Demonstration Testing of a System for the High Speed Monitoring of the Radioactive Concentration of Wastewater In Situ

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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.

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 (Ochiai *et al.*, 2013; Thakur *et al.*, 2013; Thornton *et al.*, 2013; Inoue *et al.*, 2014; Yamaguchi *et al.*, 2014; Tanaka *et al.*, 2015; Kinouchi *et al.*, 2015; Yu *et al.*, 2015; Men *et al.*, 2015). 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 liters 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 (Ministry of the Environment, 2013). The germanium semiconductor detector has a high energy resolution but lacks high sensitivity and requires apparatus unsuitable for in situ monitoring, such as equipment for liquid nitrogen cooling and lead shield (Brune *et al.*, 1965; Goulding and Stone, 1970). 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 of cesium-134 and cesium-137 together, as the

absorption spectra of cesium-134 and cesium-137 overlap (Pires *et al.*, 2003; Muminov *et al.*, 2005; Tsabaris *et al.*, 2008; Rahman *et al.*, 2009; Caffrey *et al.*, 2012).

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.

MATERIALS AND METHODS
Outline of the Cesimoni-water[®] System
Tank configuration and stirring system.

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

Photo 1. Photo of the Cesimoni-water[®] system

Several studies have identified relatively higher concentrations of radiocesium in the suspended solids in wastewater (Tsuji *et al.*, 2014; Yoshikawa *et al.*, 2014; Yoshimura *et al.*, 2015). The accurate measurement of the radioactive concentration therefore requires that the wastewater containing the suspended solids be sufficiently stirred. Figure 1 and Figure 2 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 3.0kW (Model U-244KA, SAKURAGAWA PUMP MFG. CO., LTD., Japan) and bottom pump of 2.2kW (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 fresh water 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 $\times$ 2.2 mD $\times$ 2.6 mH and 1.4 m ϕ $\times$ 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 minutes and the overall operational time of a batch, including influent and effluent times, is 18 minutes.
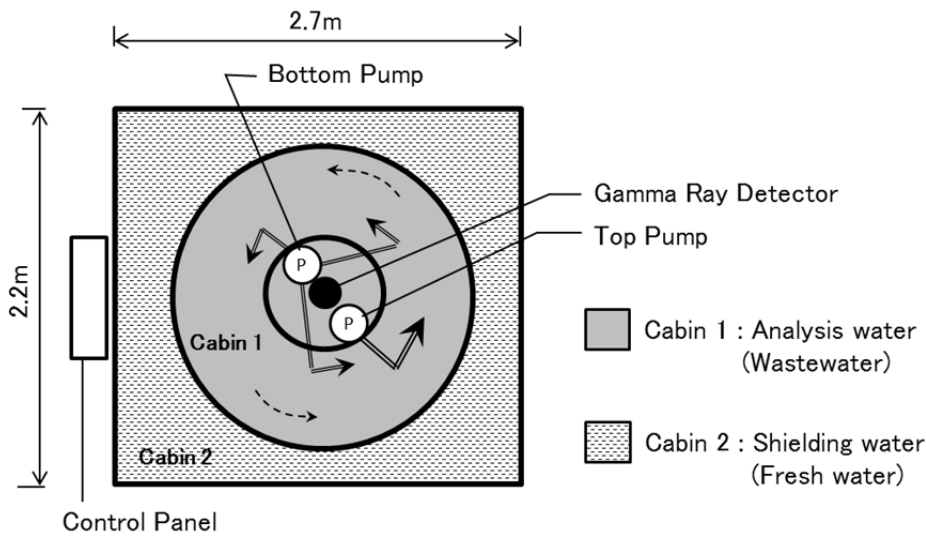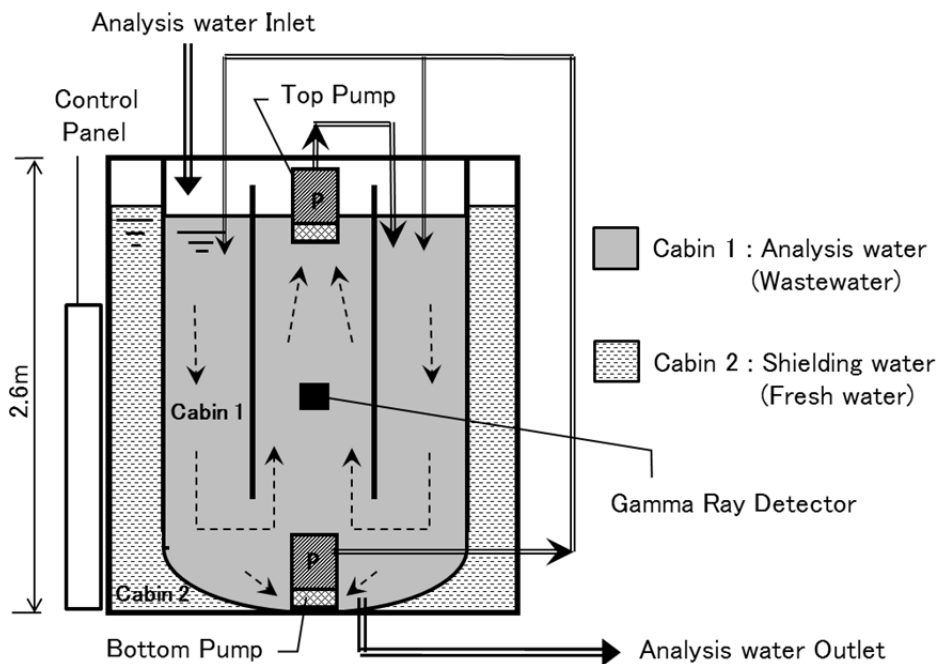

