the suspended solids. The cubic tank, meanwhile, encases the cylindrical tank. Approximately 8 m^3 of freshwater is stored in Cabin 2 between the cubic tank and cylindrical tank as shielding water to eliminate external natural background radiation. The dimensions of the cubic tank and cylindrical tank are 2.7 mW × 2.2 mD × 2.6 mH and 1.4 mϕ × 2.5 mH, respectively.

The Cesimoni-water measurement of one batch is divided into three phases: the analysis water influent process, the stirring and measurement process, and the effluent process. The gamma ray measurement takes 10 min and the overall operational time of a batch, including influent and effluent times, is 18 min.

2.1.2. Gamma ray detection system

Fig. 4 shows a schematic diagram of the gamma ray detection system. The system is composed of an underwater gamma ray detector (Fig. 5) and a control panel set beside the cubic water tank. The gamma ray detector is composed of an NaI(Tl) scintillator, a photomultiplier tube (Model R877, Hamamatsu Photonics K.K., Japan), a thermocouple thermometer, a high voltage power supply (Nikkin Flux Inc., Japan), and an

Fig. 1. Photo of the Cesimoni-water system.

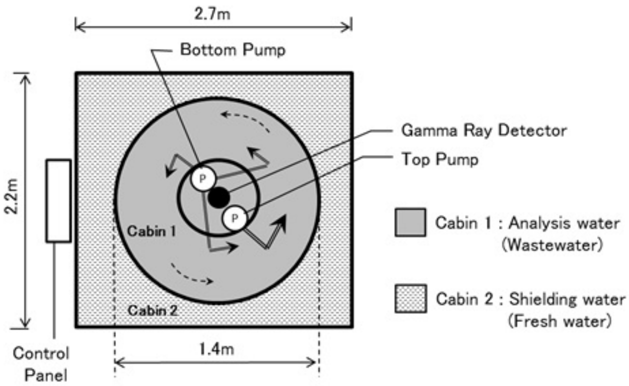

Fig. 2. Top view of the Cesimoni-water system.

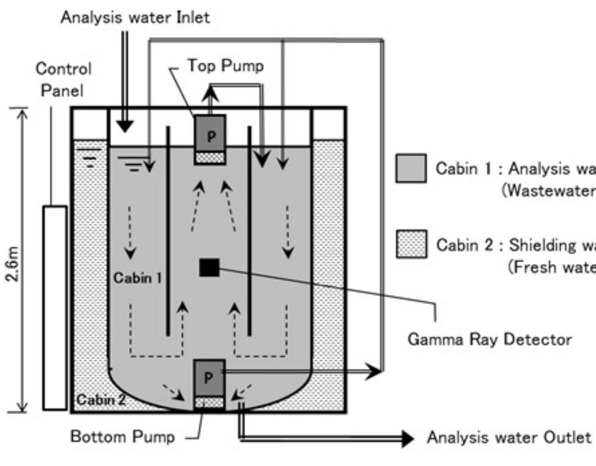

Fig. 3. Vertical cross-section view of the Cesimoni-water system.

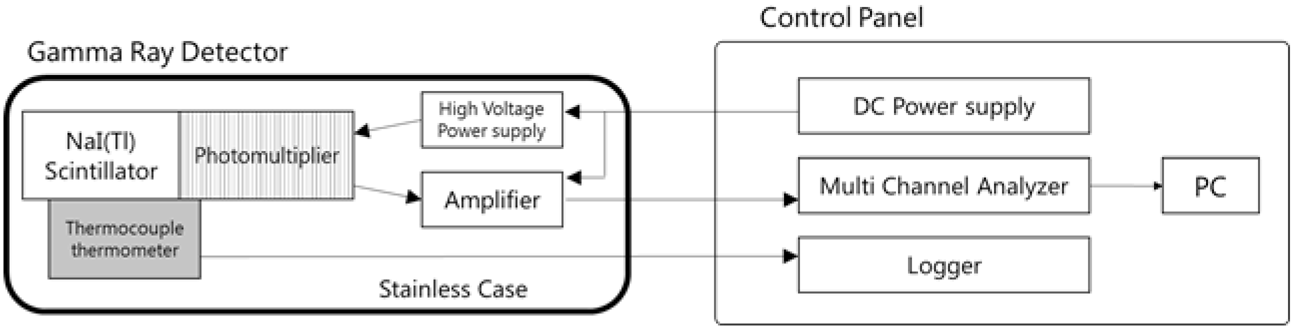

Fig. 4. Schematic diagram of the gamma ray detection system.

