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#### **KEYWORDS**

Radioactive plume; Continuous air sampling; Silicon photodiode; Detection limit **Summary** In this study, a portable radioactive plume monitor using a silicon photodiode was developed for the detection of a radioactive plume (e.g. <sup>131</sup>I, <sup>134</sup>Cs and <sup>137</sup>Cs) in an emergency situation. It was found that the background count rate was proportional to ambient dose equivalent rate and the detection limit for the monitor at 20  $\mu$ Sv h<sup>-1</sup> as an ambient dose equivalent rate was evaluated to be 187 Bq m<sup>-3</sup> using the ISO11929 method. These results suggest that the detection limit for the system can be decreased effectively by lead shielding with optimized thickness.

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## Introduction

Immediately after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, the System for Prediction of Envi-

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<sup>1</sup> Radiation Protection Department, Nuclear Fuel Cycle Engineering Laboratories, Japan Atomic Energy Agency, 4-33 Muramatsu, 319-1194 Tokai, Japan. ronmental Emergency Dose Information (SPEEDI; Imai et al., 1985) was not functioning well. The Nuclear Regulation Authority (NRA) of Japan decided not to use the SPEEDI any longer when judging whether an evacuation of residents was needed or not in a nuclear emergency (NRA, 2014). To fill the gap caused by not using the SPEEDI, many measuring devices for radioactive plume detection must be widely deployed around nuclear facilities (NRA, 2012). Thus, a compact and inexpensive radioactive plume monitor should be developed. In this study, a portable radioactive plume monitor using a silicon photodiode was developed, and the performance test of the developed monitor was carried out to determine its detection limit.

https://doi.org/10.1016/j.pisc.2019.100414

<sup>\*</sup> This article is part of a special issue entitled Proceedings of 27th International Conference on Nuclear Tracks and Radiation Measurements (ICNTRM).

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**Figure 1** Schematic diagram of the portable radioactive plume monitor.

## Materials and methods

#### Outline of the portable radioactive plume monitor

A schematic diagram of the monitor is shown in Fig. 1. The system consists of a silicon photodiode (\$3590-09, Hamamatsu Photonics Co., Ltd.), a bias supply, a preamplifier, a linear amplifier and a multi-channel analyzer (MCA) (APG7300A, Techno AP Co., Ltd.). The thickness of the depletion layer of the silicon photodiode is set at 0.3 mm. Airborne radionuclides are continuously collected on the filter using a pump which can collect the air at a flow rate of 10 L min<sup>-1</sup>, and radiation released from the radionuclides is detected by the silicon photodiode. According to the report by UNSCEAR (2014), the main radionuclides of concern for internal and external exposures because of the FDNPP accident were <sup>131</sup>I, <sup>134</sup>Cs and <sup>137</sup>Cs. It is well known that most artificial radionuclides emit beta particles as well as gamma rays. So a radioactive plume can be detected by measuring beta-particles emitted from such target radionuclides as <sup>131</sup>I, <sup>134</sup>Cs and <sup>137</sup>Cs.

#### Performance test in Fukushima Prefecture

A performance test of the developed monitor was carried out in Namie Town, Fukushima Prefecture, which was in an evacuation zone set by the Japanese government (UNSCEAR, 2014). The background counts caused by gamma-rays were measured with the monitor at 11 sites. The background measurement interval was set as 10 min. The gamma-ray pulse height distributions were obtained using a 3-in.  $\times$  3in. NaI(Tl) scintillation spectrometer (EMF-211, EMF Japan Co., Ltd.) at the same site, and these were unfolded using a response matrix method, which was reported by Minato (2001), for the evaluation of the absorbed dose rates in air. The ambient dose equivalent rates were calculated by multiplying the absorbed dose rates in air by the conversion factor of 1.25 (Omori et al., 2016). The detection limits for beta-particles were calculated using two methods which were reported by the International Organization for Standardization (ISO) (ISO11929, 2010) and Kaiser (1970). In the ISO method, the detection limit is calculated using Eqs.

(1) and (2). On the other hand, the method by Kaiser uses Eq. (3).

$$\mathbf{y}^* = \mathbf{k} \cdot \frac{1}{\mathbf{V} \cdot \varepsilon_{\beta} \cdot \varepsilon_{s}} \sqrt{\left(\frac{2r_0}{t}\right)} \tag{1}$$

$$y^{\#} = \frac{2y^{*} + k^{2}(1/(V \cdot \varepsilon_{\beta} \cdot \varepsilon_{s} \cdot t))}{1 - k^{2}\{u_{\text{rel}}^{2}(V) + u_{\text{rel}}^{2}(\varepsilon_{\beta}) + u_{\text{rel}}^{2}(\varepsilon_{s})\}}$$
(2)

$$\mathbf{y}^{\#} = \frac{(3/2)\left[(3/t) + \sqrt{(3/t)^2 + 4r_0 \cdot (2/t)}\right]}{\mathbf{V} \cdot \varepsilon_{\mathrm{B}} \cdot \varepsilon_{\mathrm{s}}} \tag{3}$$

