Investigation of contamination in spinach collected immediately following the Fukushima Daiichi nuclear disaster

Masahiro Hirotaa, Shogo Higakib, Takumi Higakic, Seiichiro Hasezawac

aResearch Center for Human and Environmental Sciences, Shinshu University, Matsumoto, Nagano 390-8621 Japan; bRadioisotope Center, The University of Tokyo, Bunkyo, Tokyo, 113-0032 Japan; cGraduate School of Frontier Sciences, The University of Tokyo, Kashiwa, Chiba, 277-8561 Japan.

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The contamination of spinach collected immediately following the Fukushima Daiichi nuclear disaster was investigated. The radionuclides deposited in the spinach were $^{132}$Te, $^{131}$I, $^{132}$I, $^{134}$Cs, $^{136}$Cs and $^{137}$Cs. Only 40% of these radionuclides were removed when the spinach was washed with water or detergent. The two outside leaves of each spinach stump were contaminated with the radionuclides, but the three inside leaves were nearly uncontaminated. The most significant contamination was observed on the concavities, creases, veins and leaf or stem injuries of the spinach. Most of the radionuclides deposited on the surface, leading to a radioactivity concentration of the epidermal tissue 9 times that of the mesophyll tissue.

Key words: nuclear disaster, crop, spinach, contamination, radioactive iodine, radioactive cesium

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1. Introduction

An earthquake of magnitude 9.0 struck eastern Japan on March 11, 2011. The Tokyo Electric Power Company’s Fukushima Daiichi nuclear Power Station suffered serious damage from the resulting tsunami, and large amounts of radionuclides were released into the environment. These radionuclides fell to the ground with dust, rain or snow, contaminating agricultural land and crops. On March 19, 2011, the Japanese Government announced that the levels of radioactive iodine 131 ($^{131}$I) present in milk produced in Fukushima prefecture, where the Power Station was located and in spinach produced in Ibaraki prefecture, adjacent to Fukushima, exceeded the provisional regulation values established by the food sanitation law. From this point onward, levels of radioactive material exceeding the provisional values have occasionally been detected from crops produced in eastern Japan.

The main radionuclides released into the atmosphere from the reactor were $^{131}$I, radioactive cesium 134 ($^{134}$Cs) and radioactive cesium 137 ($^{137}$Cs), and the radioactivity rate of these nuclides was reported as 10:1:1 or 50:1:1. The half-lives of $^{131}$I, $^{134}$Cs and $^{137}$Cs are 8.021 days, 2.065 years and 30.17 years, respectively. Contamination by $^{131}$I may have been the most pressing issue immediately following the disaster, but contamination by $^{134}$Cs and $^{137}$Cs may become more important as time passes. Furthermore, radionuclides that have fallen on the ground or on plant surfaces are gradually transferred inside of the plants through absorption by roots, leaves and stems. Therefore, while the surface contamination of radionuclides on leaves and stems may have been important immediately following the disaster, internal contamination may be more important in the following years.

The contaminated crops collected after having passed more than several months from the nuclear accident have been investigated. However the contaminated vegetables collected immediately following nuclear accident have been hardly investigated. Although only few vegetable samples contaminated by the Fukushima Daiichi nuclear disaster could be collected due to the confusion caused by the East Japan great earthquake disaster, we thought the data obtained by investigating them was valuable.
This paper describes the contamination status of spinach collected immediately following the Fukushima Daiichi nuclear disaster.

2. Materials and methods

2.1 Samples

The radioactive materials released from the Fukushima Daiichi nuclear Power Station reached the Kanto region of Japan, consisting of Tokyo, Chiba, Saitama, Ibaraki etc., on March 15 and March 21, 2011. The first rain in the Kanto region occurred following the disaster on March 21. Spinach samples were collected in Saitama (200 km southwest from the Fukushima Daiichi Power Station) on March 20 and in Ibaraki (170 km south-southwest from the Fukushima Daiichi nuclear Power Station) on March 28. Spinach collected from Saitama was used to evaluate the effectiveness of water or detergent for removing radionuclides. The distribution of deposited radionuclides was also observed for these samples. Spinach collected from Ibaraki was used to determine whether the radionuclides were deposited on the surface or in the interior tissues of the plants.

