

## Feasibility Study of Radioisotope $^{132}\text{Cs}$ Production Using Accelerator-Based Neutrons

Md Kawchar Ahmed Patwary

Department of Advanced Energy Engineering Science, Interdisciplinary Graduate School of Engineering Sciences (IGSES), Kyushu University

Kin, Tadahiro

Department of Advanced Energy Engineering Science, Interdisciplinary Graduate School of Engineering Sciences (IGSES), Kyushu University

Araki, Naoto

Department of Advanced Energy Engineering Science, Interdisciplinary Graduate School of Engineering Sciences (IGSES), Kyushu University

Aoki, Katsumi

Department of Advanced Energy Engineering Science, Interdisciplinary Graduate School of Engineering Sciences (IGSES), Kyushu University

他

<https://doi.org/10.5109/2547352>

---

出版情報 : Evergreen. 6 (4), pp.280-284, 2019-12. Transdisciplinary Research and Education Center for Green Technologies, Kyushu University

バージョン :

権利関係 : Creative Commons Attribution-NonCommercial 4.0 International

# Feasibility Study of Radioisotope $^{132}\text{Cs}$ Production Using Accelerator-Based Neutrons

Md Kawchar Ahmed Patwary<sup>1,\*</sup>, Tadahiro Kin<sup>1</sup>, Naoto Araki<sup>1</sup>, Katsumi Aoki<sup>1</sup>, Kosuke Yoshinami<sup>1</sup>, Masaya Yamaguchi<sup>1</sup>, Yukinobu Watanabe<sup>1</sup>, Masatoshi Itoh<sup>2</sup>

<sup>1</sup>Department of Advanced Energy Engineering Science, Interdisciplinary Graduate School of Engineering Sciences (IGSES), Kyushu University, 6-1 Kasuga-koen, Kasuga, Fukuoka, 816-8580, Japan.

<sup>2</sup>Cyclotron and Radioisotope Center, Tohoku University, Sendai 980-8578, Japan.

\*E-mail: patwary.md.kawchar.ahmed.760@s.kyushu-u.ac.jp

(Received August 23, 2019; Revised October 2, 2019; accepted October 27, 2019).

We propose that  $^{132}\text{Cs}$  ( $E_\gamma = 668$  keV and  $T_{1/2} = 6.5$  d) can replace  $^{137}\text{Cs}$  ( $E_\gamma = 662$  keV and  $T_{1/2} = 30$  y) as a new environmental tracer of cesium. The alternative tracer,  $^{132}\text{Cs}$ , can potentially reveal the short-time dynamics in an environment, which are considered to dominate radioactive cesium absorption after nuclear accidents. We first investigate the production yield and radioactive purity of  $^{132}\text{Cs}$  in a production experiment at the Cyclotron and Radioisotope Center of Tohoku University. The  $^{132}\text{Cs}$  was produced via the  $^{133}\text{Cs}(n,2n)$  reaction with neutrons generated by bombarding accelerated deuterons onto a 4 mm-thick neutron converter made of carbon. The generated neutrons were then irradiated onto a  $\text{Cs}_2\text{CO}_3$  sample. The experiment yielded 102.2 kBq/g of  $^{132}\text{Cs}$ , sufficient for tracing environmental cesium. The radioactive purity reached 98%, indicating negligible amounts of by-products. Next, a  $^{132}\text{Cs}$  tracer experiment was performed on three different soil samples: an andosol soil, a haplic fluvisol soil, and a gleyic fluvisol soil. This feasibility study confirmed that the new tracer can measure soil cesium distributions as adequately as the  $^{137}\text{Cs}$  tracer. Thus, we conclude that  $^{132}\text{Cs}$  is a promising alternative environmental tracer of  $^{137}\text{Cs}$ .

Keywords:  $^{132}\text{Cs}$ ,  $^{137}\text{Cs}$ , environmental tracer, accelerator-based neutrons.

