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Notes

Handmade Becquerel meter using commercial scintillation survey meter

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Abstract: After devastating nuclear disasters, the Becquerel meter should be very important for monitoring the contamination of our surrounding environment and judging the risk of exposure. However, the commercial Becquerel meters are too expensive for the citizens to purchase. Even the availability for the administrative agency is not ensured in emergency. Considering these risks, a self-produced Becquerel meter using a commercially available scintillation survey meter is proposed. The sample for measurement should be set in a cylindrical bottle having a diameter of 3 cm and the height of 5 cm. Although the shielding of the system with lead is not perfectly given, the subtraction of the background dose enables the measurement of the radiocesium concentration of the contaminated soil samples with sufficient accuracy. The theoretical error analysis revealed the relative error depends on the total counts and the contribution of the background radiation. The counts required for the measurement with 10 % of error is 4500 if the contribution of the background radiation is 80 % of the total counts, while the required counts reduced to 600 if the contribution of background is 50 %. The correction methods to apply this system to the sample with variable thickness in the bottle is also presented. Because the relationship between the counts rate and the radiocesium concentration depends on the proportion of ^{134}Cs to ^{137}Cs, the calibrations are generally required at each measurement. However, no significant difference was revealed for the calibration lines measured at Jan. 2013 and Aug. 2013 during which the proportion of ^{134}Cs highly decreased. Thus, any frequent calibrations are not necessary for the practical use of the system. When the aforementioned conditions are satisfied, the proposed Becquerel meter meets the practical demand for knowing the radioactivity of soil samples.
Key Words : radiocesium concentration, radioactive contamination, simple measurement, soil contamination, contamination of agricultural products

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1. Introduction

On unforeseen contamination by radionuclides in a nation scale such as an accident of nuclear power station, not only researchers or governmental officers who should prepare apparatus to monitor the situation, but citizens also eager to know the real situation. At the first step, they would purchase dose meter or survey meter, to monitor the surrounding environment. But it is impossible to measure the concentration of radionuclide in samples of soil or foods without having an expensive professional instrument. For the official purposes, the methods to measure the concentration of radionuclides contained in foods or soil is defined in the guidelines published by the government. For example, Ministry of Health Labor and Welfare (2012) established a guideline on the analytical method of radionuclides in foods in which the analysts must use a germanium semiconductor spectrometer (Ge spectrometer) having relative efficiency larger than 15 % and surrounded by lead shield with a thickness of 10 to 15 cm. NaI (TI) spectrometer which is much cheaper than Ge spectrometer have been also used for screening or voluntary analysis of foods. Although the lower limit of the measurable concentration by the system is as dilute as 1 Bq kg^{-1}, it is yet too expensive and bulky for citizens to purchase. The availability is also questioned after devastating accidents. In fact, the shortage of the instruments was reported just after the accident of Fukushima Daiichi Nuclear Power Station in 2011, leading researchers on the front-line to seek alternative methods to measure radioactivity of soil samples using commercially available scintillation survey meter (Nemoto et al., 2013). They proposed to measure the radiation from the surface of the 1 kg of soil sample packed and filled in a 2 L beaker. The background radiation is subtracted from the total counts to determine the net radiation from the sample. Although the principle to measure the radioactivity of samples is common for all methods, the mass of the sample, the shielding capacity, and the uniformity of