2.2 Washing

The radionuclides deposited on the spinach and their activity concentrations were analyzed using a high-resolution gamma spectrometry system consisting of a coaxial n-type high-purity germanium (HPGe) detector with 25% relative efficiency, a spectrum stabilized 8,000 multi-channel analyzer (Princeton Gamma-Tech, MCA8016) and other electronic accessories. The HPGe detector was shielded with 10 cm of lead, 0.5 cm of copper and 0.5 cm of acrylic to reduce the background radiation. The gamma spectrometry system was calibrated with a plane source made by depositing standard solutions of 131I, 134Cs and 137Cs certified by the Japan Radioisotope Association (JRIA) on a nonwoven fabric measuring 70 mm wide × 100 mm long × 1 mm thick. Each spinach sample was enclosed in a plastic bag measuring 177 mm wide × 196 mm long × 0.068 mm thick. The removal effect of the washing treatments on the radionuclides was estimated by dividing the radioactive concentration after treatment by the radioactive concentration before treatment. For the water treatment, the spinach was washed under running tap water for 5 minutes after having been submerged in tap water for 5 minutes. For the detergent treatment, the spinach was washed under running tap water for 5 minutes after having been submerged in a 0.075% neutral synthetic kitchen detergent solution for 5 minutes. This detergent consisted of alkyl ether sulfate, amine oxide and a viscosity modifier and was manufactured by NS FaFa Japan Co., Ltd.

2.3 Imaging

The spinach samples from Saitama were also used to observe the distribution of radionuclides deposition on the leaves using an imaging plate (IP) system. The IP system consisted of IPs (FUJIFILM Corporation, BAS-MS3543), an image analyzer (FUJIFILM Corporation, FLA-9000 STARION) and an IP eraser (FUJIFILM Corporation, IP ERASER3). The leaves of spinach were separated from the stock, and each leaf was enclosed in a separate plastic bag. The images were obtained by placing the leaves on the IP for 65 h, with the front side of leaves facing the IP.

2.4 Fractionation

The spinach samples from Ibaraki were separated into epidermal tissue and mesophyll tissue by peeling the epidermis after thorough washing with distilled water. The epidermal tissue and the mesophyll tissue were placed into separate 2 ml microfuge tubes and analyzed with the high-resolution gamma spectrometry system, which was calibrated with a volume source consisting of the standard solutions of 131I, 134Cs and 137Cs.

3. Result and discussion

3.1 Washing

Fig. 1 shows the radionuclides that had deposited on the spinach collected in Saitama immediately following the Fukushima Daiichi nuclear disaster and their activity concentrations. Fig. 1 (a) displays the radionuclides and activity concentrations before washing, and Fig. 1 (b) displays the radionuclides and activity concentrations after washing with water. The 132Te, 131I, 134Cs, 136Cs and 137Cs radionuclides were detected by gamma spectrometry. Before the water treatment, the radioactive concentrations of these nuclides were 70.3, 661, 62.2, 107, 12.9 and 109 Bq/kg, respectively, when they were corrected to 9:00 JST on March 20, 2011. The radioactive concentration of all nuclides taken together was 1022.8 Bq/kg. After the water treatment, the radioactive concentrations for these nuclides were 32.9, 472, 38.5, 30.6, 2.61 and 30.0 Bq/kg, respectively. The removal rates for these radionuclides were thus 53%, 29%, 38%, 71%, 80% and 72%, respectively. The radioactive concentration of all nuclides taken together was 606.7 Bq/kg, and the overall...
removal rate was 41%. Fig. 1 (c) displays the radionuclides and activity concentrations after washing with detergent. After the detergent treatment, the radioactive concentrations for these nuclides were 25.1, 526, 42.6, 23.9, 3.58 and 27.7 Bq/kg, respectively. The removal rates of these radionuclides were thus 64%, 20%, 32%, 78%, 72% and 75%, respectively. The radioactive concentration of all nuclides taken together was 649.4 Bq/kg, and the removal rate was 37%. There was no significant difference in the removal rates between the water and detergent treatments. The removal rates for radioactive iodine, radioactive cesium and all nuclides together were 30%, 70% and 40%, respectively. It was difficult to remove radionuclides completely from the spinach by washing. Averages of the removal rates of $^{131}$I and $^{137}$Cs which were presented by some researchers in Japan were 34 ± 12% and 59 ± 9% when the spinaches contaminated by the Fukushima Daiichi nuclear disaster were washed with water. The removal rates in our study showed the same tendency as the average removal rates.