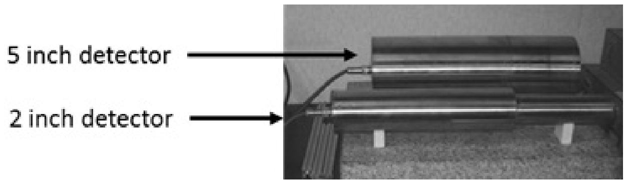

Fig. 5. Photo of the gamma ray detectors.

amplifier (Nikkin Flux Inc., Japan), all housed within a stainless case. A 2-inch-diameter or 5-inch-diameter scintillation crystal manufactured by Hilger Crystals Ltd. (UK) was used as the NaI(Tl) scintillator. The control panel consists of a DC power supply, a multichannel analyzer (Model APG7300A, Techno AP Co., Ltd., Japan), a logger and a recording PC.

2.2. Measurement tests of radioactive concentration

2.2.1. Water shield test for the external natural background radiation

Natural background radiation affects the gamma ray counting by the NaI(Tl) scintillation detector. The Cesimoni-water system adopts the water shield technique to shield out the external background radiation [21,22]. The NaI(Tl) scintillation detector is surrounded by at least 1 m of analysis water and shielding water.

Before analyzing the radioactively contaminated water, we researched the effect of the shielding on external natural background using freshwater and a point radiation source. A cesium-137 point radiation source radiating 1 MBq was set at the center of the side wall of the cubic tank. The gamma ray counting rates were measured under the two conditions shown in Table 1, namely, with Cabins 1 and 2 respectively empty (Condition 1) or respectively filled (Condition 2).

2.2.2. Radiopotassium solution test

Potassium-40 accounts for 0.0117% of natural potassium, and 1 g potassium has a radioactive concentration of 30.4 Bq [23]. Potassium chloride solutions at 0.60, 3.0, 6.0, and 13.5 g/L concentrations, the theoretical equivalents to radioactive concentrations of 1, 5, 10, and 22.5 Bq/L, were prepared and measured by the Cesimoni-water system. The measurement took 10 min. Fig. 6 shows the pulse height spectrum for the 10 Bq/L radiopotassium solution measured by the 5-inch scintillation detector. A potassium-40 photopeak with

Table 1
The conditions of Cabins 1 and 2 during NaI(Tl) scintillation testing (filled vs. empty)

	Cabin 1	Cabin 2
Condition 1	Empty	Empty
Condition 2	Filled by freshwater	Filled by freshwater

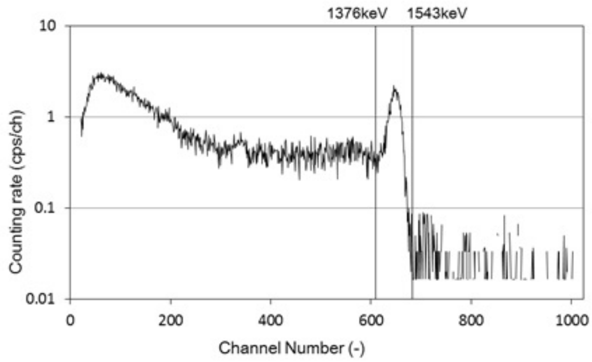

Fig. 6. The pulse height spectrum of the radiopotassium solution.

1,461 keV was observed. The gamma ray counting rate for radiopotassium was estimated by integrating the value from 1,376 to 1,543 keV.