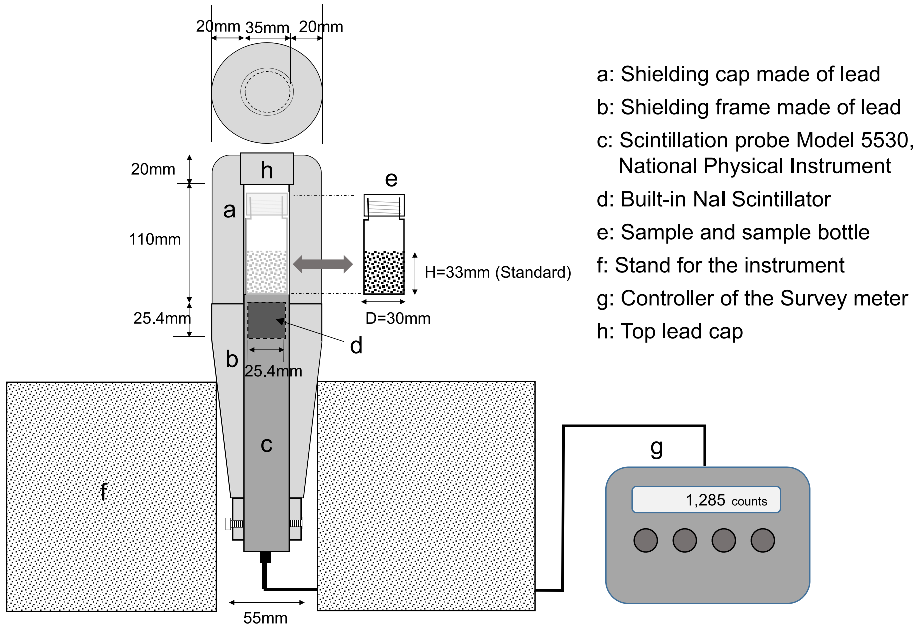

Fig. 1 Schematic of the proposed apparatus.

the geometry are the highly substantial factors to establish useful methods. The method proposed by Nemoto et al. (2013) does not require any apparatus except the scintillation survey meter. However, the large mass of soil sample is necessary to assure the consistent accuracy.

In-situ measurement of vertical profile of radiocesium concentration in soil inexpensively was also studied. Suzuki et. al. (2013) arranged photodiodes along a rod to detect the vertical radiation profile from which the concentration profile is analyzed. Due to the low sensitivity of the photodiode sensors, long counting time is required for accurate measurements. But the required time can be reduced by replacing the sensors to more sensitive ones, with additional cost.

The present report aims to provide an information about making a cheap but reliable Becquerel meter from a commercially available gamma ray survey meter and rolled lead sheet. The accuracy of the apparatus is thoroughly discussed to present the applicability of the method.

2. Design of the apparatus

Measurement of radiocesium concentration of contaminated soil samples by arranging a commercial scintillation probe (Model 5530) and survey meter (Model 5000 CYPHER) was tested. The gamma sensitivity of the scintillation probe is approximately 30,000 cpm μSv^{-1} h^{-1} for ^{137}Cs. according to the manufacturer. The survey meter including the probe approximately costs 4,000 US dollars. The GM (Geiger-Muller) tube survey meters are cheaper than scintillation survey meters, though the measuring time would have been longer due to the lower sensitivity. Fig. 1 shows the design of the instrument. The instrument consists of a shielded sample holder and shielded scintillation probe.

A cylindrical shielding cap (a) and shielding frame (b) were made of lead sheets which is soft enough to shape. The cost for the 6.5 kg of rolled lead sheet used for the shielding cap and frame was about 200 US dollars. We wrapped multiple thin lead sheets having thickness of 3 mm around a cylindrical mold having a diameter of 35 mm, each of which was glued with adhesive agent (Super Glue). The resultant thickness of the shielding wall was 20 mm, which attenuates direct gamma beam to 7 %. Because the lead wall alone is not enough to shield the background dose completely, subtraction of the background radiation is necessary. The procedure will be discussed later. The top of the cylindrical cap (a) was filled with a top lead cap (h) having thickness of 20 mm. The probe (c) must be set at the predefined position in the cylindrical shielding to be fixed by the screws when used. Thus, the probe can be taken out from the frame easily to be used as a survey meter to monitor the dose rate in environment. The NaI scintillator (d) having a diameter of 25.4 mm and a length of 25.4 mm (1 inch) is installed at the top of the probe, where gamma beam from the sample and the background is converted to light and then the light is converted to electronic pulse signal by the photomultiplier. The probe is connected to the survey meter (g) (analyzer) which displays count of photon in a predefined period. Because the system we used cannot detect the energy spectrum, the response depends on the energy. The major energies of the gamma beam from radiocesium are 661 keV (^{137}Cs), 605 keV (^{134}Cs) and 796 keV (^{134}Cs) (e.g. Malonda and Carles, 2012). Due to the