## 1. Introduction

At present, the dynamics of radioactive materials in the environment have received much attention. The behavior of radioactive cesium in soil is especially important for avoiding public radiation exposure after a nuclear accident. On March 11 of 2011, the Great East Japan Earthquake occurred and following that the Fukushima Dai-ichi Nuclear Power Plant accident happened. After that Japanese government had no choice but revises the electric power generation deal, because of large amounts of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were released along with other radioactive materials during the accident<sup>1-3)</sup>. Because the radioactive cesium isotopes  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  have longer half-lives (30 y and 2 y, respectively) than the other radioisotopes, they are dominant sources of public exposure at the present time<sup>4-7)</sup>. From an environmental recovery perspective, the  $^{137}\text{Cs}$  contribution is the most important. Radioactive cesium is strongly bound in soils<sup>8)</sup> and sediment particles containing micaceous clay minerals (such as illite and vermiculite)<sup>9)</sup>. Therefore, understanding cesium dynamics in the soil and sediment particles is important for environmental recovery. In other words, we should

investigate how cesium is absorbed, the current microscopic states of cesium in the soil, and how these states can be removed by decontamination and volume reduction of contaminated soil.

Cesium dynamics can be understood by releasing a tracer that emits gamma rays, which are then intercepted by a radiation detector. In almost all researches, the tracer is  $^{137}\text{Cs}$  itself. However, because this radioisotope has a long half-life, the radioactive waste must be measured for a long time after the research work. Thus, radioactive cesium with a shorter half-life is desired in these investigations.

To encourage this avenue of study, Nagai et al. proposed a  $^{132}\text{Cs}$  tracer generated by an accelerator<sup>10)</sup>. The  $^{132}\text{Cs}$  tracer has the following properties<sup>11,12)</sup> (see also Fig. 1 (b)):

- 1) Cesium-132 emits 668-keV gamma rays with ~98% emission probability.
- 2) The half-life of  $^{132}\text{Cs}$  is 6.5 d, very much shorter than that of  $^{137}\text{Cs}$  (~30 y).
- 3) Cesium-132 can be produced via the  $^{133}\text{Cs}(n,2n)$  reaction with accelerated neutrons.

The most intense gamma rays of  $^{132}\text{Cs}$  have very similar energies to  $^{137}\text{Cs}$  gamma rays (see Fig. 1), removing the need for correcting by the detection efficiency or the  $G(E)$  function<sup>13)</sup>. Meanwhile, the short half-life reduces the time and amount of radioactive waste management after the investigation. Moreover, the parent nuclide of the production method is  $^{133}\text{Cs}$ , which accounts for 100% of the natural cesium. Therefore, even when using natural cesium as the target material, we can expect a high production yield with few by-product impurities. However, the production of  $^{132}\text{Cs}$  from accelerator-based neutrons, and a feasibility study of  $^{132}\text{Cs}$  as a replacement of  $^{137}\text{Cs}$  tracer in different soil have not studied.

In an actual production experiment, this paper investigates the quality of  $^{132}\text{Cs}$  tracers produced via the  $^{133}\text{Cs}(n,2n)$  reaction with accelerator-based neutrons. Next, we conduct a feasibility study of  $^{132}\text{Cs}$  in andosol, haplic fluvisol, and gleyic fluvisol soil types sampled near the Fukushima Dai-ichi Nuclear Power Plant.