2.2.3. Artificial radiocesium solution test

Artificial radiocesium solutions were measured by the Cesimoni-water system. The solutions were prepared by diluting a radioactively contaminated dry sludge whose radioactive concentration had been preliminarily measured with a high purity germanium semiconductor detector (Model GC2020-7500SL, Canberra Industries Inc., USA). The dry sludge was collected from the wastewater treatment facility during the radioactive decontamination work in Fukushima, Japan. The Cesimoni-water system measured the solutions in a period of 10 min. Fig. 7 shows the pulse height spectrum for the 10 Bq/L radiocesium solution measured by the 5-inch scintillation detector. The gamma ray counting rate for radiocesium, which reflects the total radioactivity of cesium-134 and cesium-137, was estimated by integrating the value from 540 to 830 keV.

2.2.4. Wastewater test

As the final investigation, a demonstration measurement test of the Cesimoni-water system was conducted using actual wastewater collected from radioactive decontamination work. The repeatability of the Cesimoni-water system was confirmed by repeating measurements of the same wastewater.

3. Results and discussion

3.1. Result of the water shield test

Fig. 8 plots the change in the gamma ray counting rate under Conditions 1 and 2. The average gamma ray counting fell from 65.7 cps under Condition 1 to 0.161 cps under Condition 2. The results confirmed that a water shield of approximately 1 m can reduce external radiation by more than 400-fold compared with an unshielded condition.

3.2. Result of the radiopotassium solution test

Figs. 9 and 10 show the relations between the radioactive concentration of potassium-40 and the gamma ray counting rate measured by the 2-inch-diameter and 5-inch-diameter scintillation detectors, respectively. Both regression curves can be regarded as linear in the 1–22.5 Bq/L range. The results suggest that Cesimoni-water is a feasible system for radiopotassium measurement.

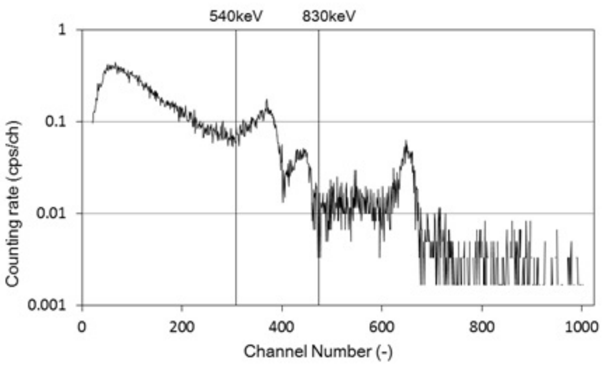

Fig. 7. The pulse height spectrum of the radiocesium solution.

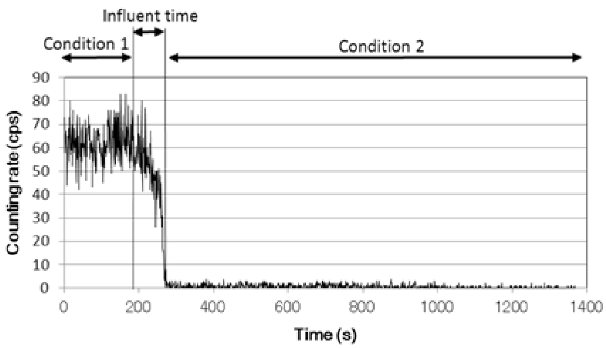

Fig. 8. Result of the water shield test.

3.3. Result of the artificial radiocesium solution test

Figs. 11 and 12 show the relation between the radioactive concentration of radiocesium, which is the total of cesium-134 and cesium-137, and the gamma ray counting rate measured by the 2-inch and 5-inch scintillation detectors, respectively. Both regression curves can be regarded as linear. The limit of detection (LOD) of the gamma ray counting rate was calculated by Eq. (1) [24]. The LODs of the Cesimoni-water system

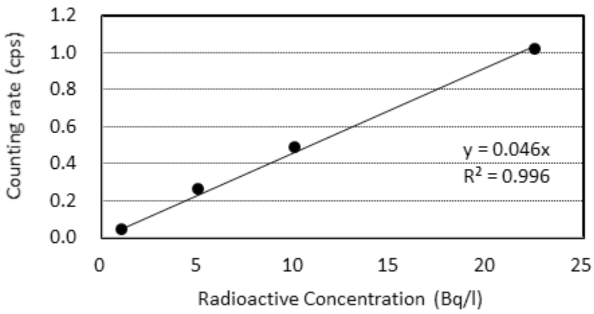

Fig. 9. Relation between the radioactive concentration of potassium-40 and the gamma ray counting rate measured by the 2-inch scintillation detector.

