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Simple method for estimating soil mass loading onto plant surface using magnetic material content as a soil indicator — Influence of soil adhesion to vegetation on radioactive cesium concentration in forage

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ABSTRACT

A simple technique for estimating soil mass loading on vegetation was developed using magnetic material content as an indicator of soil adhesion. Magnetic material contents in plant and soil samples were determined by a magnetic analyzer. High recovery rates of 85–97% were achieved in a recovery test in which additional soil was added to powdered plant materials [stem of forage corn (*Zea mays* L.), aboveground part of Italian ryegrass (*Lolium multiflorum* Lam.)] at addition rates of 12.3–200 g dry soil kg^{-1} dry plant material including soil. Samples of different Japanese cultivated soils were tested and showed a range of magnetic contents of 1.27–16.1 g kg^{-1} on a dry weight basis. These levels are considered adequate for determining soil contamination in plant materials. Then, we applied this method for confirming the effect of soil adhesion on radioactive cesium concentrations in plant samples obtained at the area affected by the 2011 nuclear accident in Japan. The mean soil mass loading (±standard deviation) on forage rye (*Secale cereale* L.) showing mild lodging was 0.8 ± 0.6 g kg^{-1}, but was 7.4 ± 5.0 g kg^{-1} for plants with serious lodging. No soil loading was detected on rye plants that showed no lodging. Radioactive cesium concentrations in the rye samples increased linearly with the increase in soil mass loading caused by plant lodging, and consequently mean radioactive cesium concentration for rye plants with serious lodging was about 2.7 times higher than that with no lodging. Cesium radioactivity in forage was affected by variations in soil mass loading onto forage plants caused by changes in plant growth and differences between plant species.

1. Introduction

Soil-borne radionuclides can be resuspended into the atmosphere and can be redeposited onto vegetation by wind action and raindrop splash. Radioactive soil particles probably adhere to plant surfaces after crop lodging and mechanical disturbance in areas exposed to radioactive fallout. Forage crop fields and grasslands in eastern Japan (Tohoku and Kanto regions) were widely contaminated by radioactive cesium (hereafter, radiocesium) that was released during the 2011 nuclear accident at the Fukushima Daiichi Nuclear Power Station (FDNPS) of the Tokyo Electric Power Company. To avoid or minimize radiocesium contamination of beef and dairy products via the consumption of roughage, Japan's Ministry of Agriculture, Forestry, and Fisheries set a provisional regulation in 2011 to limit the radiocesium concentration of roughage for cattle to 300 Bq kg^{-1} at a water content of 0.8 kg kg^{-1}. The value was subsequently lowered to 100 Bq kg^{-1}, corresponding to 500 Bq kg^{-1} on a dry weight (DW) basis, in February 2012 (MAFF, 2012, 2014). Not surprisingly, there is some concern that contamination of plant surfaces by radioactive soil can raise the radiocesium concentration in forage produced in areas affected by the nuclear accident. As complicating factors, transfer rates of radiocesium to ruminants across their gut are different among the different forms of radiocesium; the transfers of radiocesium in soil particles tend to be lower than those in forage crops (Beresford et al., 1992, 1995, 2000). Estimation of soil mass in forage is important to clarify the transfer of radiocesium to beef and dairy products via ingestion of forage-

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attached soil particles polluted with radiocesium. Kubo et al. (2016) reported that radiocesium of buckwheat (*Fagopyrum esculentum* Moench) grain of lodged plants can be decreased by removing soil from the grain surface by polishing and winnowing. Therefore, a simple method for estimating soil mass in crops is required to determine the safety of forage production and to consider countermeasures for soil adhesion to edible crops as well as forage crops in radiation-polluted areas of Japan.

Titanium (Ti) is commonly used as an indicator of soil-adhered vegetation. Soil ingestion by grazing animals was estimated for evaluation of intake of soil-borne contaminants, such as toxic metals (Thornton and Abrahams, 1983) and radionuclides (Green and Dodd, 1988), using Ti as an indicator for soil. Cook et al. (2009) examined Ti contamination of plant materials through laboratory processes: sample grinding using a Wiley mill and wet digestion with nitric and sulfuric acids increased Ti concentration by about 4 mg kg^{-1} and by 5–6 mg kg^{-1}, respectively. Ti uptake via roots of seedling shoots of tapertip hawksbeard (*Crepis acuminata* Nutt.) averaged 5 mg kg^{-1}. It was concluded that plant samples with Ti concentrations of >10 mg kg^{-1} are probably contaminated by soil. These results indicate that Ti has the potential to contaminate plant samples through laboratory processes and that plants can absorb some Ti through the root system. Cary et al. (1986) reported that hydrofluoric acid (HF) was essential for the digestion procedure used for quantitative determination of Ti in soil contamination of plants, and in analysis of rumen fistulas and feces samples. Extreme care must be taken when handling HF because of its high toxicity.