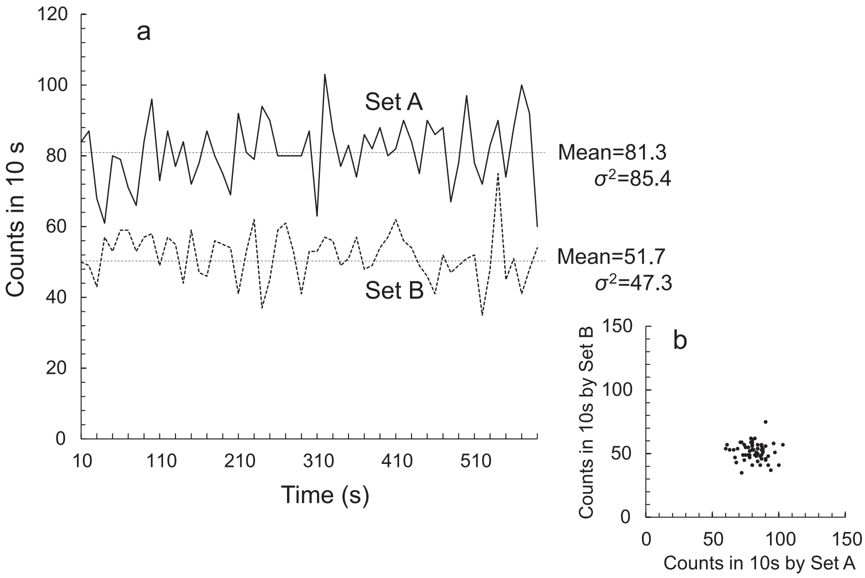

Fig. 2 Blank counts per 10 seconds; (a) Fluctuation during 600 seconds; (b) Correlation in counts per 10 seconds between a couple of the apparatus put next to each other.

dependency of the sensitivity on energy, the relationship between the counts and the dose can change when the proportion of ^{134}Cs and ^{137}Cs changes due to time. This drawback will be discussed later. A couple of the measurement sets: probes, caps, frames and the controller, were constructed to compare the individual specificity.

3. Methods of calibration and accuracy test

The contaminated soil samples were taken from a profile of a paddy field located in Iitate Village, Fukushima, Japan and the bottom of a reservoir in Motomiya City, Fukushima Japan. The samples were subject to drying in an oven at 105 °C. The highly contaminated sediment of the agricultural reservoir was mixed with non-contaminated volcanic ash soil to make a specimen with variety of predefined radiocesium concentration, which was 100 %, 80 %, 60 %, 40 %, 20 % and 10 % of the original sediment by weight. The volcanic ash soil was taken from the experimental field of The University of Tokyo located in Nishi-Tokyo city before the accident of Fukushima Daiichi Nuclear Power Station. The radiocesium concentration of these samples were measured by the Ge semiconductor detector run by Radioisotope (RI) Center of the Graduate School of Agricultural and Life Sciences, The University of Tokyo. The samples were also subject to the counting of photons by the proposed system. The contribution of the sample to the detected count of photons were evaluated by subtracting the background from the total. The background can be determined by measuring the radiation without setting the samples, to be called blank. Among the expected factors influencing the accuracy, duration of the measurement, the background dose rate, and the thickness of the sample in the bottle were examined. All the measurements were performed in an unshielded laboratory in the University of Tokyo located in Bunkyo-ku, Tokyo. Only the Set A was used for the measurement except for the comparison between the probes.

4. Results and discussion

The variation of the background radiation was examined using a couple of the proposed apparatus which were the same model of the survey meter but the sensibility to the gamma ray fluence are different due to the shielding capability. Fig. 2 shows the temporal variation of the background detected by the probes (Set A and Set B) in 10 seconds. Observed numbers of photons in space at predefined time interval is widely known to follow Poisson distribution (e.g. Malonda and Carles, 2012) represented by $P(\lambda)$. When the probability variable X follows the Poisson distribution and the mean value is λ, the variance of X is also λ. The observed mean (Mean) and variance (σ_b^2) shown in Fig. 2a are consistent with this theoretical knowledge. Because the mean count is proportional to the duration of the measurement, the theory tells that the standard deviation increases proportionally with the root square of duration of measurement. This means that the longer we spent for the measurement, the less the relative error becomes. This principle is important in measuring dilute samples. Because the count due to the sample alone (N_s) is evaluated by subtraction of the count due to the background (N_b) from the measured count (N):

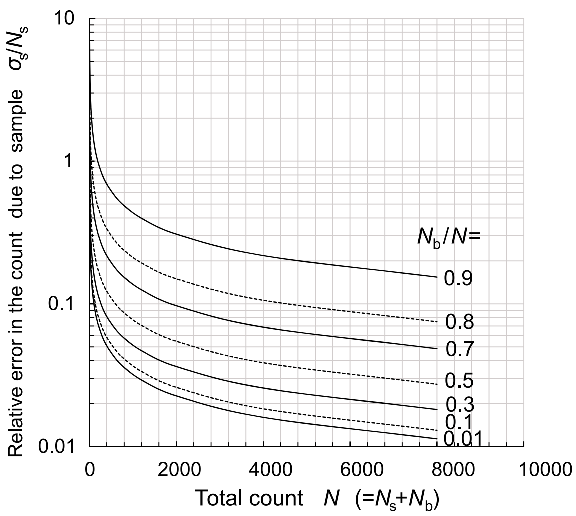

Fig. 3 Theoretical estimate of relative error in the count due to sample N_s as a function of the proportion of the background radiation N_b to the total count N.