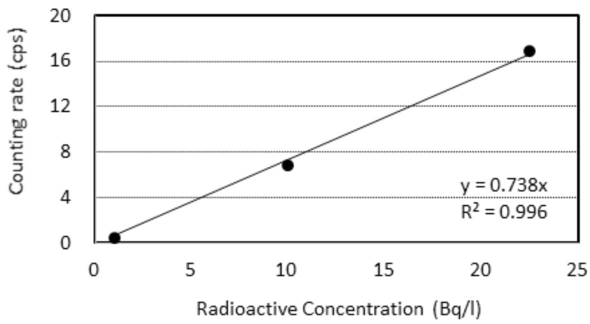

Fig. 10. Relation between the radioactive concentration of potassium-40 and the gamma ray counting rate measured by the 5-inch scintillation detector.

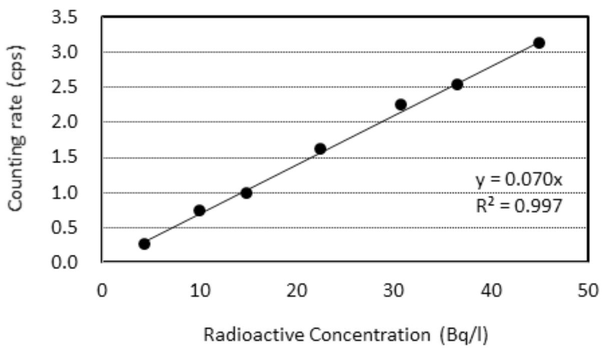

Fig. 11. Relation between the radioactive concentration of radiocesium and the gamma ray counting rate measured by the 2-inch scintillation detector.

equipped with the 2-inch and 5-inch scintillation detectors were 1.94 and 0.34 Bq/L, respectively, for radiocesium wastewater. The measurement error by the Cesimoni-water system equipped with the 5-inch scintillation detector was within 2% in the 0.34–45 Bq/L range.

$$C_l = \frac{3}{2}\left\{\frac{3}{T_s} + \sqrt{\left(\frac{3}{T_s}\right)^2 + 4C_b\left(\frac{1}{T_s} + \frac{1}{T_b}\right)}\right\} \quad (1)$$

where C_l is the LOD of the gamma ray counting rate (cps); C_b is the background gamma ray counting rate (cps); T_s is the measurement time of the solution (s); and T_b is the measurement time of the background radioactivity (s).

3.4. Result of the wastewater test

Fig. 13 shows the findings of the repeatability test by the Cesimoni-water system. The ±3σ (standard deviation) region of the gamma ray counting rates was confirmed to be <0.69 cps which was equivalent to 1.0 Bq/L of the radioactive concentration [25]. The findings suggest that the Cesimoni-water system is feasible for the radiocesium measurement of actual wastewater.

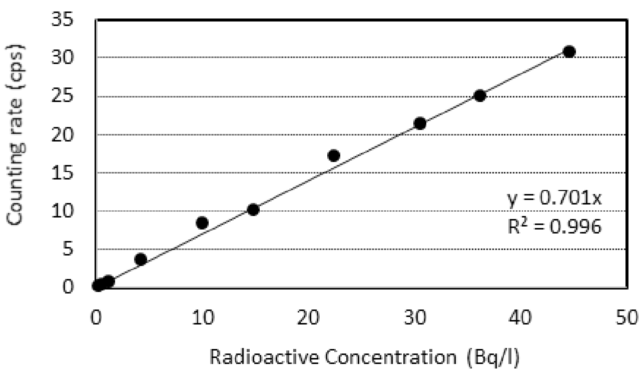

Fig. 12. Relation between the radioactive concentration of radiocesium and the gamma ray counting rate measured by the 5-inch scintillation detector.

