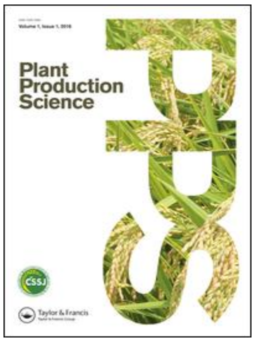

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Decreasing radioactive cesium in lodged buckwheat grain after harvest

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ABSTRACT
This study assessed soil contamination with high radioactive cesium (R–Cs) concentration in buckwheat grains by lodging, and assessed the possibility of R–Cs reduction in grain through post-harvest preparation. Analysis of buckwheat grain produced in farmers' fields and reports from farmers indicated that grain from fields that had lodging showed higher R–Cs than grain from fields with no lodging. A field experiment demonstrated that R–Cs in grain after threshing and winnowing (TW) was about six times higher in lodged plants than in nonlodged plants. In lodged plants, R–Cs in grain was decreased to about one-fourth by polishing, and was decreased to about one-seventh by ultrasonic cleaning, compared with R–Cs in grain after TW. These results demonstrate that R–Cs of buckwheat grain of lodged plants can be decreased by removing soil from the grain surface by polishing and winnowing.

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KEYWORDS
Buckwheat; cross-contamination; lodging; polishing

Radioactive cesium (^{134}Cs and ^{137}Cs, R–Cs) was released into the environment in eastern Japan by an accident at the Tokyo Electric Power Company's Fukushima Dai-ichi (No. 1) nuclear power plant, caused by the Great East Japan Earthquake and tsunami on 11 March 2011. The principal route of internal human exposure to the R–Cs can be the consumption of food containing R–Cs (Zhu & Smolders, 2000). Although the half-life of ^{134}Cs is 2.1 yrs, the half-life of ^{137}Cs is long: 30.2 yrs. For that reason, it has lasting effects on humans and the environment. Therefore, it is important to elucidate the R–Cs dynamics in agricultural environment and to reduce R–Cs concentration in foods.

Buckwheat (*Fagopyrum esculentum* Moench) is an important field crop in eastern Japan. A survey of R–Cs of buckwheat grain samples in 2011 (330 samples) and 2012 (2918 samples) revealed that 0.9 and 0.5% of buckwheat samples exceeded 100 Bq kg^{-1} (the standard limit of radioactive nuclides for food materials from April, 2012), respectively (MAFF, 2013; MHLW, 2012). Since then, countermeasure was rapidly developed to decrease R–Cs in buckwheat grain by the application of potassium (Kubo et al., 2015). No samples from the surveys of 1394 and 848 samples taken respectively in 2013 and 2014 exceeded the standard limit (MAFF 2015a, 2015b).

However, a few buckwheat samples exhibited relatively high R–Cs concentrations (under the standard limit) in spite of production with sufficient potassium application. Because we had an impression that buckwheat was cultivated in a variety of ways depend on farmers, hearing investigations of farmers was conducted to assess their cultivation and harvest practices. According to the results, we devoted attention to the cross-contamination of R–Cs in soil on the grain surface by lodging as a factor affecting the grain R–Cs concentration, because the lodging increases the risk of soil adherence to grain by bringing grain close to soil. For this study, we tested a hypothesis by which adherence of soil with high R–Cs concentration to grain caused by lodging affects the grain R–Cs concentration, and by which contamination of R–Cs on the grain surface can be decreased by post-harvest preparation.

Materials and methods

For these analyses, 19 grain samples of buckwheat produced in farmers' fields of Tochigi prefecture in 2012 were used. These samples were kindly provided by Tochigi prefectural office. Hearing investigations were conducted of farmers on the lodging during cultivation (presence or

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Table 1. R–Cs concentrations in buckwheat grain differ in lodging and contamination of extraneous substances collected from 19 farmers' fields in 2012.

Lodging	Contamination of extraneous substance	Number of farmers	R–Cs concentration in buckwheat grain (Bq kg^{-1})
Yes	Yes	12	47.7 ± 6.3
Yes	No	4	56.6 ± 14.7
No	No	3	26.5 ± 6.4

Values of R–Cs show the average ± standard error.

absence), the contamination of extraneous substances (chips of shoots) in harvested grains (presence or absence), type of field (upland or lowland), previous crop (rice, wheat or buckwheat), depth of plowing, and processing methods of earlier crops (plow under or carry out). Regarding the lodging during cultivation and the extraneous substance in harvested grains, the judgments of farmers depended on visual check. The R–Cs concentrations of grain samples were measured using germanium semiconductor detectors (GC4020; Canberra Japan KK, Tokyo, Japan / GEM20-70, Ortec; Ametek Inc., Berwyn PA) after filling the grain samples into 100-mL polystyrene containers (D24-365-080; Sekiya Rika Co., Ltd., Tokyo, Japan/5-093-02; Yamayu-umano Co. Ltd., Osaka, Japan). The values of the R–Cs concentrations in the grain were corrected with water contents of 15%. The R–Cs concentrations of grain samples were measured again after ultrasonic cleaning of grain samples collected from farmers which had lodging. Ultrasonic cleaning was conducted for 5 min with tap water using an ultrasonic cleaner (VS-100III; As One Corp., Osaka, Japan). Then samples were rinsed with distilled water.

