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Measurement system for alpha and beta emitters with continuous air sampling under different exposure situations

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A R T I C L E I N F O

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A B S T R A C T

In the present study, a continuous radioactive aerosol measurement system (CRAMS) was developed for measurements of radioactive plume (e.g. ^{131}I, ^{134}Cs and ^{137}Cs) under the emergency situation, and measurements of radon/thoron progeny under the existing situation. As a result, it is suggested that the CRAMS could follow the variation of radon concentration, and the detection limit of the CRAMS under ambient dose equivalent rate of 20 μSv h^{-1}, where the temporary evacuation is required within one week in the Japanese regulation, was evaluated to be 129 Bq m^{-3} in the manner of ISO11929.

1. Introduction

After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred in 2011, prompt detection of the radioactive plume was required for the safe evacuation of general public in the framework of nuclear disaster prevention. Since it was difficult to know the amount of radionuclides released from the nuclear power plant and due to the uncertainty of the meteorological information, the System for Prediction of Environmental Emergency Dose Information (SPEEDI; Imai et al., 1985) was not used effectively for general public immediately after the FDNPP accident. In October 2014, the Nuclear Regulation Authority (NRA) in Japan decided not to use the SPEEDI any longer when judging whether evacuation is needed or not (NRA, 2014). Given such circumstances, many measuring devices for radioactive plume detection need to be widely deployed around nuclear facilities. In addition, radon is a radioactive noble gas that is taken into the body by breathing and it is well-known as a hazardous material that can cause lung cancer through internal exposure (WHO, 2009). Thus, there is a need for a continuous radon measurement system under the existing situation.

In the present study, a dual-purpose continuous radioactive aerosol measurement system (CRAMS) was developed for measurements of radioactive plume (e.g. ^{131}I, ^{134}Cs and ^{137}Cs) under an emergency situation, and for measurements of radon under the existing situation. Furthermore, basic characteristics of the CRAMS were also investigated.

2. Materials and methods

2.1. Outline of the CRAMS

A schematic diagram of the CRAMS is shown in Fig. 1. This system consists of collection and detection parts. The collection assembly includes a pump, a membrane filter (Millipore DA, Merck Millipore Ltd.) and a filter holder. The detection head accommodates a filter holder which the detector faces for continuous air sampling. The detection assembly consists of a silicon semiconductor detector (CAM-490-AM, CANBERRA Industries Inc.), a bias supply (CANBERRA 3102D), a pre-amplifier (ORTEC 142), a linear amplifier (ORTEC 570) and a multi-channel analyzer (MCA) (APG7300A, Techno AP Co., Ltd.). Beta particles emitted from radionuclides which were collected on the filter are measured by the silicon semiconductor detector, and the energy spectrum of beta particles is analyzed and registered at the MCA. The thickness of depletion layer for Si semiconductor detector is set as 300 μm with a bias voltage of +70 V. The geometrical efficiency of the CRAMS is evaluated to be 10.5% using the values of the effective diameter of detector (26.0 mm), the effective diameter of filter (18.0 mm) and the distance between detector and filter (11.5 mm).

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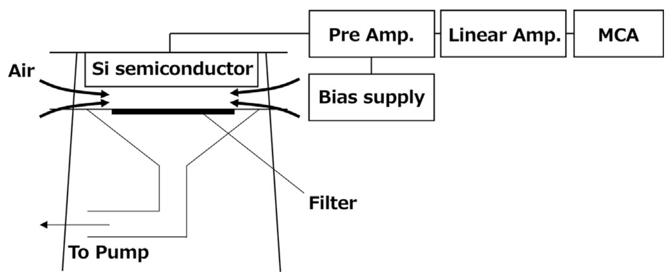

Fig. 1. A schematic diagram of the CRAMS.

Table 1
The main nuclides for internal and external exposures to the residents and ^{214}Pb which is one of radon progenies.

Radionuclides	β-ray maximum energy (MeV)	Remarks
^{131}I	0.606 (89.4%)	
^{134}Cs	0.658 (70.2%)	
^{137}Cs	0.514 (94.4%)	
^{214}Pb	0.667 (46.5%), 0.729 (41.1%)	Radon progeny

According to a recent UNSCEAR report (UNSCEAR, 2014), the main nuclides for internal and external exposures to residents as a result of the 2011 Great East-Japan earthquake and tsunami were found to be ^{131}I, ^{134}Cs and ^{137}Cs. Thus, these nuclides are targeted for evaluation by the CRAMS. It is well known that most artificial radionuclides emit beta particles as well as gamma radiations. So, the region of interest (ROI) of beta-energies of target nuclides, which are shown in Table 1, is decided according to the result obtained for ^{214}Pb in the previous study (Kurosawa and Tokonami, 1991). As shown in Table 1 (BIPM, 2013, 2008, 2006, 2004), the energies of target nuclides are close to the energy of ^{214}Pb. The energy window of the CRAMS is optimized by adjusting the energy window of the beta region. The ROI of the CRAMS is decided as 100 − 600 keV for the following reasons: (1) to minimize the influence of electronic noise generated from the system, (2) to detect targeted beta particles effectively after taking into account their maximum energy. In fact, the background count rate in the region is 0.38 cps with ambient dose equivalent rate of 0.04 µSv/h. Furthermore, the region was set after considering beta energy emitted from artificial radionuclides such as ^{131}I, ^{134}Cs and ^{137}Cs.