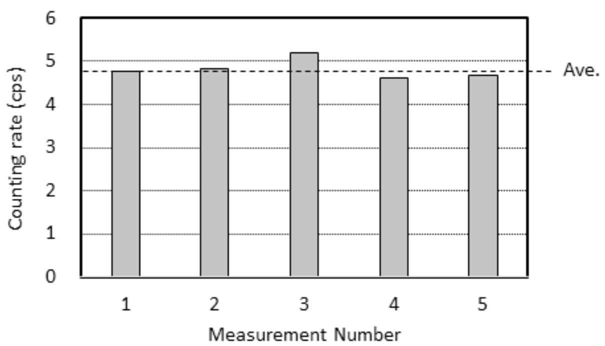

Fig. 13. Result of the repeatability test for actual wastewater.

4. Conclusion

The findings of this study suggest that the Cesimoni-water system is capable of measuring a low radioactive concentration of <10 Bq/L as the limit of quantitation at an operating rate of more than 10 m^3/h. The Cesimoni-water system appears to be suitable for use as a continuous monitoring system for radioactive effluent water. The Cesimoni-water system has good potential for adoption in interim storage facilities, sewage treatment plants, and many other wastewater treatment facilities as a convenient system for measuring radioactivity in situ.

Acknowledgments

We would like to thank Ministry of the Environment of Japan for providing a place and wastewater for our measurement tests. This development was supported by SENTAN, Japan Science and Technology Agency (JST).

References

[1] S. Ochiai, S. Nagao, M. Yamamoto, T. Itono, K. Kashiwaya, K. Fukui, H. Iida, Deposition records in lake sediments in western Japan of radioactive Cs from the Fukushima Dai-ichi nuclear power plant accident, Appl. Radiat. Isot., 81 (2013) 366–370.

[2] P. Thakur, S. Ballard, R. Nelson, An overview of Fukushima radionuclides measured in the northern hemisphere, Sci. Total Environ., 458–460 (2013) 577–613.

[3] B. Thornton, S. Ohnishi, T. Ura, N. Odano, S. Sasaki, T. Fujita, T. Watanabe, K. Nakata, T. Ono, D. Ambe, Distribution of local ^{137}Cs anomalies on the seafloor near the Fukushima Dai-ichi Nuclear Power Plant, Mar. Pollut. Bull., 74 (2013) 344–350.

[4] M. Inoue, H. Kofuji, K. Fujimoto, Y. Furusawa, K. Yoshida, S. Nagao, M. Yamamoto, Y. Hamajima, M. Minakawa, Delivery mechanism of (134)Cs and (137)Cs in seawater off the Sanriku Coast, Japan, following the Fukushima Dai-ichi NPP accident, J. Environ. Radioact., 137 (2014) 113–118.

[5] M. Yamaguchi, A. Kitamura, Y. Oda, Y. Onishi, Predicting the long-term (137)Cs distribution in Fukushima after the Fukushima Dai-ichi nuclear power plant accident: a parameter sensitivity analysis, J. Environ. Radioact., 135 (2014) 135–146.

[6] K. Tanaka, H. Iwatani, A. Sakaguchi, Q. Fan, Y. Takahashi, Size-dependent distribution of radiocesium in riverbed sediments and its relevance to the migration of radiocesium in river systems after the Fukushima Daiichi Nuclear Power Plant accident, J. Environ. Radioact., 139 (2015) 390–397.

[7] T. Kinouchi, K. Yoshimura, T. Omata, Modeling radiocesium transport from a river catchment based on a physically-based distributed hydrological and sediment erosion model, J. Environ. Radioact., 139 (2015) 407–415.

[8] W. Yu, J. He, W. Lin, Y. Li, W. Men, F. Wang, J. Huang, Distribution and risk assessment of radionuclides released by Fukushima nuclear accident at the northwest Pacific, J. Environ. Radioact., 142 (2015) 54–61.

