Elution of Radioactive Cesium from Tofu by Water Soaking

Mitsuru Yoshida¹, Hitomi Kaino¹, Saori Shidara¹, Kazuhiro Chiku¹, Mayumi Hachinohe², Shioka Hamamatsu³

¹Department of Food Science and Technology, Faculty of Applied Life Science, Nippon Veterinary and Life Science University, 1-7-1 Kyonancho, Musashino 180-8602, Japan
²Food Research Institute, National Agriculture and Food Research Organization, 2-1-12 Kannondai, Tsukuba 305-8642, Japan
³Headquaters, National Agriculture and Food Research Organization, 3-1-1 Kannondai, Tsukuba 305-8517, Japan

Elution of cesium-137 (¹³⁷Cs) from tofu into water was investigated to know the behavior of ¹³⁷Cs during preservation and cooking. The food processing retention factor (Fr) reached 0.55 when tofu was soaked in water at a ratio of 1:2 w/w for 24 h at 4°C. Doubling the amount of water did not further significantly decrease Fr. When tofu was held in water at a ratio of 1:2 w/w at a temperature of 80°C for 50 min, Fr was 0.72. Increasing the amount of water to 10 times the tofu weight did not further reduce Fr significantly. Cesium-137 is mostly bound to tofu and does not freely diffuse into water. Tofu was then soaked in water at a ratio of 1:2 w/w at 4°C for 24 h, placed in new water at a ratio of 1:2 w/w, and held at 80°C for 50 min, resulting in an Fr 0.33. This value is close to an estimated Fr calculated by multiplying the Fr of 0.55 from soaking at 4°C by the Fr of 0.72 from the hot water treatment. The calculated Fr from soybeans sequentially processing into tofu, soaking tofu at 4°C for 24 h and in hot water at 80°C for 50 min was about 0.1, indicating 90% removal of ¹³⁷Cs. Degree of decrease in ¹³⁷Cs during preservation and cooking of tofu demonstrated in this study will be useful for exposure assessment of ¹³⁷Cs through oral intake of contaminated soybeans after processing and cooking.

Key words: ¹³⁷Cs, bean curd, elution, soaking, tofu, radioactive cesium

1. Introduction

Environmental pollution with radionuclides was caused by the tsunami that severely damaged the Fukushima Daiichi Nuclear Power Plant after the Great East Japan Earthquake on March 11, 2011. Contamination of agricultural products, mainly with radioactive cesium and iodide, became an issue in Fukushima Prefecture and the surrounding East Japan area just after the accident. The Japanese government set provisional standard values for radioactive cesium (¹³⁴/¹³⁷Cs) and iodine (¹³¹I) in drinking water and food on March 17, 2011. These values were determined assuming a 5 mSv upper limit for annual radiation dose from drinking water and food and using intake estimates for Japanese consumers. Contamination with ¹³¹I was resolved within a few months due to its short half-life of 8 days. Radioactive cesium isotopes, however, have longer half-lives, 2 and 30 years for ¹³⁴Cs and ¹³⁷Cs, respectively. Standard limits were newly set on...
April 1, 2014, decreasing the limit of annual radiation dose to 1 mSv and assuming that 50% of general foods and 100% of drinking water, milk and baby food are contaminated at limit values\textsuperscript{1}). The standard limits for radioactive Cs ($^{134}$Cs + $^{137}$Cs) are 10 Bq/kg for drinking water, 50 Bq/kg for milk and baby food, and 100 Bq/kg for general food.

Cases where water or food exceeds standard limits are now rare due to natural radioactive decay, efforts for environmental remediation, and regulations based on monitoring of food by the government. Radioactive contamination of food is not currently a serious problem in Japan, but the accident of the nuclear facility illustrated the importance of intake estimation of radionuclides for setting standard limits for regulation. Standard limits for general food are applied to both raw and processed food, and processing raw materials generally decreases radioactive pollutants\textsuperscript{2–10}). More precise intake estimation of the pollutants should be possible by considering contaminant loss during processing and cooking.