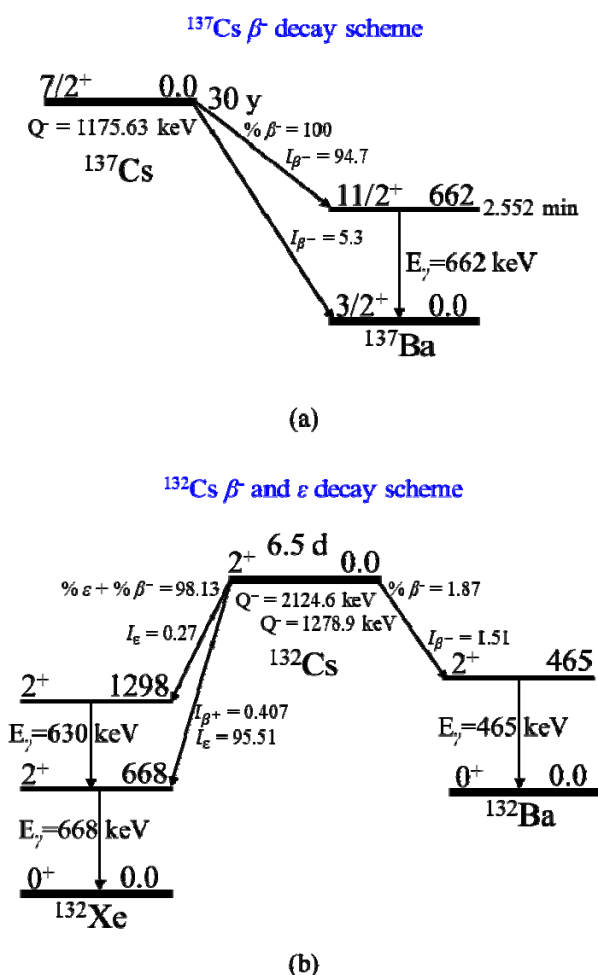


Fig. 1: Decay schemes of (a)  $^{137}\text{Cs}$  and (b)  $^{132}\text{Cs}$ . Due to its short half-life  $^{132}\text{Cs}$  is a promising tracer of  $^{137}\text{Cs}$ .

## 2. Cesium-132 Production Experiment by Accelerator-based Neutron

We first conducted a  $^{132}\text{Cs}$  production experiment. Cesium-132 can be produced via the  $^{133}\text{Cs}(\gamma,n)^{132}\text{Cs}$ <sup>14,15)</sup> and  $^{133}\text{Cs}(p,pn)^{132}\text{Cs}$ <sup>16)</sup> reactions by several routes. At this time, the optimal routes that maximize the  $^{132}\text{Cs}$  yield have not been established, although the accelerator-based neutron method shows promise, so was chosen for the present study. More specifically, the  $^{132}\text{Cs}$  was produced via the  $^{133}\text{Cs}(n,2n)$  reaction enabled by an accelerator neutron source. The neutrons were generated by bombarding deuterons onto a thick carbon target, initiating  $\text{C}(d,n)$  reactions. Figure 2 shows the neutron-induced reaction cross sections on  $^{133}\text{Cs}$  stored in EAF-2010<sup>17)</sup>. We selected the  $\text{C}(d,n)$  reaction that generates the neutrons most effectively. Note also that the neutron-energy distribution of the  $\text{C}(d,n)$  reaction has a broad peak around half of the incident deuteron energy<sup>18)</sup>. Therefore, we can select the incident energy that optimizes the energy of the neutrons. To obtain the suitable neutron-energy distribution (around 17 MeV), we expected incident deuteron energies between 20 and 35 MeV.

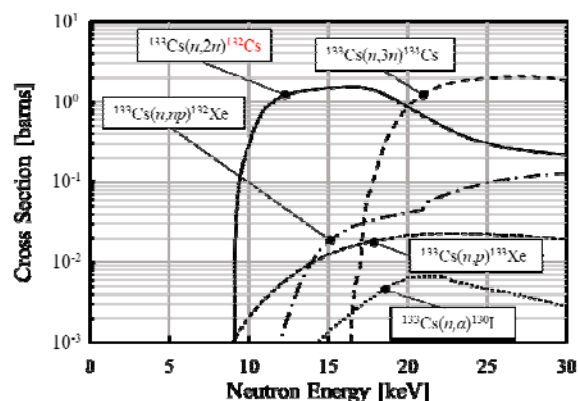


Fig. 2: Neutron-induced reaction cross section of  $^{133}\text{Cs}$ . The threshold energy of the  $^{132}\text{Cs}$  production cross-section is approximately 9 MeV; from that point, the cross-section increases with neutron energy to its maximum at approximately 17 MeV.