Figure 1. Top view of the Cesimoni-water® system

Figure 2. Vertical cross-section view of the Cesimoni-water® system

Gamma ray detection system

Figure 3. shows a schematic diagram of the gamma ray detection system. The system is composed of an underwater gamma ray detector (Photo 2) 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 amplifier (Nikkin Flux Inc., Japan), all housed within a stainless case. A 2-inch- or 5-inch-diameter scintillation crystal manufactured by Hilger

Crystals Ltd. was used as the NaI(Tl) scintillator. The control panel consists of a DC power supply, a multi channel analyzer (Model APG7300A, Techno AP co., Ltd., Japan), a logger and a recording PC.
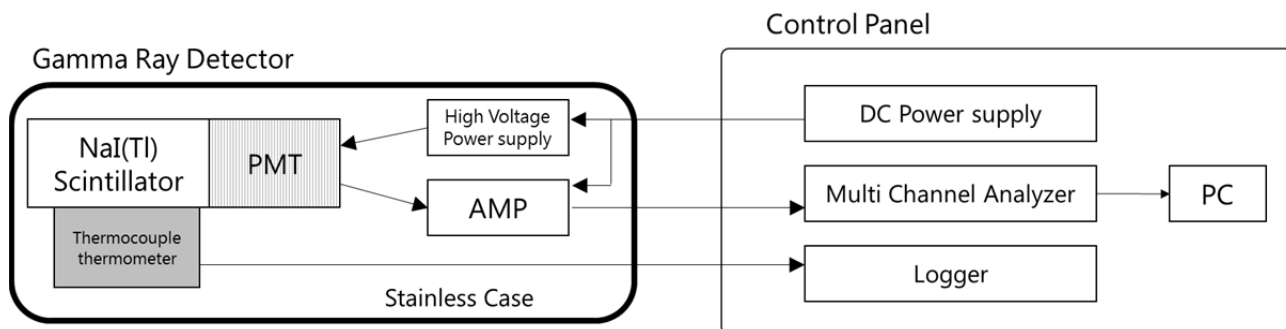

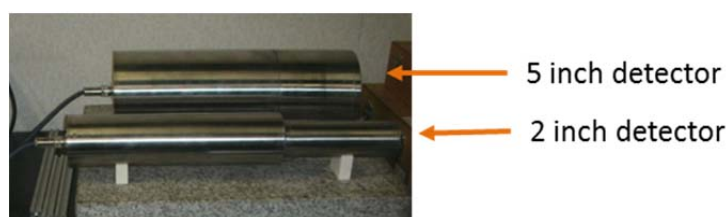Figure 3. Schematic diagram of the gamma ray detection system

Photo 2. Photograph of the gamma ray detectors

Measurement Tests of Radioactive Concentration

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 (Shapiro, 2002; Shahbazi-Gahrouei *et al.*, 2013). 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 fresh water 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).