Here,  $y^*$  is the decision threshold (Bq m<sup>-3</sup>), k is the quantiles of the standardized normal distribution for the probability of 0.95 (i.e. k = 1.65), V is the sampling volume (m<sup>3</sup>),  $\varepsilon_{B}$ is the counting efficiency,  $\varepsilon_s$  is the collection efficiency,  $r_0$ is the background count rate (cps),  $t_g$  is the measurement time (s),  $y^{\#}$  is the detection limit (Bq m<sup>-3</sup>), and  $u_{rel}(x)$  is the relative uncertainty for the parameter x. In this study, the relative uncertainties for the sampling volume, the counting efficiency and the collection efficiency were assumed as zero because these values were not evaluated. The sampling volume, the counting efficiency for beta-particles and the collection efficiency were assumed to be 0.1 m<sup>3</sup>, 0.05 and 1, respectively, because it was supposed that the sampling flow rate and measurement time were  $10 L min^{-1}$  and 10 min, and the collection efficiency was not accurately calculated. The filter which had a collection efficiency of almost 1 was used.

## Performance test in Okinawa Prefecture

The continuous measurement of airborne radionuclides using a pump which had a maximum air flow velocity of  $5L \text{min}^{-1}$  was carried out in Okinawa Prefecture. According to the previous report (Furukawa et al., 2015), absorbed dose rates in air in Okinawa Prefecture were higher compared with the other prefecture. The sampling flow rate was set as  $2.5 L \text{min}^{-1}$ . The radiation emitted from the radionuclides collected on the membrane filter was measured for two days.

#### **Results and discussion**

#### Performance test in Fukushima Prefecture

The relationship between the ambient dose equivalent rates evaluated using a 3-in.  $\times$  3-in. Nal(Tl) scintillation spectrometer and the count rates of the portable radioactive plume monitor is shown in Fig. 2. The background count rates caused by gamma-rays were directly proportional to the ambient dose equivalent rates ( $y = 174x^{0.708}$ ,  $R^2 = 0.877$ ). The result suggests that this monitor would be suitable for gamma-ray measurements.

The detection limits calculated by the ISO method and Kaiser's equation are shown in Fig. 3. The detection limit for  $20 \,\mu$ Sv h<sup>-1</sup> as an ambient dose equivalent rate, which was decided by the NRA for early protective measures, was evaluated to be  $187 \text{ Bq m}^{-3}$  using the ISO method. This value was higher than the detection limit of  $100 \text{ Bq m}^{-3}$  at  $20 \,\mu$ Sv h<sup>-1</sup>



Figure 2 The relationship between the ambient dose equivalent rates evaluated using a 3-in.  $\times$  3-in. Nal(Tl) scintillation spectrometer and the count rates of the portable radioactive plume monitor. The dotted line indicates a power approximation.



Figure 3 The relationship between the ambient dose equivalent rates evaluated using a 3-in.  $\times$  3-in. Nal(Tl) scintillation spectrometer and the detection limits calculated by the ISO method and Kaiser's equation.

as an ambient dose equivalent rate under emergency situation decided by the NRA (2015). It is suggested that the detection limit for the system can be decreased effectively by using lead shielding optimized for the thickness. Furthermore, the counting efficiency and the collection efficiency should be calculated to evaluate a more accurate detection limit.

#### Performance test in Okinawa Prefecture

The spectrum obtained when measuring the radiation released from airborne radionuclides collected on a membrane filter for continuous air sampling in Okinawa Prefecture is shown in Fig. 4. This result showed that alpha particles from radon progenies (<sup>212</sup>Po, <sup>214</sup>Po and <sup>218</sup>Po), which are risk factors for lung cancer (WHO, 2009), were detected



**Figure 4** The spectrum obtained measuring radiation from airborne radionuclides collected on a membrane filter for continuous air sampling in Okinawa Prefecture.

by the silicon photodiode. Generally, a silicon semiconductor detector is used for the measurement of radon progenies (Tokonami et al., 1996). However, the result suggests that the portable radioactive plume monitor using the silicon photodiode would be suitable for measurements of radon progeny concentrations. It is planned to carry out an experiment to determine the equilibrium factor, which is the ratio between radon concentration and equilibrium equivalent radon concentration.

# Conclusion

A portable radioactive plume monitor using a silicon photodiode was developed and its performance tests were carried out to determine the detection limit. A strong correlation was observed between the ambient dose equivalent rates evaluated by a 3-in.  $\times$  3-in. NaI(Tl) scintillation spectrometer and the count rates measured by this monitor. The detection limit was evaluated to be 187 Bq m<sup>-3</sup> using the ISO method. This suggests the need for lead shielding optimized for thickness. Alpha particles can be detected by this monitor, so the portable radioactive plume monitor would be suitable for measurement of radon progeny concentration. In the future, it is necessary to consider the following matters: (1) the counting efficiency and the collection efficiency; (2) the accurate evaluation of the relative uncertainty; and (3) how much the detection limit of this monitor will decrease if it is covered by a lead shield. The counting efficiency for beta particles of target radionuclides will be calculated by using a beta particle standard source of <sup>137</sup>Cs or by measuring beta particles from <sup>214</sup>Pb which has similar maximum beta energy to that of the target radionuclides.

## Acknowledgment

This work was partially supported by JSPS KAKENHI Grant Number JP16K15368.

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