The  $^{132}\text{Cs}$  production experiment was conducted at the Cyclotron and Radioisotopes Center of Tohoku University, Japan. Deuterons were accelerated to 30 MeV by the accelerator and bombarded onto a circular-shaped thick carbon target having 40 mm in diameter and 4 mm thick. The generated neutrons were irradiated onto a 12-g cesium carbonate ( $\text{Cs}_2\text{CO}_3$ ) sample placed 30 mm downstream of the carbon target and  $0^\circ$  to the beam axis. The average deuteron beam current during the irradiation was  $1.2 \mu\text{A}$ . Figure 3 is a photograph and a schematic of the irradiation setup. After irradiating for 2 hours, the gamma rays emitted from the sample were measured by a high-purity germanium (HP Ge) detector

(ORTEC GMX50P4-83-RB-B-PL). The measured sample was placed 150 mm from the front head of the HP Ge detector. To derive the peak detection efficiency for this sample, a 227-kBq standard gamma-ray source of  $^{137}\text{Cs}$  was placed and measured at the same position from the detector. The detector and the measured sample were surrounded by lead blocks to reduce the natural background radiation.

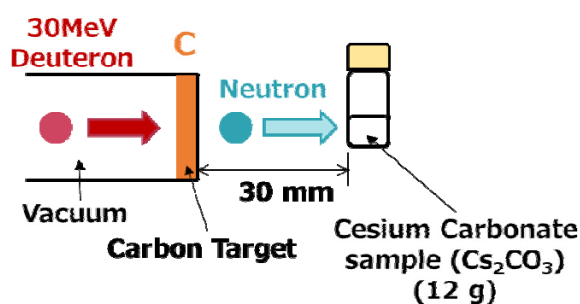
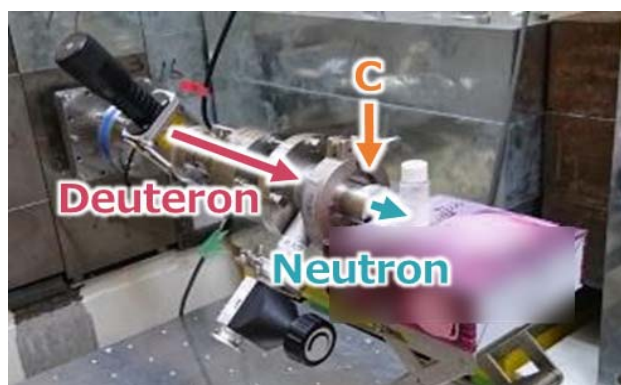


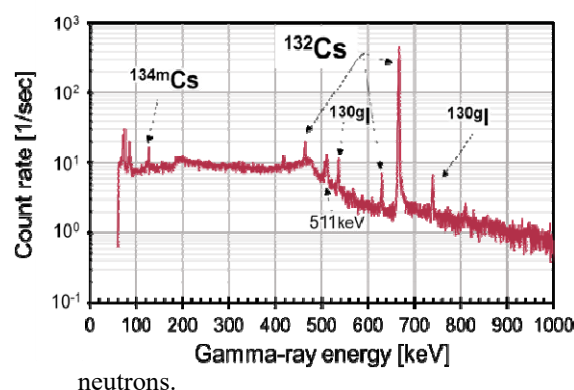
Fig. 3: Photograph and schematic view of the irradiation setup.