$$N_s = N - N_b \tag{1}$$

the range of error in the count due to the sample can be estimated by eq. (2) based on the principle of propagation of error:

$$\sigma_s^2 = \sigma^2 + \sigma_b^2 \tag{2}$$

where $\sigma_s{}^2$, σ^2 and $\sigma_b{}^2$ are variance of N_s, N and N_b, respectively. Since $N = \sigma^2$ and $N_b = \sigma_b{}^2$,

$$\sigma_s^2 = N + N_b \tag{3}$$

Then the relative error in N_s can be written as

$$\frac{\sigma_s}{N_s} = \frac{\sqrt{N+N_b}}{N-N_b} \tag{4}$$

The diagram of this relationship is provided as Fig. 3, which is useful to estimate the minimum total count to obtain the count due to the sample with practical accuracy. We should usually wait until the total count meets the condition because N_b/N cannot be reduced. For instance, the total counts required for the concentration measurement with 10 % of error ($\sigma_s/N_s = 0.1$) is 4500 if the contribution of the background (N_b/N) is 80 %, while the required counts reduced to 600 if the N_b/N is 50 %. In general, the relative contribution of N_b : N_b/N is not known in advance. Thus, the rate of counts should be measured for the sample with the minimum radioactivity among the samples for some minutes to roughly estimate N_b/N.

Although two probes were set on the same table, the counts in 10 seconds measured by the both probes were never synchronized to each other. Fig. 2b examines the correlation between the counts by both probes. It is obvious no correlation was found in the temporal trends of the counts in 10 seconds detected at the neighboring location. This basic characteristic show real-time subtraction of the background does not work to increase the accuracy of measurements.

Table 1 shows the variation in the averaged blank counts per minute (CPM) due to the measurement dates. The measurement was kept for an hour and the counts per every minute were averaged. The variation due to the dates can be negligible compared to the fluctuation observed in every minute. Therefore, frequent measurement of blank CPM is not required unless the environmental radiation is known to be considerably change. However, needless to write, the measurements of samples and blanks must be done in the same place because the background radiation highly depends on the place.

Fig. 4 shows the relationship between total radiocesium concentration of mixture of the contaminated sediment and andosol measured by Ge semiconductor detector and Counts in 400 seconds by the proposed apparatus. The temporal decay of the concentration is corrected based on the date of the measurements. The relationship was so linear that the CPM obtained by the proposed apparatus can be converted to concentration. However, we should note that the relationship could change depending on height, density and $^{134}Cs/^{137}Cs$ ratio of samples.

The thickness (height) of the sample perpendicular to the scintillator affects the self-attenuation of the gamma

Table 1 Variation in averaged blank CPM due to the measurement dates.

		2016/3/11 PM	2016/3/14 AM	2016/3/14 PM	2016/3/15 AM	2016/3/21 PM
Set A	Mean	490	491	490	488	492
	σ	21.7	23.3	25.5	24.3	21.7
Set B	Mean	311	309	312	310	307
	σ	18.0	17.1	18.1	17.0	17.6

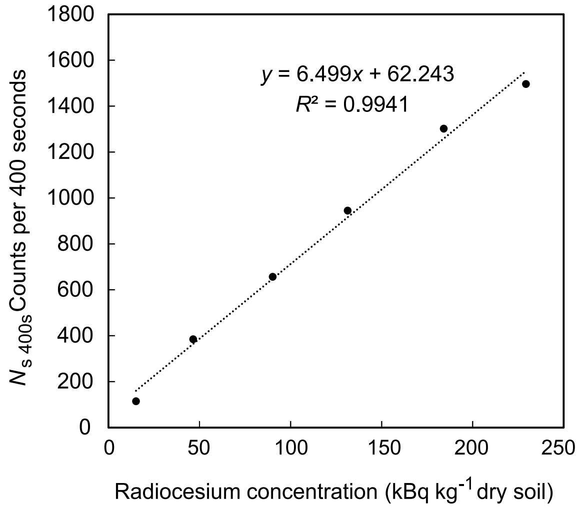

Fig. 4 Relationship between total radiocesium concentration measured by Ge semiconductor detector and counts per 400 seconds by the proposed apparatus (Set A). The sample was the mixture of sediment in an agricultural reservoir and non-contaminated upland soil. The temporal decay of the concentration is corrected based on the date of the measurements.