Soybean is a major bean consumed in Japan. It is processed into bean curd, “tofu,” and “natto” through fermentation, in addition to oil, soy sauce, and “miso,” a fermented soybean paste. Home cooking also yields boiled soybean, “nimame.” Reduction of $^{134}$Cs and $^{137}$Cs in soybeans during processing into tofu, natto, and nimame has been reported\textsuperscript{11).} For soybean processing into tofu, 40% of radioactive Cs in soybeans, which distributed into water by washing and soaking beans and tofu refuse, is removed. Elution of Cs into water was newly added to the soaked soybeans to bring the total weight to 1000 g. The mixture was homogenized with a food processor (DLC-IJ Mini-Prep Processor; Conair Japan G.K., Tokyo, Japan). The homogenate was heated on an induction heating cooking heater (KZ-PH33; Panasonic Co., Osaka, Japan) after addition of 10 mL canola oil (J-Oil Mills Inc., Tokyo, Japan) as an antifoaming agent. After the homogenate temperature reached 85°C, heat was maintained for 15 min. The heated homogenate was filtered with a nylon meshed sheet to obtain soymilk. Soymilk was cooled in an ice bath, and 0.5% by weight of d-glucono-1,5-lactone (FU-JIFILM Wako Pure Chemical Co., Osaka, Japan) after addition of 10 mL canola oil (J-Oil Mills Inc., Tokyo, Japan) as an antifoaming agent. After the homogenate temperature reached 85°C, heat was maintained for 15 min. The heated homogenate was filtered with a nylon meshed sheet to obtain soymilk. Soymilk was was then heated in a boiling water bath for about 1 h until it solidified into tofu. The tofu was cut into $4 \times 6 \times 3.5$ cm (about 100 g) pieces and soaked in ion-exchange purified water under prescribed conditions. Soaking experiments were run in triplicate using different batch of tofu.

2.3 Radioactivity Measurement

Tofu was packed into a 100-mL U-8 container (PP-U8; Sekiya Rika, Co. Ltd., Tokyo, Japan), and soaking water was decanted into a 0.7-liter Marinelli container (SMAX-Y07, Sekiya Rika Co. Ltd.). Concentrations of $^{137}$Cs were measured by $\gamma$-ray spectrometry with germanium semiconductor detectors (GC2020, GC2520, GC4020; Canberra, Meriden, CT, USA) coupled to a multichannel analyzer (DAS1000; Canberra). Cesium-137 concentrations were calculated using Spectrum Explorer 1.71 software (Canberra) integrated with the germanium semiconductor detector. The $\gamma$-ray peak used in $^{137}$Cs measurements was 661.6 keV. Counting times were set so that the counting error was <10% of the total count, and $^{137}$Cs concentration is reported as the $\gamma$-activity at 0:00:00 on February 1, 2019 in Japanese Standard Time.

The processing factor ($Pf$) was calculated as follows: $Pf = A/B$, where $A$ is the $^{137}$Cs concentration after processing (Bq/kg [fresh wt]), and $B$ is the $^{137}$Cs concentration before processing (Bq/kg [fresh wt]). The food processing retention factor ($Fr$), was calculated as follows: $Fr = C/D$, where $C$ is the $^{137}$Cs content after processing (Bq), and $D$ is the $^{137}$Cs content before processing (Bq) based on sample mass. The data from the same batch of tofu were regarded as a set, and $Pf$ and $Fr$ were calculated using data within a set. Significant differences ($p < 0.05$) in $Fr$ between two treatments were assessed with a paired t-test as the values from the same batch of tofu were regarded as a pair.

3. Results and Discussion

The average concentration of $^{137}$Cs in prepared tofu was 16 Bq/kg. The $Pf$ of tofu prepared from dried soybeans was 0.10, which is similar to values reported by Hachinohe et al.\textsuperscript{11).} The Fr for tofu prepared in this study was 0.28, which is about half of that previously reported (0.64$)^{11).}$ This dif-
ference may be due to differences in conditions of soaking, homogenization, and separation of soymilk. Recovery of $^{137}$Cs in all soaking experiments calculated using $Fr$ of tofu and soaking water was 0.83–1.28.

A loaf of tofu was soaked in water at 4°C for 24 h, simulating storage at home. After soaking in water with twice the weight of tofu, $Fr$ reached 0.55 (Fig. 1, closed circle). Increasing the amount of water up to four times the weight of tofu did not further decrease $Fr$ significantly ($p \geq 0.05$). The $Fr$ should be 0.33 and 0.20 when $^{137}$Cs concentration in tofu becomes equal to that in water which is twice and four times the weight of tofu, respectively (Fig. 1, dotted line). Thus, more than half the $^{137}$Cs is considered to be trapped in tofu.

When the temperature was raised to 37°C, $Fr$ was reduced to 0.33 in water twice the weight of tofu for 24 h (Fig. 1, closed square). The residual ratio of $^{137}$Cs in tofu at 37°C is lower than that at 4°C, but a temperature of 37°C is not hygienic for the preservation of tofu.