Figure 4 shows the spectrum of the gamma rays emitted from the irradiated  $\text{Cs}_2\text{CO}_3$  sample. The gamma rays from  $^{132}\text{Cs}$  are clearly observed, and the other photo-peaks correspond to the decay of gamma rays from  $^{130}\text{gI}$  ( $E_\gamma = 536.07$  keV and  $T_{1/2} = 12.36$  h) and  $^{134\text{m}}\text{Cs}$  ( $E_\gamma = 127.5$  keV and  $T_{1/2} = 2.912$  h). The activity of the  $^{132}\text{Cs}$  produced in the irradiation experiment was approximately 102.2 kBq/g, with a radioactive purity of 98%. This value is comparable to the estimated activity of  $^{132}\text{Cs}$  as we calculated before the experiment by considering the experimental conditions. The dominant by-products were  $^{130}\text{gI}$  (1.88 kBq/g) and  $^{134\text{m}}\text{Cs}$  (0.311 kBq/g). The activity ratios of  $^{130}\text{gI}$  and  $^{134\text{m}}\text{Cs}$  to  $^{132}\text{Cs}$  were only 1.8% and 0.3% respectively. Besides their low activity, these by-products have shorter half-lives than  $^{132}\text{Cs}$ , so their contributions can be reduced by cooling after the  $^{132}\text{Cs}$ -production irradiation. Accordingly, the influence of the by-products was negligible. Although the longer half-life of the Cs isotope  $^{134\text{m}}\text{Cs}$  ( $T_{1/2} = 2.07$  y), is problematic for disposal handling, its contribution to the  $^{132}\text{Cs}$  production was also negligible. Therefore, the by-products of this production experiment are

unimportant in waste management.

In conclusion, neutrons generated by a state-of-the-art accelerator-based neutrons enable the production of high-quality and manageable  $^{132}\text{Cs}$ .

Fig. 4: Gamma-ray spectrum of the cesium carbonate sample irradiated with accelerator-based



### 3. Feasibility study of $^{132}\text{Cs}$ tracer

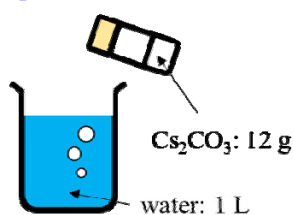
#### 3.1 Experiment

In the next step, the success of  $^{132}\text{Cs}$  as an environmental tracer was investigated in a feasibility study. For this purpose, we conducted a soil-absorption experiment using the produced  $^{132}\text{Cs}$  tracer. The soil samples were andosol, haplic fluvisol, and gleyic fluvisol were provided by the Fukushima Agricultural Technology Centre, Japan and sampled at their experimental sites where no contamination observed by the Fukushima Dai-ichi Nuclear Power Plant accident pollution.

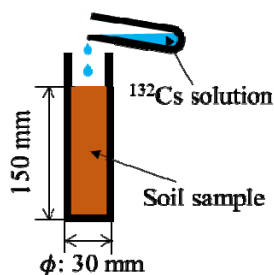
Figure 5 is a schematic view of the measurement setup. First, the sampled soils were packed into individual acrylic cylinders ( $\Phi 30$  mm  $\times$  200 mm) to a height of 150 mm. Density of the packed soil into the column is 1.215 g/cm<sup>3</sup> for haplic fluvisol, 1.076 g/cm<sup>2</sup> for andosol, and 1.224 g/cm<sup>2</sup> for gleyic fluvisol soil. Next, an aqueous solution of  $^{132}\text{Cs}$  was made by dissolving the irradiated  $\text{Cs}_2\text{CO}_3$  sample in 1 L of distilled water. This aqueous solution was dropped into the packed soil samples, maintaining a mass ratio of 5:1 between the soil and the solution.

The vertical cesium distributions were derived from the gamma rays emitted by the  $^{132}\text{Cs}$  tracer. Measurements were taken at 10-mm intervals in the depth direction (starting from the soil surface) using a NaI (TI) detector (EMF211, EMF Japan Co., Ltd.). A lead collimator was placed in front of the NaI(Tl) detector to eliminate  $^{132}\text{Cs}$  gamma rays outside the region of interest.