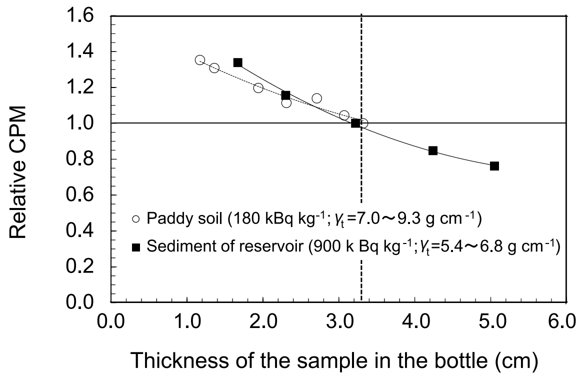

Fig. 5 Dependency of CPM on thickness of sample in bottle. Normalized to the CPM for the sample with thickness of 3.3 cm. Set A is used for these measurements.

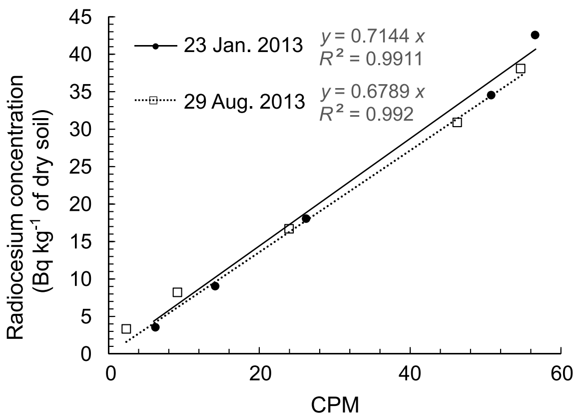

Fig. 6 Temporal change in relationship between CPM by the proposed apparatus (Set A) and total radiocesium concentration. The samples were taken from a profile of a contaminated paddy field. The concentrations were corrected to the day when the CPM measurement was performed.

beam. Fig. 5 shows the relative decrease in the CPM per unit weight of soil with an increase in the thickness of the sample in the bottle. Thickness of 33 mm was applied as the reference. The 10 % of decrease in thickness caused 5 % increase in CPM for paddy soil; the 30 % decrease caused 15 % of increase in CPM for the sediment. The dependency of CPM on the thickness had a slight difference between the samples having different bulk density. The dependency of CPM on the thickness for the denser sample (paddy soil) was less than that for the looser sample (sediment). If the thickness of the sample is adjusted to the standard thickness, the difference in the soil density is practically less important.

Because the measurement of numerous samples usually took so much time that the radiocesium concentration of the samples were subject to the effect of natural decay. Thus, the measured concentrations should be converted into the ones at the reference date by considering the half-life period and existing ratio of ^{134}Cs and ^{137}Cs. However, since the survey meter we used cannot detect the energy spectrum, the response depends on the component of the energy of the gamma ray. Fig. 6 compares the difference in the relationship between the radiocesium concentration and CPM due to the measurement date: 23 Jan. and 29 Aug. 2013, during which the proportion of ^{134}Cs to the total Cs should decreased from 0.34 to 0.28. Because the covariance analysis reveals no significant difference in the regression coefficients, the calibration is not frequently required unless the measurement spans many years.

5. Conclusion

Self-produced Becquerel meter using commercially available scintillation detector is proposed. Sufficient accuracy can be ensured if the measurement is conducted for adequate time determined from the background radiation and the concentration of the sample. Shieling by lead sheet helps reduce the effect of background radiation and shorten the time required for the accurate measurement. Although any types of detector can be principally used, the NaI scintillation probe should be the best regarding both the cost and the sensitivity. Detectors with a function of counting photons in any time interval is highly recommended to produce the system.

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要　旨

原子力事故の後には，周辺環境の汚染モニタリングや被爆リスクの判断のためにベクレルメータが不可欠である．しかし，市販のベクレルメータは一般市民には高価であり，しかも非常時には行政機関でさえもすぐに入手することはできない．これらのリスクを考慮し，比較的入手が容易な市販のシンチレーションサーベイメータを用いてベクレルメータを自作し，その精度を検証した．作成したベクレルメータは，シンチレーションプローブの周囲に鉛板を巻き付けることで，簡易な遮蔽を行うとともに，ブランク測定によるバックグラウンドを差し引くことで，汚染試料の測定において実用上十分な精度を得ることができた．

キーワード：放射性セシウム濃度，放射能汚染，簡易計測，土壌汚染，農産物汚染