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Short communication

Analyses and countermeasures for decreasing radioactive cesium in buckwheat in areas affected by the nuclear accident in 2011

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ABSTRACT

Tokyo Electric Power Company's Fukushima Dai-ichi nuclear power plant released radioactive cesium (Cs) into the environment after the Great East Japan Earthquake of March 11, 2011. After radioactive Cs fell onto agricultural fields, radioactive nuclide levels in some buckwheat (*Fagopyrum esculentum* Moench) grain subsequently exceeded new standard limits for radioactive materials, including that for radioactive Cs (100 Bq kg^{-1}) established by the Japanese government in 2012. A survey of soils and buckwheat grain from 68 farmers' fields in 2012 revealed that soil exchangeable potassium (K) concentration was significantly and negatively correlated with radioactive Cs concentration in the grain. The effect of K application on the reduction of radioactive Cs concentration in the grain was confirmed by pot and field experiments conducted respectively in 2012 and 2013. This effect might result from the similarity of ion forms of K and Cs and/or the lower exchangeable Cs concentrations at higher exchangeable K concentrations in soil. Based on these results, farmers were recommended to establish a soil exchangeable K concentration of 250 mg kg^{-1} (300 mg kg^{-1} in K_2O) before applying basal fertilizer. After this recommendation, the soil K concentrations of farmers' fields increased, causing an overall decrease in radioactive Cs concentrations of buckwheat grain produced in 2013. Consequently, no grain sample from this year exceeded the standard limits for radioactive Cs level across the affected area in Japan. These efforts contributed greatly to the rehabilitation and reconstruction of agriculture in the area contaminated with radioactive Cs.

1. Introduction

The Great East Japan Earthquake and tsunami on March 11, 2011 triggered an accident at the Tokyo Electric Power Company's (TEPCO) Fukushima Dai-ichi (No. 1) nuclear power plant (FDNPP). Radioactive cesium (^{134}Cs and ^{137}Cs) was emitted from the plant and diffused into the environment, including agricultural areas in eastern Japan. ^{137}Cs has a longer half-life, i.e. 30.2 years, compared with ^{134}Cs (2.1 years). Radioactive Cs has high biological availability, and the consumption of food containing radioactive Cs represents the principal route of human exposure to the element (Zhu and Smolders, 2000). The Japanese government first established the provisional regulation value for radioactive Cs in grains as 500 Bq kg^{-1} on March 17, 2011, and changed it to 100 Bq kg^{-1} as a new standard on April 1, 2012 (Ministry of Health, Labour and Welfare MHLW, 2012a). Extensive monitoring of the radioactivity of agricultural products has been conducted in contamination-affected areas. Only products that meet the new standard limits for radioactive Cs are distributed in markets. Agricultural researchers from various disciplines have attempted to decrease the radioactive Cs in crops and soils using their accumulated knowledge and newly acquired rapid communications (e.g., Yamaguchi et al., 2012).

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Buckwheat (*Fagopyrum esculentum* Moench) is one of Japan's most common crops. Buckwheat was produced on 56,400 ha of arable land in 2011, 14% of which belonged to prefectures predominantly contaminated by radioactive Cs (Ministry of Agriculture, Forestry and Fisheries MAFF, 2013). After TEPCO's FDNPP accident, some samples of buckwheat produced in 2011 exceeded the maximum acceptable level (500 Bq kg^{-1}, the provisional regulatory value for food materials in 2011) of radioactive nuclides (MHLW, 2012b). Development of techniques to decrease radioactive Cs in buckwheat began emerging as an objective for the rehabilitation and reconstruction of agriculture in areas contaminated by radioactive Cs.

Regarding the accumulation of radionuclides in buckwheat, Yudintseva et al. (1980) showed that lime and slag applications decrease the uptake of ^{90}Sr (strontium) and ^{137}Cs. However, information related to decreasing radioactive Cs concentrations in buckwheat in Japanese environmental conditions has been severely lacking. Research into countermeasures against radioactive Cs accumulation began in paddy rice after TEPCO's FDNPP accident in 2011 (Endo et al., 2013; Fujimura et al., 2012, 2013; Ohmori et al., 2014a,b; Saito et al., 2012). Some of these reports describe studies in which potassium (K) application prominently decreased radioactive Cs in rice.

First, we hypothesized that soil exchangeable K affects radioactive Cs in buckwheat and surveyed buckwheat samples collected from farmers' fields in areas affected by radioactive Cs in 2012. Next, we rapidly conducted a pot experiment using soils with highly radioactive Cs during winter 2012 to demonstrate that K application before sowing was effective for the reduction of radioactive Cs in buckwheat. Using these approaches, we quickly established guidelines for buckwheat cultivation in Japanese regions affected by radioactive Cs and promoted them through the MAFF, which encouraged farmers to apply additional K before sowing buckwheat. Concurrently, we conducted a field experiment at an area with high radioactive Cs in summer 2013 to ensure the efficacy of K application. Another survey of buckwheat samples in farmers' fields, where is similar area with 2012, was conducted in autumn 2013. This report presents discussion of the mechanisms of radioactive Cs transfer into buckwheat grains.