Plutonium (Pu) isotopes can be used as tracers of soil particles on the surface of plants grown in Pu-contaminated soil. The mass loading of soil particles on plant surfaces was estimated using Pu-238 (Pinder and McLeod, 1988, 1989; Pinder et al., 1991). However, Pu analysis is difficult (MEXT, 2002), and information for Pu isotopes in Japanese soils is very limited after the FDNPS accident (Zheng et al., 2012). Hinton (1992) reviewed soil mass loadings that were determined by several techniques and found a range of 2–500 g kg^{-1}.

Magnetic minerals (ferromagnetic substances), such as magnetite (Fe_3O_4), maghemite (γFe_2O_3), and the titanomagnetites, are present in most soils (Mullins, 1977). Because these minerals are unlikely to be absorbed into the plant body via the root system, magnetic material content can be used as a specific indicator for soil mass loading on plant surfaces. Because these magnetic materials show high magnetic susceptibility in an external magnetic field, magnetic susceptibility measurements of soil have been used in studies of soil formation (Mullins, 1977; Maher and Taylor, 1988; Singer and Fine, 1989; Dearing et al., 1996) and soil pollution (Hanesch and Scholger, 2005; Vodyanitskii, 2013); however, these were not quantitative evaluations for magnetic minerals in soil.

In the present study, we developed a simple method for estimating soil mass on plant surfaces using magnetic material content as a soil indicator. Measurements were recorded using a magnetic analyzer and the data were used to clarify the effects of soil-borne radiocesium contamination in forage.

2. Materials and methods

2.1. Measurements of magnetic material content and radiocesium content in soil and plant samples

Fresh soil samples were passed through a 2-mm sieve (Testing sieve; Tokyo Screen, Tokyo, Japan) made from a nonmagnetic stainless steel to remove coarse materials and plant matter, and then air-dried. Plant samples were dried in an oven at 70–90 °C for 72 h, and then were ground in a Wiley mill with a 2-mm screen (1029-JC; Yoshida Seisakusho, Tokyo, Japan). Soil and plant samples were analyzed for both magnetic material content (hereafter, magnetic content) and radiocesium content.

Magnetic contents in soil and plant samples were measured using a magnetic analyzer (MA-1040; Micromeritics, GA, USA). Soil and plant samples were oven-dried before analysis for magnetic content. Sample material was added to a sample holder (approximate volume 110 mL), which was weighed, and the net mass of the sample was calculated and input into the magnetic analyzer. The sample was inserted into the air core coil of the device, and the magnetic content as iron equivalent was displayed immediately. Following sample measurement, the measurement procedure was repeated with an empty sample holder to obtain a blank value. The magnetic content of a sample was then determined by subtracting the blank value from the value observed for the sample. For soil samples with high magnetic contents, the actual level was above the reading capacity of the instrument. As a result, ten times the net weight of the sample was input to the device, and the magnetic content of the soil sample was determined by multiplying the measured value by ten. Samples that gave a negative value for magnetic contents were considered to contain no magnetic material.

Contamination of magnetic materials such as metallic iron through the sample-grinding process was examined using cellulose powder (38 μm, 036-2225; Wako Pure Chemical Industries, Osaka, Japan). Cellulose powder (100 g) was injected into a Wiley mill, and grinding continued until almost all of the powder passed through a 2-mm screen. Magnetic contents of the cellulose powder were compared before and after grinding.

In handling finely ground samples, powder was observed to adhere to the inside and outside of the sample holder due to static electricity. To avoid contamination of the measurement device with sample materials and to eliminate the need to clean the sample holder, we also examined the use of a cellulose thimble filter (No.84; Toyo Roshi Kaisha, Tokyo, Japan; i.d. 30 mm, o.d. 33 mm, length 120 mm) as an inner vessel of the sample holder. A sample was added to the thimble filter and dried in an oven at 105 °C overnight. The oven-dried thimble filters were weighed with and without powder samples to determine sample weights. Empty thimbles were measured as blanks by the magnetic analyzer and measurement of magnetic content for sample was determined by subtracting reading for the empty thimble as the blank value from that for the sample in the thimble. Measured values of magnetic content for 15 plant samples and 9 soil samples using the thimble were compared with those by the basic measuring method without the thimble.

Soil samples were placed in U8 containers and analyzed for cesium-134 (^{134}Cs) and cesium-137 (^{137}Cs) using a germanium detector (GEM20P4-70; ORTEC, Oak Ridge, TN, USA). Plant samples were packed into 1-L or 2-L Marinelli vessels, and the total ^{134}Cs and ^{137}Cs contents were determined using a germanium detector (GC2020; Canberra Industries, Meriden, CT, USA) as well as the aforementioned detector for soil analysis. The ^{134}Cs and ^{137}Cs concentrations of the samples were corrected according to the time of harvest to compensate for radioactive decay assuming a half-life of 2.0652 years for ^{134}Cs and 30.17 years for ^{137}Cs.

Magnetic and radiocesium contents in soil and plant samples were expressed on a DW basis.

2.2. Recovery test (experiment 1)

Soil sample was collected at the Institute of Livestock and Grassland Science (NILGS, 36°55′N, 139°55′E), National Agriculture and Food Research Organization, Tochigi Prefecture, Japan, in 2010, before the FDNPS accident. The soil type was a Brown Lowland soil,