THESIS

RADIOCESIUM SOIL TO UNDERSTORY PLANT TRANSFERS IN FUKUSHIMA FORESTS

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ABSTRACT

RADIOCESIUM SOIL TO UNDERSTORY PLANT TRANSFERS IN FUKUSHIMA FORESTS

The objective of this research was to investigate the activity of radiocesium in plants and soil found in forested areas near the Fukushima Dai-Ichii Nuclear Power Station (FDNPS). Plant samples and 15-cm soil core samples were collected at four sampling locations.

The quantity of soluble and exchangeable amounts of ¹³⁷Cs in soil was measured by performance of two sequential extractions using de-ionized water and ammonium acetate respectively. The remaining radiocesium bound to the soil following the extractions was considered strongly bound to soil particles and whose movement into either soluble or exchangeable concentrations is slow. The distribution of the different forms of radiocesium were developed throughout each 15-cm soil core as measured using a High Purity Germanium Detector (HPGe).

An analysis of correlation between plant activity versus plant root area and exchangeable radiocesium resulted in limited success. It remains probable that quantifying plant root radiocesium distribution can aid in predicting plant uptake, but it is believed that the concentrations of competing ions in soil should also be quantified and parameterized as well as additional possible routes of intake (throughfall).

Concentration ratios (¹³⁷Cs plant activity/¹³⁷Cs soil activity) of 12 plants were calculated and exhibited a broad range from 0.01 to 2.5. The broad range of plant concentration falls within the International Atomic Energy Agency Parameter Handbook values of herbaceous plants in

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clay soils. Concentration ratios appear similar within sampling locations which suggests that additional soil characteristics (clay content, competing ions) can be used to describe plant uptake of radiocesium.

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INTRODUCTION

The 2011 Fukushima Dai-Ichii Nuclear Power Station (FDNPS) accident resulted in the deposition of a significant quantity of radiocesium (¹³⁷Cs and ¹³⁴Cs) over areas of mainland Japan (UNSCEAR, 2014). It is estimated that forests cover ~70% of the land area receiving the heaviest deposition of FDNS derived fallout (Hashimoto, 2012). The forest ecosystem, therefore, plays an important role in the long term fate of accidentally released radiocesium deposited on land. The objective of this was research to determine if understory plant uptake of of radiocesium (¹³⁷Cs) can by described by the vertical distribution of radiocesium in soil and the distribution pf plant root surface area. The data collected suggests that these parameters alone do not fully characterize radiocesium understory plant uptake.

FDNPP Accident

On March 11, 2011 the fourth largest earthquake (M 9.1) in recorded history occurred 70 km from the Pacific coast of Honshu, Japan. The magnitude 9.1 earthquake (USGS, 2016) resulted in a massive tsunami that struck the coast of the Tohoku region of Japan. The tsunami's toll on human life was extensive and resulted in nearly 20,000 fatalities (Nakahara, 2013), and hundreds of thousands were forced to evacuate (Zare, 2012).

The height of the tsunami was greater than the 6 m sea wall protecting the Fukushima Dai-Ichii Nuclear Power Station. The FDNPS consisted of 6 boiling water reactors (BWR) with a power generation capacity of 4.7 gigawatts electricity. The tsunami disabled backup electrical power generation to the three operational reactors. The lack of offsite power generation and additional system damage resulted in the loss of all cooling systems. Heat build up resulted in melting of reactor fuel core material and the subsequent release of fission and activation products

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into terrestrial and marine environments. The temperatures reached within the fuel cores was such that a majority of the total 520 PBq of released radioactivity consisted of mostly the volatile elements found in a fission reactor fuel core (Steinhauser et al. 2014). These volatile elements include ¹³¹I, ¹³²Te, ¹³⁴Cs, and ¹³⁷Cs and additional short lived isotopes of noble gases. Amounts of intermediate volatility ⁹⁰Sr and refractory element plutonium (²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu) releases were estimated to be orders of magnitude lower than of radicosesium or radioiodine (Steinhauser et al, 2014).

The releases of radionuclides from the FDNPP accident to the environment occurred over a prolonged period yet the most significant releases occurred within the first 5 days following March 11, 2011. Atmospheric releases of radionuclides were primarily the result of hydrogen buildup and subsequent explosion occurring at FDNPS Units 1 and 3, and from the efforts to depressurize the containment vessels in Units 1, 2 and 3. The marine environment received FDNPS derived radionuclides as a result of atmospheric deposition, direct transfer of reactor coolant, and groundwater flows.