A field experiment was conducted at a farmer's field in Date City, Fukushima Prefecture, within the area affected by the dispersal of R–Cs in 2013. Soil R–Cs concentration was 4,085 Bq kg^{-1} before sowing (Kubo et al., 2015). Buckwheat cultivar 'Aizunokaori' developed in Fukushima prefecture in 2007 (Yamauchi, 2012) was used in the experiment. The lodging resistance of 'Aizunokaori' is estimated as high because it has short plant height compared with conventional cultivars (Osawa, 2011). Characteristics of the field and experimental design were described in Kubo et al. (2015). The lodging of buckwheat in the field was caused by the typhoon No.18 (Man-yi), which had 13.2 m s^{-1} of the greatest wind velocity in Fukushima prefecture (Japan Meteorological Agency, 2013), in 16 September 2013 (around flowering stage of buckwheat). At maturity (21 October 2013; at the moment of 70% of grain blackened), grains were sampled separately from lodged plants and nonlodged plants, at the plot with 1.713 t ha^{-1} K_2O application before sowing, which correspond to 65 mg K_2O 100 g^{-1} soil. Grains of lodged plants were collected about 200 g per plot by hand. Grains of nonlodged plants were harvested from 1.5 m^2 area for each plot. Grain samples were collected with threshing and winnowing (TW) from air-dried plants. The grain yield of nonlodged plants was 129.5 kg 10 a^{-1}. The water contents of the grain was 14.4%.

Analyses of R–Cs concentrations in grain were conducted three times for each sample: after TW; after threshing, winnowing, and polishing using a polishing machine (F1; Fukubishi Co., Ltd., Fukushima, Japan) (TWP); and after threshing, winnower polishing and ultrasonic cleaning (TWPU). Ultrasonic cleaning was conducted as described above. The R–Cs concentrations of grains were measured using a germanium semiconductor detector (GC2520-7500SL; Canberra Japan KK, Tokyo, Japan) after filling the grain samples into 100-mL polystyrene containers. Measurement of R–Cs concentrations had a 10% range of error. The R–Cs concentrations were decay-corrected to October 21, 2013, using the half-life of R–Cs on a 15% water content basis.

The significance of differences among the treatments was analyzed using analysis of variance with the general linear model procedure and multiple comparisons with the Ryan–Einot–Gabriel–Welsch procedure. The difference between treatments was analyzed using t-test. Pearson's correlation analysis was used to analyze the relations between two characteristics. All analyses were conducted using software (IBM SPSS ver. 22 for Windows; IBM Japan Ltd., Tokyo, Japan).

Results and discussion

Reports from 19 farmers obtained in 2012, revealed that the type of field, the previous crop species, the depth of plowing, and the processing method of the previous crop had little effect on grain R–Cs (data not shown). The respective numbers of farmers who reported both lodging and contamination of extraneous substances, who reported only lodging, and who reported no lodging or contamination were 12, 4, and 3 (Table 1). No farmer reported contamination of extraneous substances without lodging. Grain R–Cs concentrations tended to be higher in samples collected from farmers who reported lodging than those from the farmers who did not have lodging, which indicates that lodging produces an increased potential for raising R–Cs concentration in buckwheat grain. The presence or absence of contamination of extraneous substance did not affect to grain R–Cs concentration, which might indicate that the contamination of shoot chips has little

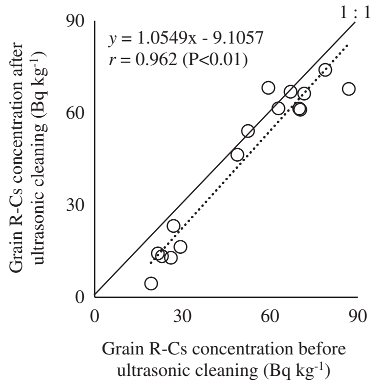

Figure 1. Relation of R–Cs concentration in grain before and after ultrasonic cleaning, collected from 16 farmers who had lodging in 2012.

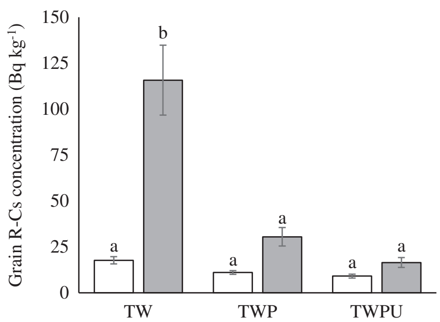

Figure 2. Grain R–Cs concentration after TW, TWP, and TWPU from lodged plants and from nonlodged plants in a field experiment in 2013. □ Nonlodged plants, ■ Lodged plants. TW, TWP threshing, winnowing and polishing, TWPU threshing, winnowing, polishing and ultrasonic cleaning. Bars show standard error (n = 3). Common letters are not significantly different according to the multiple tests of Ryan–Einot–Gabriel–Welsch (P < .05).

effect on the grain R–Cs concentration compared with lodging because the R–Cs concentration in shoots is not high (Kubo et al., 2015). From these results, we inferred that the reasons for high R–Cs in grain samples from lodged plants were (1) adherence of soil with high R–Cs to grain and/or (2) difference in degree of grain fertility. The adherence of soil with high R–Cs to grain was confirmed using grain samples collected from 19 farmers and the field experiment. The degree of grain fertility was evaluated using data obtained from the field experiment.