[9] W. Men, J. He, F. Wang, Y. Wen, Y. Li, J. Huang, X. Yu, Radioactive status of seawater in the northwest Pacific more than one year after the Fukushima nuclear accident, Sci. Rep., 5 (2015) 7757.

[10] Ministry of the Environment, Guideline for Method of Measurement of Radioactive Concentration, 2013, p. 46.

[11] D. Brune, J. Dubois, S. Hellstrom, Improvements in Applied Gamma-Ray Spectrometry with Germanium Semiconductor Detector, Aktiebolaget Atomenergi, Stockholm, Sweden, 1965, AE-174.

[12] F.S. Goulding, Y. Stone, Semiconductor Radiation Detectors: basic principles and some uses of a recent tool that has revolutionized nuclear physics are described, Science, 170 (1970) 280–289.

[13] L.F. Pires, J.R. de Macedo, M.D. de Souza, O.O.S. Bacchi, K. Reichardt, Gamma-ray-computed tomography to investigate compaction on sewage-sludge-treated soil, Appl. Radiat. Isot., 59 (2003) 17–25.

[14] I.T. Muminov, A.K. Muhamedov, B.S. Osmanov, A.A. Safarov, A.N. Safarov, Application of NaI(Tl) detector for measurement of natural radionuclides and (137)Cs in environmental samples: new approach by decomposition of measured spectrum, J Environ. Radioact., 84 (2005) 321–331.
[15] C. Tsabaris, C. Bagatelas, T. Dakladas, C.T. Papadopoulos, R. Vlastou, G.T. Chronis, An autonomous in situ detection system for radioactivity measurements in the marine environment, Appl. Radiat. Isot., 66 (2008) 1419–1426.
[16] M.S. Rahman, G. Cho, B.S. Kang, Deconvolution of gamma-ray spectra obtained with NAI(Tl) detector in a water tank, Radiat. Prot. Dosim., 135 (2009) 203–210.
[17] J.A. Caffrey, K.A. Higley, A.T. Farsoni, S. Smith, S. Menn, Development and deployment of an underway radioactive cesium monitor off the Japanese coast near Fukushima Dai-ichi, J. Environ. Radioact., 111 (2012) 120–125.
[18] H. Tsuji, T. Yasutaka, Y. Kawabe, T. Onishi, T. Komai, Distribution of dissolved and particulate radiocesium concentrations along rivers and the relations between radiocesium concentration and deposition after the nuclear power plant accident in Fukushima, Water Res., 60 (2014) 15–27.
[19] N. Yoshikawa, H. Obara, M. Ogasa, S. Miyazu, N. Harada, M. Nonaka, ^{137}Cs in irrigation water and its effect on paddy fields in Japan after the Fukushima nuclear accident, Sci. Total Environ., 481 (2014) 252–259.
[20] K. Yoshimura, Y. Onda, A. Sakaguchi, M. Yamamoto, Y. Matsuura, An extensive study of the concentrations of particulate/dissolved radiocaesium derived from the Fukushima Dai-ichi Nuclear Power Plant accident in various river systems and their relationship with catchment inventory, J. Environ. Radioact., 139 (2015) 370–378.
[21] J. Shapiro, Radiation Protection: A Guide for Scientists, Regulators and Physicians, 4th ed., Harvard University Press, 2002, pp. 58–59.
[22] D. Shahbazi-Gahrouei, M. Gholami, S. Setayandeh, A review on natural background radiation, Adv. Biomed. Res., 2 (2013) 65.
[23] S.B. Samat, S. Green, A.H. Beddoe, The ^{40}K activity of one gram of potassium, Phys. Med. Biol., 42 (1997) 407–413.
[24] Ministry of Health, Labour and Welfare, Manual on the Radiation Measurement of Tap Water, 2011, p. 49 (in Japanese).
[25] J. Uhrovčík, Strategy for determination of LOD and LOQ values—some basic aspects, Talanta, 119 (2014) 178–180.