The primary radionuclides responsible for significant potential public exposures were releases of ¹³¹I, ¹³⁴Cs, and ¹³⁷Cs. With an 8 day half-life the exposure risk associated with the release of ¹³¹I is well passed. Protection from the release of ¹³¹I was effectively managed through evacuation and monitoring of food stuffs in the period immediate following the accident. The primary radionuclide of concern for long term potential radioactive exposure to humans and the environment is therefore the medium lived ¹³⁷Cs isotope ($T_r = 30.0$ y) and to a lesser extent the short lived ¹³⁴Cs isotope ($T_r = 2.0$ y)

Radiocesium

The fissioning of both ²³⁵U and ²³⁹Pu results in both the release of a large amount of energy and the formation of a vast number of radioactive fission fragments ranging from ⁷²Zn to ¹⁵⁸Gd (Johnson, 2017). Radioactive decay of the fission fragments produces daughter radionuclides referred to as fission products. The isotopes of cesium produced within a nuclear reactor as fission products are provided in the following table with their respective half-lives.

Cesium Isotope	Half-life
¹³³ Cs	Stable
¹³⁴ Cs	2.06 years
¹³⁵ Cs	2.3×10^6 years
¹³⁶ Cs	13.16 days
¹³⁷ Cs	30.08 years

Table 1: Isotopes of Cesium Fission Products

Only ¹³⁷Cs and ¹³⁴Cs tend to be a radiation exposure concern over their environmental lifespan. ¹³⁴Cs is yielded in low amounts from fission 0.0000121% per fission) and is primarily produced due to the ¹³³Cs neutron capture reaction. ¹³³Xe (T_{1/2} 5.2 days) is yielded at 6.61% per fission and decays to ¹³³Cs. ¹³³Cs has a thermal neutron capture cross section of 30.3 barns. Given enough time and thermal neutrons the isotope ¹³⁴Cs is produced. The amount of ¹³⁴Cs present within a nuclear reactor fuel core depends on the age of the fuel and fuel composition as it relies on the build up of ¹³³Cs in the fuel. With a moderate radioactive half-life ¹³⁷Cs emits a high energy beta particle in transition to daughter ¹³⁷mBa (T_r = 2.55 min) which in turn emits a high energy gamma radiation (661 keV).

 137 Cs is yielded directly with the greatest fission yield of the cesium fission isotopes at 6.34% per fission of 235 U. 137 Cs is considered the major contributor of man-made environmental

radiation doses to living organisms (NCRP, 2006). All radioactive cesium behaves effectively chemically identical to stable cesium. As an alkali metal (i.e. Na⁺, K⁺, Rb⁺) Cs⁺ is often taken up by living organisms. Cs will accumulate in muscle tissues of animals when uptaken. Notable releases of ¹³⁷Cs to the environment have occurred from nuclear power plant accidents and atmospheric nuclear weapons testing. The Chernobyl nuclear power plant accident and the FDNPS accident resulted in releases of 85 PBq (Steinhauser et al, 2014) and 19-24 PBq (Aoyama et al. 2016), respectively of ¹³⁷Cs. In comparison, atmospheric nuclear weapons tests performed by the United States and Soviet Union in the 1950's and 1960's input an estimated total of 948 PBq of ¹³⁷Cs into the biosphere (NCRP, 2006).

The ratio of the activity inventory of ¹³⁴Cs and ¹³⁷Cs released from the FDNPP at the time of the accident was estimated to be 0.98 (Steinhauser et al. 2014). Immediately following the FDNPP accident the activities of ¹³⁴Cs and ¹³⁷Cs in the environment were very similar. At seven years beyond the accident ¹³⁴Cs has experienced nearly four half lives of decay and ¹³⁷Cs remains as the major contributor to environmental radiation doses.

Forest Ecosystem

Deposition of radiocesium from the FDNPP accident occurred primarily over a 40 km by 10 km area extending northwest of the power plants. Areas impacted included residential zones, agricultural areas, and natural and artificial (plantation) forests. The impacted region contains many artificial and natural forests which due to their total area comprise a majority of the impacted land type. Currently, within the evacuation zones large scale decontamination is taking place in residential and agricultural areas (Yoschenko, 2017). The decontamination in uninhabited forested areas are not a decontamination priority as there is limited direct exposure

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hazard to people. However, monitoring and understanding of the movement of radiocesium within forest ecosystems can provide value in evaluating future forestry management decisions.

While tree species comprise a majority of the total standing biomass in the forests, a biomass of understory plants exist that uptake radiocesium. The objective of this research was to examine the role of plant root distribution and radiocesium distribution in soils on understory plant transfers of radiocesium. However, a correlation between exchangeable radiocesium, root surface area and plant activity were not evident. Additional soil parameters are required to adequately describe plant uptake.

METHODS

2.1 Sampling Locations

Table 1 provides the locations where samples were collected and the estimated deposition of ¹³⁷Cs as estimated from a 2013 airborne dose rate survey (Nuclear Regulation Authority, 2013) as well as the predominate soil type at the location. Figure 1 displays the location of sampling locations overlaying the deposition estimates.