### 1. Preparation of $^{132}\text{Cs}$ Aqueous Solution (AS)



### 2. Pouring $^{132}\text{Cs}$ AS



### 3. Cs distribution measurement in soil

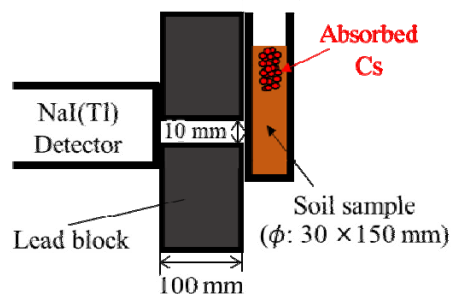


Fig. 5: Measuring the vertical distribution of  $^{132}\text{Cs}$  activity in soil samples. Distributions were measured in three types of soil samples: haplic fluvisol, andosol, and gleyic fluvisol soil.

## 3.2 Results and discussion

Figure 6 shows the vertical distributions of the  $^{132}\text{Cs}$  activities in the haplic fluvisol, andosol, and gleyic fluvisol soil samples in which their total activity is 30.72 kBq, 33.57 kBq, and 46.19 kBq respectively. In all samples, the  $^{132}\text{Cs}$  was absorbed near the soil surface. Almost 100% of the  $^{132}\text{Cs}$  was absorbed within 40 mm from the soil surface. The  $^{132}\text{Cs}$  distribution decreased along the vertical direction. The different absorption patterns reflect the different taxonomies and compositions of the three soils. However, as is widely known, cesium is absorbed within the first 10 cm in the dominant soil types (andosol soil, forest and grassland soil, and fluvisol soil)<sup>19-22</sup>.

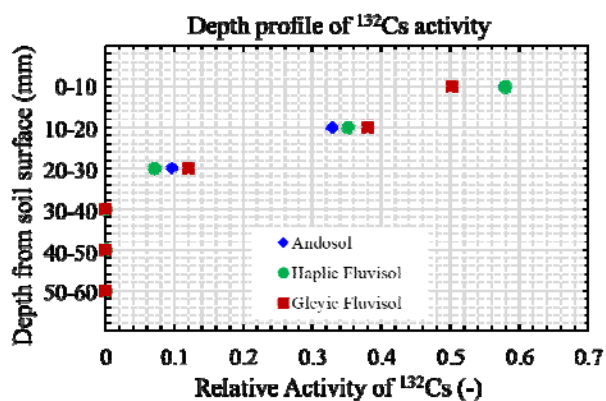


Fig. 6: Vertical distributions of  $^{132}\text{Cs}$  activity in the three soil samples. The measured points are the relative activities of  $^{132}\text{Cs}$  at the respective soil depths. The activity distribution remains steady after 40 mm.

Previous studies have investigated the soil absorption of  $^{137}\text{Cs}$  gamma rays<sup>23-27</sup>. For example, Ohno et al.<sup>24</sup> analyzed the  $^{137}\text{Cs}$  distribution in soil samples collected

within a 60-km west of the Fukushima Dai-ichi Nuclear Power Plant at one month after the accident. Almost 100% of the  $^{137}\text{Cs}$  in their experiment was detected within 40 mm from the ground surface. Similar behavior was observed in the present experiment using the  $^{132}\text{Cs}$  tracer. Therefore, we concluded that  $^{132}\text{Cs}$  is a plausible alternative tracer to  $^{137}\text{Cs}$  to investigate Cs-adsorptive nature of the soil.