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 Fresh water	Filled by Fresh water

Radiopotassium solution test

Potassium-40 accounts for 0.0117 % of natural potassium, and 1g-potassium has a radioactive concentration of 30.4 Bq (Samat *et al.*, 1997). 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 minutes. Figure 4 shows the pulse height spectrum for the 10 Bq/l radiopotassium solution measured by the 5-inch scintillation detector. A potassium-40 photopeak with 1461 keV was observed. The gamma ray counting rate for radiopotassium was estimated by integrating the value
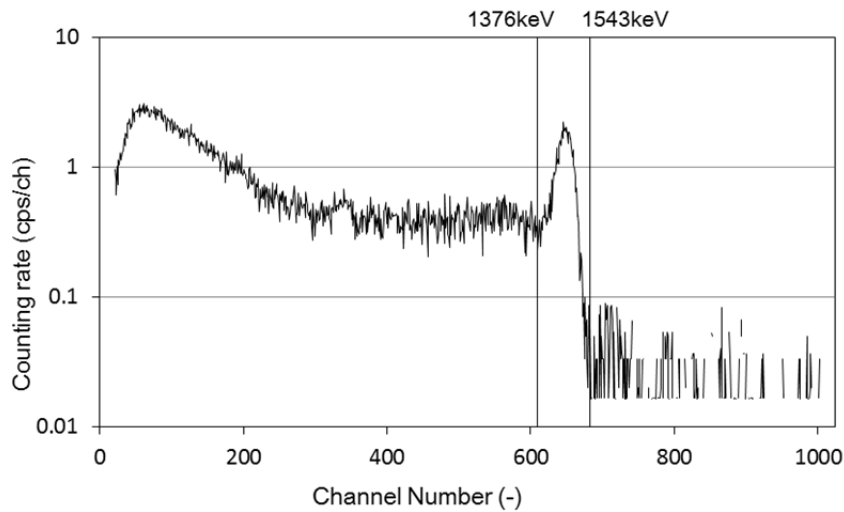
from 1376 to 1543 keV.

Figure 4. The pulse height spectrum of the radiopotassium solution

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.). 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 minutes. Figure 5 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.
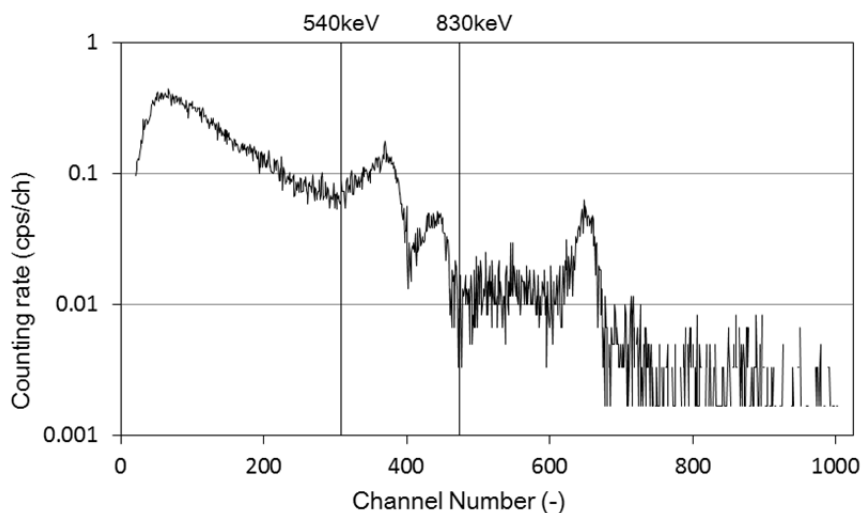

Figure 5. The pulse height spectrum of the radiocesium solution

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 determined by repeating measurements of the same wastewater.