Surface contamination was investigated by using leafy vegetables contaminated experimentally with $^{131}$I and $^{137}$Cs. The removal rates of $^{131}$I were 18%, 17%, 19%, 29% and 33% and the removal rates of $^{137}$Cs were 64%, 63% 36%, 7% and 8%, respectively when washed with 1 L of distilled water 3, 7, 24, 72 and 144 h after depositing these radionuclides. The spinaches were washed 5 d (120 h) after depositing radionuclides in our study. Therefore, the removal rates in our study were compared with those of 72 and 144 h after deposition in Singhal et al.’s. The removal rate of 29% for $^{131}$I in our study agreed with that of 72 or 144 h after deposition in Singhal et al.’s. However the removal rate of 72% for $^{137}$Cs in our study did not agree with that of 72 or 144 h in Singhal et al.’s, and approximately agreed with that of 3 h.

### 3.2 Imaging

Fig. 2 shows images of the radionuclide distribution on the spinach leaves. The two outside leaves in the stump were contaminated with the radionuclides as shown in Fig. 2 (I) and (II), but the three inside leaves were nearly uncontaminated as shown in Fig. 2 (III). The inside leaves may have emerged after the fall of the radionuclides. Furthermore, the inside leaves were smaller and less extended than the outside leaves. Therefore, unlike the outside leaves, the inside leaves were not significantly contaminated.

Fig. 3 displays those points where contamination was most significant on the Saitama spinach samples. The contamination was especially prevalent on the concavities, creases, veins and injuries of the leaves. The radionuclides deposited on these points were not removed despite thorough washing of the leaves. Removing these portions may be an effective measure to decontaminate radionuclides from spinach.

### 3.3 Fractionation

Fig. 4 (a) shows the radionuclides that had deposited on the epidermal tissue and their activity concentrations, and Fig. 4 (b) shows the radionuclides that had deposited on the mesophyll tissue and their activity concentrations. $^{132}$Te, $^{131}$I, $^{134}$I, $^{136}$Cs and $^{137}$Cs radionuclides were detected in the epidermal tissue of the Ibaraki spinach samples, matching the results from the Saitama spinach samples. The radioactive concentrations for these nuclides were 1.32, 13.4, 1.32, 1.30, 0.0685 and 1.34 kBq/kg, respectively, when corrected to 9:00 JST on March 28, 2011.
Only the ¹³¹I, ¹³⁴Cs, ¹³⁶Cs and ¹³⁷Cs radionuclides were detected in the mesophyll tissue. The radioactivity concentrations for these nuclides were 1.31, 0.273, 0.0007 and 0.290 kBq/kg, respectively. After these radionuclides had soaked into the soil through rainfall, they may have been taken up by the roots to the inside of the plants. The spinach from Ibaraki, collected after the rain, may have taken up more radionuclides than the Saitama spinach samples, collected before the rain. However, the radioactivity concentration in the epidermal tissue is 9 times of that in the mesophyll tissue, suggesting that most of the radionuclides deposited on the surface of the plants.

90% of ¹³¹I was deposited on the epidermal tissue of leafy vegetables exposed to ¹³¹I in the field at night, while 60% was deposited in the mesophyll tissue of leafy vegetables exposed to ¹³¹I in the illuminated laboratory. From these results, Hungate et al. described that the uptake of ¹³¹I into the interior of plant
increased when the stomata opened and when $^{131}$I was gaseous\textsuperscript{10}. The stomata usually open when it is light. The spinaches used in our study had opportunities to take up radionuclides from the stomata because 1–2 weeks passed after depositing them. Therefore, most of radionuclides might deposit to the surface because the radionuclides were particulate.

4. Conclusion

The contamination of spinach collected immediately following the Fukushima Daiichi nuclear disaster was investigated. The radionuclides that deposited on the spinach were $^{132}$Te, $^{131}$I, $^{132}$I, $^{134}$Cs, $^{136}$Cs and $^{137}$Cs. Only 40\% of these radionuclides were removed by washing with water or detergent, demonstrating the difficulty of completely removing radionuclides from the spinach by washing. The two outside leaves of each spinach stump were contaminated with radionuclides, but the three inside leaves were nearly uncontaminated. The most significant contamination was observed on the concavities, creases, veins and leaf or stem injuries of the spinach. Most of the radionuclides deposited on the surface of the plants, and the radioactivity concentration of the epidermal tissue was 9 times that of the mesophyll tissue.

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