Tofu is often served in hot seaweed-based broth as “yudofu” in Japan. We investigated elution of $^{137}$Cs into hot water over shorter periods of time. Tofu was added to twice its weight of boiling water, and the temperature was then maintained at 80°C for 50 min. The core temperature of the tofu reached 80°C in 20 min and was maintained for 30 min in this condition. This treatment decreased $Fr$ to only 0.72 from that of tofu before added to boiling water. Increasing the amount of water up to 10 times the weight of tofu did not further significantly ($p \geq 0.05$) reduce $Fr$ (Fig. 1, closed triangle). Cesium-137 did not freely diffuse from tofu into hot water over periods of time of less than 50 min. How $^{137}$Cs is trapped in tofu requires investigation in further research.

We further investigated the combined effects of soaking and cooking in water on release of $^{137}$Cs from tofu. Tofu was soaked in twice its weight of water at 4°C for 24 h, then shifted into twice its weight of boiling water and maintained at 80°C for 50 min resulting in an overall $Fr$ of 0.33. This value is close to that estimated by multiplying the $Fr$ of 0.55 from refrigeration by the $Fr$ of 0.72 from the hot water treatment. The calculated $Fr$ from soybeans through processing into tofu, soaking the tofu at 4°C for 24 h, and then cooking in water at 80°C for 50 min was about 0.1, indicating 90% removal of $^{137}$Cs.

Decrease in radioactive Cs by processing soybeans into tofu and during preservation and cooking of tofu in water is demonstrated in this study. This information will be useful for exposure assessment of $^{137}$Cs by oral intake of contaminated soybeans after processing and cooking.

Acknowledgments

The authors would like to thank Enago (www.enago.jp) for the English language review.

Conflict of Interest

The authors have no conflict of interest.

References

1. Ministry of Health, Labour, and Welfare. Current situation and protective measures for radioactive materials in foods. https://www.mhlw.go.jp/english/topics/2011eq/dl/food-130926_1.pdf. Accessed April 1, 2020.
2. Green N, Wilkins BT. Effects of processing on radionuclide content of food - implications for radiological assessments. Radiation Protection Dosimetry. 1996; 67(4): 281–286. doi:10.1093/oxfordjournals.rpd.a031829
3. Malek MA, Nakahara M, Nakamura R. Removal of $^{137}$Cs in Japanese catfish during preparation for consumption. J Radiat Res. 2004; 45(2): 309–317. PMID:15304975, doi:10.1269/jrr.45.309
4. Patel AA, Prasad SR. Decontamination of radioactive milk--a review. Int J Radiat Biol. 1993; 63(3): 405–412. PMID:8095292, doi:10.1080/09553009314550531
5. Pröhl G, Muller H, Voigt G, Vogel H. The transfer of $^{137}$Cs from barley to beer. Health Physics. 1997; 72(1): 111–113. PMID:8972835, doi:10.1097/00004032-199701000-00015
6. Steinhauser G, Steinhauser V. A simple and rapid method for reducing radiocesium concentrations in wild mushrooms (Cantharellus and Boletus) in the course of cooking. J Food Prot. 2016; 79(11): 1995–1999. PMID:28221918, doi:10.4315/0362-028X.JFP-16-236
7. Hachinohe M, Okunishi T, Hagiwara S, Todoriki S, Kawanmoto S, Hamamatsu S. Distribution of radioactive cesium ($^{134}$Cs plus $^{137}$Cs) in rice fractions during polishing and cooking. *J Food Prot*. 2015; 78(3): 561–566. PMID:25719881, doi:10.4315/0362-028X.JFP-14-275

8. Nabeshi H, Tsutsumi T, Hachisuka A, Matsuda R. Variation in amount of radioactive cesium before and after cooking dry shiitake and beef [in Japanese]. *J Food Hyg Soc Jpn. (Shokuhin Eiseigaku Zasshi)*. 2013; 54(1): 65–70. PMID:23470875, doi:10.3358/shokueishi.54.65

9. Nabeshi H, Tsutsumi T, Hachisuka A, Matsuda R. Reduction of radioactive cesium content in beef by soaking in seasoning [in Japanese]. *J Food Hyg Soc Jpn. (Shokuhin Eiseigaku Zasshi)*. 2013; 54: 298–302. doi:10.3358/shokueishi.54.298

10. Sato I, Sasaki J, Satoh H, Okada K. Effects of treatment time and thickness of meat on the removal of radioactive cesium from beef slices by boiling and water extraction. *J Food Prot*. 2019; 82(4): 623–627. PMID:30917040, doi:10.4315/0362-028X.JFP-18-427

11. Hachinohe M, Kimura K, Kubo Y, et al. Distribution of radioactive cesium ($^{134}$Cs Plus $^{137}$Cs) in a contaminated Japanese soybean cultivar during the preparation of tofu, natto, and ni-mame (Boiled Soybean). *J Food Prot*. 2013; 76(6): 1021–1026. PMID:23726198, doi:10.4315/0362-028X.JFP-12-441