RESULTS AND DISCUSSION
Result of the water shield test
     Figure 6 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 to an unshielded condition.
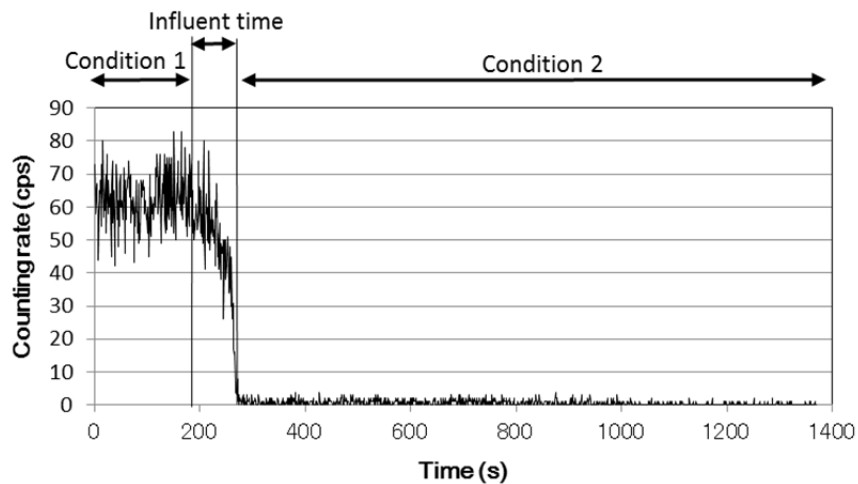

Figure 6. Result of the water shield test

Result of the radiopotassium solution test
     Figures 7 and 8 show the relations between the radioactive concentration of potassium-40 and the gamma ray counting rate measured by the 2-inch and 5-inch diameter scintillation detectors, respectively. Both regression curves can be regarded as linear. The results suggest that Cesimoni-water® is a feasible system for radiopotassium measurement.
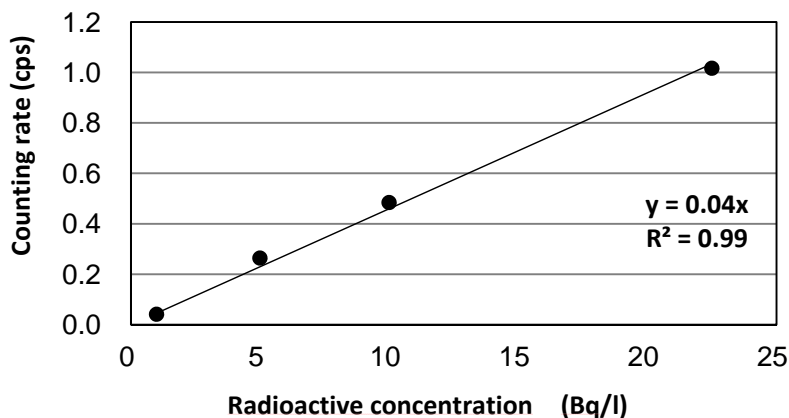

Figure 7. Relation between the radioactive concentration of potassium-40 and the gamma ray counting rate measured by the 2-inch scintillation detector
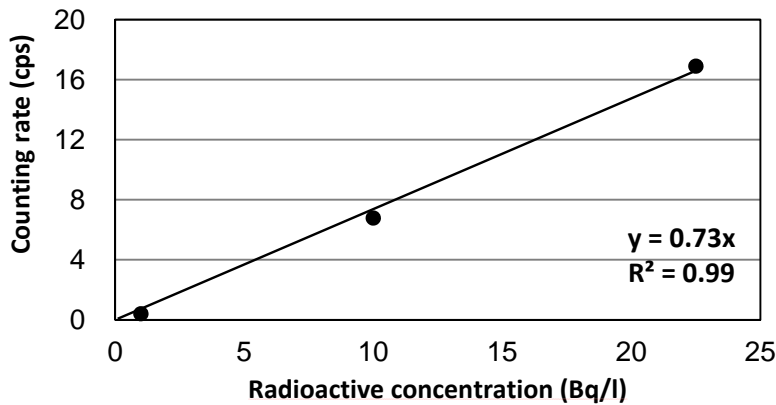
Figure 8. Relation between the radioactive concentration of potassium-40 and the gamma ray counting rate measured by the 5-inch scintillation detector

Result of the artificial radiocesium solution test

Figures 9 and 10 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 equation (1) (Ministry of Health, Labour and Welfare, 2011). The LODs of the Cesimoni-water® system equipped with the 2-inch and 5-inch scintillation detectors were 1.94 Bq/l and 0.34 Bq/l, respectively, for radiocesium wastewater.