Location ID	Forest Type	Coordinates of Location (WGS84)	2013 ¹³⁷ Cs Deposition Density Estimate (Bq/m ²)	Soil Type
Tomioka hinoki	Cypress	37.37419, 140.9946	1.5E+06	Allophanic Black Soil
Yamikiya	Sugi	37.5884, 140.7108	6.6E+05	Allophanic Black Soil
Tomioka	Sugi	37.36942, 140.9461	4.9E+05	Brown forest soil
Juman-yama	Sugi	37.4407, 140.9130	4.7E+05	Brown forest soil
Tsushima	Sugi/Pine	37.5528, 140.7890	1.9E+06	Allophanic Black Soil

Table 2: General Sampling Location Characteristics



Figure 1: Sampling Locations and ¹³⁷Cs Deposition Density.

This figure was created using the open source GIS software, QGIS (2018). The Sample Location coordinates were provided courtesy of V. Yoschenko. The March 11, 2013 Deposition Density Data is downloaded via the Nuclear Regulatory Authority (2013) report. GIS data can be downloaded in the WGS84 coordinate system.

2.2 Sample Selection

Plants samples that were relatively isolated were chosen to decrease the influence of nonselected plants on determining plant root surface area. In most cases plants were selected such that no other understory plants were within 30-cm, and were a minimum distance of 2-m from trees. In cases where a large grouping of the same species were found in a location, the plant sample was collected regardless of the lack of isolation. There is an abundance of vegetation at all the sampling locations. The ability to ensure the roots from only the intended plants is limited. In some cases the roots from the sampled plant were unique in color and form, and could be visually separated from other roots.



Figure 2: A plant (D3) selected for sampling: *Polystichum ovatopaleaceum* var. *coraiense*.

Once a plant sample was identified for sampling, the aboveground portion of the plant was collected. The surface layer below the plant, consisting primarily of organic litter, was cleared. A pair of steel hand clippers was used to cut the plant from the root stalk above the soil surface. The plant sample was placed in a plastic bag and labelled with date and identification number.

Following above ground plant sample collection, samples of the soil through the first 15cm of depth below ground surface were collected. Samples were collected using an ICT International 0192K1 Root Auger (Armidale, New South Wales, Australia), which allows for collection of a nearly undisturbed 15-cm soil layer and can push the soil core back out of the device by means of a hand crank. The root auger was centered over the location of the sampled plant and driven into the ground by hand until maximum depth below the ground was achieved (15-cm), then the device was removed from the ground. The core was then pushed out in 1-cm intervals and each interval was partitioned from the core using a combination of a sharp steel knife and steel clippers. Care was taken to ensure all roots belonging to each partition were collected. In some cases, the soil layers experienced compression resulting in fewer than a total of 15 samples per core. Each partition was collected in a stainless-steel pan and transferred to a plastic sample bag labeled with appropriate plant identification and representative depth. The pan was cleaned with paper towel between partition collection.



Figure 3: Root auger, collection pan, steel clippers, and garden knife.



Figure 4: Soil core collection and partition into 1-cm intervals. The ICT International 0192K1 Root Auger.

2.4 Sample Processing

The collected plant sample was brought to the lab and thoroughly washed under tap water being careful to remove any attached soil particles. Following the wash, plant samples were placed into stainless steel sample pans and dried in an oven at 100°C for 24 hours. The total mass of the dried plant and pan was recorded. Dried plant material was cut up and placed into a 100 mL U-8 plastic sample counting container (Figure 5). The total dry plant mass was recorded. In one case the plant sample (D12) volume was greater than the capacity U-8 container and the entire sample was homogenized, and an aliquot was selected and placed into the counting container.



Figure 5: Typical 100 mL U-8 Sample Container for gamma spectroscopy using a High Purity Germanium detector. Soil, vegetation and aqueous extractions were counted in these containers.

The masses of the total undried soil partitions were determined using a gram scale and stainless steel pan. Each interval was homogenized by mixing with a steel spoon. From the total sample an approximately 10 g soil aliquot was removed. Any roots found within the aliquot were returned to the bulk sample using tweezers. Each soil aliquot was dried at 100°C for a minimum of 24 hours. Total dry soil mass (d.w.) of the aliquot was recorded and was also determined between each of the sequential extractions described below. Data of sample and aliquot mass is in included as results in Appendix A. Two extractions were performed on the soil aliquots.

De-ionized Water

The extraction collected in this step contains the fraction of cations in soil of a soluble form. Dried soil aliquots were loaded into 50 mL centrifuge tubes. Forty mL of de-ionized water

was added to each centrifuge tube and set on an orbital shaker. Each sample set was shaken for 24 hours.