## 4. Summary and Conclusions

Cesium-132 was proposed as a promising alternative environmental tracer to  $^{137}\text{Cs}$ . To study its tracing feasibility, we first determined the production amount and radioactive purity of  $^{132}\text{Cs}$  in a production experiment. The  $^{133}\text{Cs}(n,2n)^{132}\text{Cs}$  reaction, initiated by irradiating 30 MeV deuterons on a carbon neutron converter, yielded 102.2 kBq/g of  $^{132}\text{Cs}$  with a radioactive purity of 98% after irradiation. Since the activity of  $^{134g}\text{Cs}$  after decay of  $^{134m}\text{Cs}$  has very small and it is negligibly small compared to the activity of  $^{132}\text{Cs}$ , so it can be eliminated after a few days of cooling. Next, we conducted a feasibility study of the produced  $^{132}\text{Cs}$ . The cesium absorptions to andosol, haplic fluvisol, and gleyic fluvisol soil particles were measured by an NaI(Tl) gamma-ray detector. Almost 100% of the  $^{132}\text{Cs}$  was distributed within 40 mm from the surfaces of all soil samples. The results are consistent with previous studies on  $^{137}\text{Cs}$  soil absorption. We conclude that  $^{132}\text{Cs}$  can replace  $^{137}\text{Cs}$  as an environmental tracer.

In future work, we will apply the  $^{132}\text{Cs}$  product to cesium dynamics in different soil samples in the environment. This work will contribute to the environmental recovery near the Fukushima Dai-ichi Nuclear Power Plant accident. Additionally, we plan to measure the Cs transition from soils to plants using the  $^{132}\text{Cs}$  tracer concept. We will further investigate the Cs dynamics in living organisms, providing valuable and

much needed data for understanding Cs dynamics in humans.

### Acknowledgements

The soil samples were provided by Fukushima Agricultural Technology Centre, Japan. The authors would like to thank Y. Suzuki and M. Sato for their kind effort to provide it. The authors are grateful to accelerator operators of CYRIC, Tohoku University, Japan to provide high quality beam during the irradiation.