The relation between the R–Cs concentrations before and after ultrasonic cleaning of 16 grain samples collected from the farmers which reported lodging showed significant positive correlation (Figure 1). These results indicate that most R–Cs in grain is absorbed by roots from the soil. However, the R–Cs concentration of many samples decreased after ultrasonic cleaning, and the grain R–Cs concentration after ultrasonic cleaning decreased to 23.2% of that before ultrasonic cleaning in the most conspicuous sample. The slope and intercept of a regression line was 1.0549 and −9.1057, respectively. This result suggests that lodging has the risk to cause grain surface contamination by the adhesion of substances with high R–Cs, such as soil particles irrespective of the degree of internal contamination by root absorption. It is considered that the degree of grain surface contamination with soils depends on some factors such as the severity of lodging, soil property, and water status of soil.

In the field experiment in 2013, the TW R–Cs concentration of grains from lodged plants was 115.7 Bq kg^{-1}, and was about six times higher than that of nonlodged plants (17.7 Bq kg^{-1}) (Figure 2). The difference was statistically significant. The TWP R–Cs concentration of grains from lodged plants was 30.5 Bq kg^{-1}. It was about quarter of the TW R–Cs concentration of grains from lodged plants. The TWP R–Cs concentration of grains from nonlodged plants also decreased slightly with polishing (11.1 Bq kg^{-1}).

Figure 3. Grain samples after TW (upper) and TWP (lower) of lodged plants collected from a field experiment in 2013.

No significant difference in TWP R–Cs concentration was found between grains from lodged plants and from nonlodged plants. Additionally, TWPU decreased the R–Cs concentrations in grains from lodged plants and from nonlodged plants (16.5 and 9.1 Bq kg^{-1}, respectively). However, significant differences in R–Cs were not found between TWP and TWPU in grains from lodged plants and from nonlodged plants. Figure 3 presents grain samples from lodged plants after TW and TWP. Grains of TW have a drab color compared to grains of TWP. Some grime was apparently attached to the grain surface in grain of TW. R–Cs concentration of substances removed from grains by polishing was about 1.5 times higher in lodged plants than in nonlodged plants (lodged plants 1,491 Bq kg^{-1}, nonlodged plants 1,057 Bq kg^{-1}). These results confirmed that lodging induced the buckwheat grain surface contamination. In addition, the surface contamination was reduced by polishing even if the grains were harvested from lodged plants. Although the polishing of harvested grain is generally suggested method of grain preparation in buckwheat, there are some small-scale farmers who do not conduct polishing, according to the hearing investigation. Polishing is very important to produce buckwheat grain with low R-Cs concentration stably.

The hundred-grain weights of lodged plants and nonlodged plants were, respectively, 3.14 ± 0.04 and 3.29 ± 0.02 g, and the difference was significant according to *t*-test ($p = 0.037$). The hundred-grain weight of lodged plants was about 5% smaller than that of nonlodged plants. Although a difference in grain fertility between lodged plants and nonlodged plants related to grain R–Cs was found, results suggest that the effect was small compared with the adhesion of soil particles with high R–Cs.

It was also inferred that lodging prevention can reduce the surface contamination of substances with high R–Cs in buckwheat. Murayama et al. (2004) reported that the use of varieties with high lodging resistance, an appropriate amount of seeding, and fertilizer application is important to prevent lodging in buckwheat. The reason that there were lodging and no lodging plants in the same managed field experiment might be the unevenness of the emergence rate and growth of buckwheat affected by the unevenness of soil conditions. 'Nijiyutaka', a variety with high lodging resistance bred in 2011 (Yui et al., 2012), may be an alternative variety to reduce lodging. In addition, consideration of varieties of determinate growth (Osawa, 2011) and changing the sowing time may also contribute increased lodging resistance. Attention to these points during cultivation is expected to reduce the lodging of buckwheat.

The fate of R–Cs during the milling was investigated in wheat (Kimura et al., 2012). The study showed that the concentrations of R–Cs were found to be highest in the bran fractions, and the R–Cs concentration of flour was less than half of the concentration in whole grain. Also in buckwheat, R–Cs concentration of grain may be affected by the processing treatment such as the removal of hull, and further testing is required to clarify it.

In conclusion, to prevent cross-contamination of R–Cs in buckwheat, cultivation management to prevent lodging, in addition to careful preparation of grain with a polisher after harvesting is important to produce buckwheat with sufficiently low R–Cs.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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