2. Materials and methods

2.1. Analyses of samples collected from farmers' fields

In 2012, samples of buckwheat grain and soil just around the plant root at the maturity stage were collected from 68 farmers' fields, covering a wide distribution of the farmers' field in areas affected by radioactive Cs across northeastern and eastern Japan. The ^{134}Cs and ^{137}Cs concentrations of grain samples were measured using germanium semiconductor detectors (GC4020; Canberra Japan KK, Tokyo, Japan/GEM20-70, Ortec; Ametek Inc., Tennessee, USA) 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) with the samples. The values of the ^{134}Cs and ^{137}Cs concentrations in the grain were corrected with water contents of 15%. The soil samples collected at 0–15 cm depth were passed through a 2.0-mm sieve and measured for total ^{134}Cs and ^{137}Cs concentrations, exchangeable ^{134}Cs and ^{137}Cs concentrations, radioactive Cs interception potential (RIP; Cremers et al., 1988), pH, total carbon and nitrogen concentrations, cation-exchange capacity (CEC), exchangeable K concentration, exchangeable magnesium (Mg) concentration, exchangeable calcium (Ca) concentration, and soil particle distribution (percent ratios of coarse sand, fine sand, silt, and clay). The ^{134}Cs and ^{137}Cs concentrations of the soil were measured in the same manner as those of the buckwheat grain. RIP was measured in accordance with the methods described by Waegeneers et al. (1999). Measurements of soil pH, C/N ratio, CEC, exchangeable K, Mg, and Ca concentrations extracted by 1 M neutral ammonium acetate (NH_4OAc), and soil particle distribution were conducted according to the methods of Sparks (1996). Exchangeable ^{134}Cs and ^{137}Cs concentrations were determined after extraction using 1 M (NH_4OAc) at the soil solution ratio of 1:10.

In 2013, buckwheat grain and the soil just around the plant root at the maturity stage were collected from 68 farmers' fields. Of these fields, 17 were repeated cultivation of buckwheat, and provided the samples both 2012 and 2013. The ^{134}Cs and ^{137}Cs concentrations of the grain samples were measured in the same manner as those of the 2012 samples. In the soil samples, exchangeable K concentration, ^{134}Cs and ^{137}Cs concentrations, and exchangeable ^{134}Cs and ^{137}Cs concentrations were measured. The transfer factor (TF) was calculated as the proportion of the ^{134}Cs and ^{137}Cs concentrations of the grain (15% moisture content) to the ^{134}Cs and ^{137}Cs concentrations of the soil (dry matter). For ^{134}Cs and ^{137}Cs concentrations of the grain less than the lower detection limit, the sample concentration was shown with the lower detection limit. TF was not calculated for these samples.

2.2. Pot experiment in 2012

The application of additional K was identified as the hopeful practice for decreasing radioactive Cs in buckwheat from analyses of samples collected from farmers' fields in 2012. A pot experiment was conducted in a glasshouse at the Fukushima Agricultural Technology Centre, Koriyama, Japan (37.5° N, 140.4° E), using Humic Andosols collected from Nishigo village, Fukushima Prefecture (total ^{134}Cs and ^{137}Cs concentration in the soil: 4093 Bq kg^{-1}) to test the reduction of radioactive Cs in buckwheat grain attributable to K application. After being passed through a 2.0-mm sieve, the soil was mixed with fertilizers and put in Wagner pots (0.05 m^2). Application rates of N (ammonium sulfate, 21.0% N) and P_2O_5 (calcium superphosphate, 20.5% P_2O_5) were, respectively, 40 and 80 kg ha^{-1}. The experimental treatments comprised four application levels of K_2O: 0, 200, 400, and 600 mg kg^{-1}. K_2O was applied as potassium sulfate (50.0% K_2O). The experimental design was a randomized complete block with three replications. One replication consisted of six pots. In each pot, 24 seeds of the buckwheat cultivar 'Aizunokaori', a major cultivar of Fukushima prefecture, were sown on December 13, 2012. Growing conditions in the glasshouse were 20 °C (day)/15 °C (night) under natural lightning. Irrigation was conducted by hand to maintain appropriate soil moisture for buckwheat growth without leaching. At the maturity stage of the buckwheat (February 25, 2013), all grains from each pot were collected by hand and prepared for the measurement of ^{134}Cs and ^{137}Cs concentrations. The soils in the pots before and after buckwheat cultivation were measured for exchangeable K concentration, as well as ^{134}Cs and ^{137}Cs concentrations. The ^{134}Cs and ^{137}Cs concentrations of grains and soils were measured using a germanium semiconductor detector (GC 2020; Canberra Japan KK, Tokyo, Japan).

2.3. Field experiment in 2013

A field experiment was conducted in 2013 to test the effects of K and zeolite applications on the reduction of radioactive Cs in buckwheat grain at a farmer's field in Date City, Fukushima Prefecture, within the area affected by the dispersal of radioactive Cs. The field soil was a Gray Lowland soil. The total ^{134}Cs and ^{137}Cs in the soil was 4085 Bq kg^{-1} before sowing. We applied zeolite as well as K because zeolite appeared to reduce the transfer of radioactive Cs from the soil to rice plants in a previous pot