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

        C_l : LOD of the gamma ray counting rate (cps)
        C_b : Background gamma ray counting rate (cps)
        T_s : Measurement time of the solution (s)
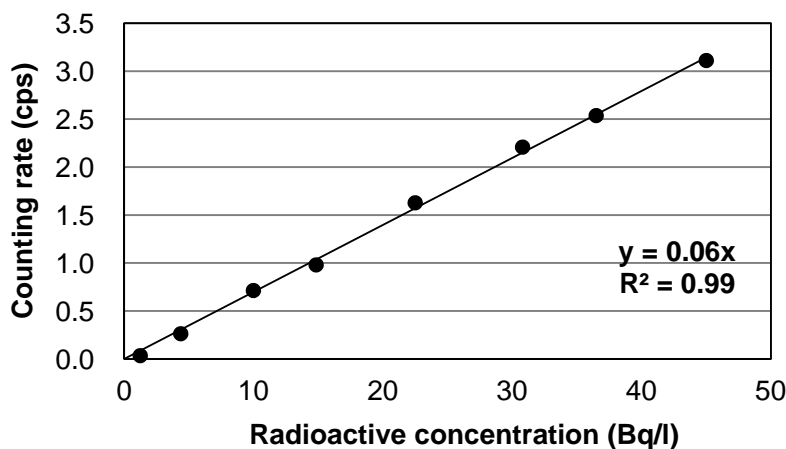 T_b : Measurement time of the background radioactivity (s)

Figure 9. Relation between the radioactive concentration of radiocesium and the gamma ray counting rate measured by the 2-inch scintillation detector
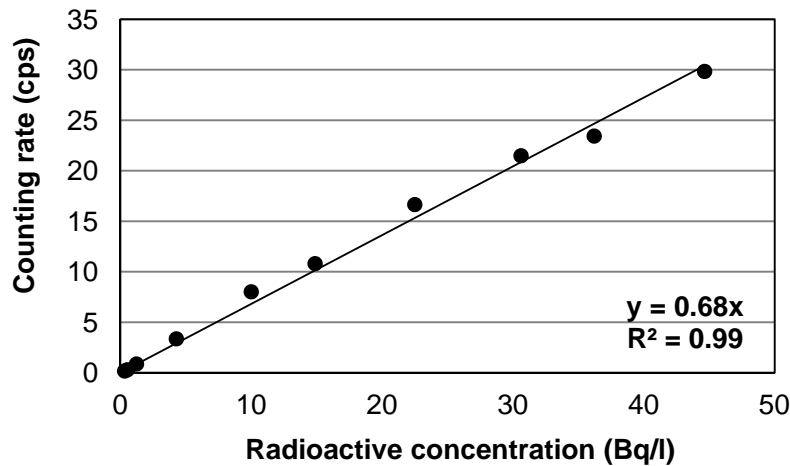
Figure 10. Relation between the radioactive concentration of radiocesium and the gamma ray counting rate measured by the 5-inch scintillation detector

Result of the wastewater test

Figure 11 shows the findings of the repeatability test by the Cesimoni-water® system. The $\pm 3 \sigma$ ※ region of the gamma ray counting rates was confirmed to be less than 0.5 (Currie, 1995; Uhrovčík, 2014). The findings suggest that the Cesimoni-water® system is feasible for the radiocesium measurement of actual wastewater.
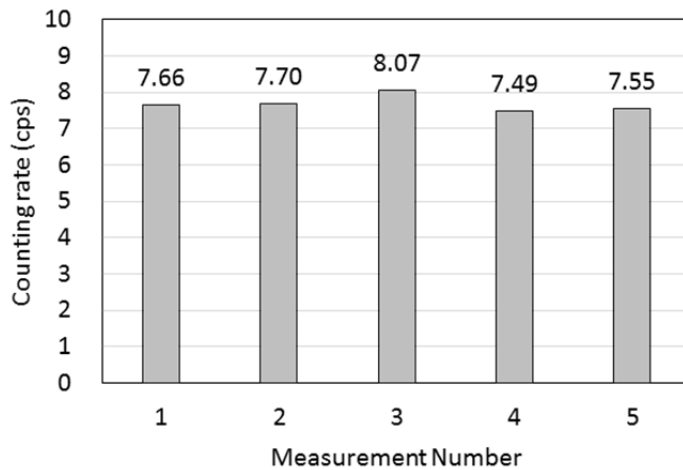
※ σ : Standard deviation

Figure 11. Result of the repeatability test for actual wastewater

CONCLUSION

The findings of this study suggest that the Cesimoni-water® system is capable of measuring a low radioactive concentration of less than 10 Bq/l as the limit of quantitation at an operating rate of more than 10 m³/hour. 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.

ACKNOWLEDGEMENT

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