2.2. Performance test under the existing situation

Beta particles emitted from radon progeny are measured by the CRAMS in Toki City, Gifu Prefecture, which is an area of Japan with relatively high radon concentration. The measurement time interval and the flow rate are adjusted to 10 min and 10 L min^{-1}, respectively, and then the count rates caused by radon progeny collected on the filter are evaluated from the spectrum of beta particles. Radon concentration in the atmosphere is simultaneously measured using a pulse type ionization chamber (AlphaGUARD, Saphymo GmbH, Germany).

2.3. Performance test at an evacuation zone in Fukushima Prefecture

The performance test of the CRAMS is also carried out in Namie Town, Fukushima Prefecture, which was decided as an evacuation zone by Japanese government after the FDNPP accident. The total count rates by the CRAMS at several measurement points in Namie Town are compared with the ambient dose equivalent rates obtained by a 3-in. × 3-in. NaI(Tl) scintillation spectrometer (EMF-211, EMF Japan Co., Japan) to evaluate a conversion factor from count rate to dose rate. In this case, the measurement interval for the CRAMS are set as 10 min for background measurement. The gamma-ray pulse height distribution obtained by a 3-in. × 3-in. NaI(Tl) scintillation spectrometer is unfolded using a response matrix method for the evaluation of the absorbed dose rate in air. The details of the response matrix method were described in the previous reports (Hosoda et al., 2016, 2015; Minato, 2001). The ambient dose equivalent rate was calculated by multiplying the absorbed dose rate in air by a conversion factor (Omori et al., 2016). To evaluate the detection limit of the CRAMS, the ambient dose equivalent rate obtained at each measurement point in Namie Town is converted to the total count rate. The detection limits for several dose rates were calculated using Eq. (1) and Eq. (2) which were reported by the International Organization for Standardization (ISO) (Calmet et al., 2008; ISO, 2010). In addition, the detection limits were calculated using Eq. (3) which was reported by Kaiser (1970). The Kaiser's equation was often used for radiation safety management in Japan.

$$y^* = k \cdot \frac{1}{V \cdot \varepsilon_\beta} \cdot \sqrt{\frac{2r_0}{t}} \tag{1}$$

$$\eta^*_{\mathrm{ISO11929}} = \frac{2y^* + \left(\frac{1}{V \cdot \varepsilon_\beta \cdot t}\right) \cdot k^2}{1 - k^2 \cdot \{u^2_{\mathrm{rel}}(V) + u^2_{\mathrm{rel}}(\varepsilon_\beta)\}} \tag{2}$$

$$\eta^*_{\mathrm{Kaiser}} = \frac{\frac{3}{2}\left\{\frac{3}{t} + \sqrt{\left(\frac{3}{t}\right)^2 + 4r_0 \cdot \frac{2}{t}}\right\}}{V \cdot \varepsilon_\beta} \tag{3}$$

Here, y^* is the decision threshold (Bq m^{-3}), 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^3), ε_β is the counting efficiency, r_0 is the background count rate (s^{-1}), t is the counting time (s), η^* is the detection limit (Bq m^{-3}), $u_{\mathrm{rel}}(x)$ is the relative uncertainty for the parameter x. Since the relative uncertainties for the sampling volume (i.e. flow rate), the counting efficiency of the CRAMS are not evaluated in this study, these values are assumed as zero for the evaluation of the detection limit according to the ISO approach. Furthermore, the counting efficiency of the CRAMS is assumed as 10% according to the previous report (Kurosawa and Tokonami, 1991). In the study, the beta counting efficiency for ^{214}Pb was estimated to be about 10.5% in the similar ROI (100–300 keV) and geometric arrangement.

3. Results and discussion

3.1. Performance test under the existing situation

The daily variations of the count rates measured by the CRAMS and the radon concentrations measured every hour by AlphaGUARD are shown in Fig. 2. These measurements were continuously carried out over 48 h. The daily variations of count rates obtained by the CRAMS were in relatively good agreement with the variations of the radon concentration. These results suggest that the CRAMS developed in this study is able to evaluate the variation of radon progeny concentration.

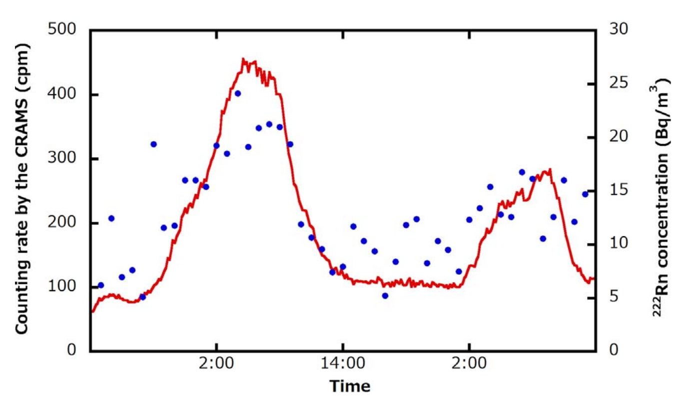

Fig. 2. The daily variations of the count rates measured by the CRAMS and the radon concentrations measured every hour by AlphaGUARD.