### References

- 1) T. J. Yasunari, A. Stohl, R. S. Hayano, J. F. Burkhart, S. Eckhardt, and T. Yasunari, *PNAS*, **108**, 19530 (2011).
- 2) K. Adachi, M. Kajino, Y. Zaizen, and Y. Igarashi, *Sci. Rep.*, **3**, 2254 (2013).
- 3) R. Yoneda, *Evergreen*, **4**, 16 (2017).
- 4) Y. Onda, H. Kato, M. Hoshi, Y. Takahashi and M-L. Nguyen, *J. Environ. Radioact.*, **139**, 300 (2015).
- 5) H. Kato, Y. Onda, and T. Gomi, *Geophys. Res. Lett.*, **39**, L20403 (2012).
- 6) H. Kato, Y. Onda, and M. Teramage, *J. Environ. Radioact.*, **111**, 59 (2012).
- 7) G. Katata, M. Ota, H. Terada, M. Chino, and H. Nagai, *J. Environ. Radioact.*, **109**, 103 (2012).
- 8) S. Hashimoto, T. Matsuura, K. Nanko, I. Linkov, G. Shaw, and S. Kaneko, *Sci. Rep.*, **3**, 2564 (2013).
- 9) A. Konoplev, V. Golosov, G. Laptev, K. Nanba, Y. Onda, T. Takase, Y. Wakiyama, and K. Yoshimura, *J. Environ. Radioact.*, **151**, 568 (2016).
- 10) Y. Nagai, H. Makii, S. Namiki, O. Iwamoto, N. Iwamoto, and H. Sawahata, *J. Phys. Soc. Jpn.*, **81**, 085003 (2012).
- 11) E. Browne and J.K. Tuli, *Nuclear Data Sheets*, **108**, 2173 (2007).
- 12) M. P. Unterweger, D. D. Hoppes, F. J. Schima, *Nucl. Instrum. Methods Phys. Res. Sect. A*, **312**, 349 (1992).
- 13) S. Moriuchi, M. Tsutsumi, and K. Saito, *Jpn. J. Health Phys.*, **44**, 122 (2009).
- 14) T. Iinuma, T. Nagai, T. Ishihara, K. Watari, and M. Izawa, *J. Radiation Res.*, **6**, 73 (1965).
- 15) M.T.F. da Cruz and I.D. Goldman, *Phys. Rev. C.*, **42**, 869 (1990).
- 16) Y. Nagai, K. Hashimoto, Y. Hatsukawa, H. Saeki, S. Motoishi, N. Sato, M. Kawabata, H. Harada, T. Kin, K. Tsukada, T. K. Sato, F. Minato, O. Iwamoto, N. Iwamoto, Y. Seki, K. Yokoyama, T. Shiina, A. Ohta, N. Takeuchi, Y. Kawauchi, N. Sato, H. Yamabayashi, Y. Adachi, Y. Kikuchi, T. Mitsumoto, and T. Igarashi, *J Phys. Soc. Jpn.*, **82**, 064201 (2013).
- 17) J.-Ch. Sublet, L. W. Packer, J. Kopecky, R. A. Forrest, A. J. Koning, and D. A. Rochman, *EASY Documentation Series CCFE-R*, **10**, 5 (2010).
- 18) S. Nakayama, H. Kouno, Y. Watanabe, O. Iwamoto, and K. Ogata, *Phys. Rev. C*, **94**, 014618 (2016).
- 19) A. T. Campocosio and H. H. Leon-Santiesteban, "Fungal Bioremediation: Fundamentals and Applications," CRP Press, 2019 [https://books.google.co.jp/books?id=NOSLDwAAQBAJ&printsec=frontcover&source=gbs\\_ge\\_summary\\_r&cad=0#v=onepage&q&f=false](https://books.google.co.jp/books?id=NOSLDwAAQBAJ&printsec=frontcover&source=gbs_ge_summary_r&cad=0#v=onepage&q&f=false)
- 20) A. V. Konoplev, V. N. Golosov, V. I. Yoschenko, K. Nanba, Y. Onda, T. Takase, and Y. Wakiyama, *Eurasian Soil Sci.*, **49**, 570 (2016).
- 21) J. Koarashi, M. Atarashi-Andoh, T. Matsunaga, T. Sato, S. Nagao, and H. Nagai, *Sci. Total Environ.*, **431**, 392 (2012).
- 22) M. K. Carradine, "Vertical Distribution of Radiocesium in Soil Deposits on the Contaminated Areas after the Fukushima Daiichi Nuclear Power Plant Accident," Colorado State University, 2017. [https://mountainscholar.org/bitstream/handle/10217/185676/Carradine\\_colostate\\_0053N\\_14491.pdf?sequence=1&isAllowed=y](https://mountainscholar.org/bitstream/handle/10217/185676/Carradine_colostate_0053N_14491.pdf?sequence=1&isAllowed=y)
- 23) K. Fujii, S. Ikeda, A. Akama, M. Komatsu, M. Takahashi, and S. Kaneko, *Soil Sci. Plant Nutr.*, **60**, 751 (2014).
- 24) T. Ohno, Y. Muramatsu, Y. Miura, K. Oda, N. Inagawa, H. Ogawa, A. Yamazaki, C. Toyama, and M. Sato, *Geochem. J.*, **46**, 287 (2012).
- 25) K. Tanaka, Y. Takahashi, A. Sakaguchi, M. Umeo, S. Hayakawa, and H. Tanida, *Geochem. J.*, **46**, 73 (2012).
- 26) S. Shiozawa, in *Agricultural Implications of the Fukushima Nuclear Accident*, 1<sup>st</sup> ed., eds. by T. M. Nakanishi, K. Tanoi, Springer, Tokyo, p. 49 (2013).
- 27) K. Rosén, I. Öborn, and H. Lönsjö, *J. Environ. Radioact.*, **46**, 45 (1999).