Following mixing, the soil and water were poured into a Nalgene vacuum flask (model 166-0045) and the water was filtered from the soil through a double layer of 45 um filters. The filtrate was transferred to a 100 mL U-8 container, with the mass of recovered solution measured on a gram scale. The soil sample was re-collected from the filter setup by rinsing with D.I water. The top filter of the filter set-up was discarded after use, and the built-in filter set up was used for an entire sample set consisting of about 10 samples. The re-collected soil aliquot was placed in a 100°C oven and allowed to dry for 24 hours prior to the next extraction.



Figure 6: Thermo Scientific[™] Nalgene[™] Rapid-Flow[™] Sterile Disposable Filter Unit with 0.45 um PES filter + 0.45 um Omnipore Membrane Filter.

Ammonium acetate (NH₄Ac) extraction

The extraction collected in this step contains the fraction of cations bound to exchangeable sites of soil minerals. The soil aliquot, having dried, was mixed with 40 mL of 1M ammonium acetate (NH₄Ac) and shaken for 2 hours on the orbital shaker seen in Figure 7. Following mixing the solution and soil were filtered using a vacuum filter flask set-up (Figure 6). The soil aliquot was again recovered from the filter set up, and oven dried for 24 hours. The recovered filtrate was placed into sample counting containers in preparation for gamma spectroscopy.

Slowly Available or Fixed Radiocesium

The dried soil was transferred to a U-8 sample container and labeled for gamma spectroscopy analysis. The activity of radiocesium remaining in the soil aliquot following the two extractions represents the least soluble or exchangeable sources of radiocesium for plant uptake.



Figure 7: An orbital shaker with timer function used for mixing soil and extraction solutions.

All dried soil, dried plant material, and extraction solutions were analyzed via gamma spectrometry for ¹³⁷Cs and ¹³⁴Cs activity. Measurements were completed using a CANBERRA GC3018 high purity germanium (HPGe) detector. The time of measurement was set to ensure a statistical uncertainty of less than 10%.

It should be noted that it is unknown if the calibration of the instrument was adjusted for the differing geometries of the samples counted. For example, 40 mL of solution versus a few grams of plant matter would have different counting efficiencies. For this reason, the results presented are comparable to each other but may not correspond directly to other results using varying calibration.

2.5 Root Surface Area Measurements

A device to aid in the separation of roots from soil was constructed at the Institute of Environmental Radioactivity (IER).. In general, soil samples containing plant roots are loaded into a vertical PVC pipe—water is added and air is used to agitate the soil and water. After a period of agitation the water is turned on and filtered through a mesh screen. Plant roots are collected on the mesh screen and then further processed.



Figure 8: Root washing apparatus.



Figure 9: From root ball to root slide: A scanned image of laminated roots is converted to an entirely black and white image . Image processing and counting of the number of black pixels is performed with ImageJ software.

Roots separated from soil were placed between sheets of thermal lamination paper and sent through a lamination machine. Roots larger than 5 mm in diameter were set aside as these were too large to be fed through the lamination machine.

The prepared root slides were then scanned using a digital scanner (Ricoh MP C3004) at 600 dots per inch resolution and saved in a lossless image format (i.e. ".tiff"). The larger roots that had been set aside and not included in the lamination were placed on the scanner bed and included in the scanned image. Scanned imaged were processed and analyzed for two dimensional root surface area with ImageJ software.

An open source software platform designed for biological-image analysis known as "ImageJ" (Schindelin, J et al. 2012) was used in determining the two dimensional area of roots found on each root slide. A 256 color image of the roots is converted to a black and white image (Figure 1). Pixels above a color threshold of 155 become black pixels (000) and white background remains as white pixels (255). The number of black pixels in each image was counted and represents the two-dimensional root surface area belonging to a soil interval. Conversion from pixels to units of square centimeter are possible given the resolution of each image is known, in this case 600 dpi.

Metal wires of known length and diameter (diameters of wire ranged from 0.1 mm to 2 mm) were clipped and bent to approximate typical roots and a laminated slide was prepared of the wires. This "standard" image was then processed in the same manner as the root slides. Using the default values (threshold at 155) the surface area measured through ImageJ analysis was compared to the calculated 2-d area for the wires and found to be within 1%.

RESULTS

3.1 Deposition

Table 2 provides a comparison of two estimates of the amount of FDNPS ¹³⁷Cs deposited at the sampling locations: Radiocesium deposition estimates conducted by the Nuclear Regulation Authority in March 2013 (Nuclear Regulation Authority, 2013) and radiocesium deposition estimated by the 15-cm soil core samples collected for this research. The 2017 data has been decay corrected to allow for comparison with the March 2013 data. The 2017 deposition area activity is calculated by summing the total activity in each ~15 cm soil core and dividing by the area sampled via the root auger (an 8-cm diameter cylinder).

Location	2013 Deposition Density (Bq/m ²)	2017 Decay Corrected Estimated Average Unit Area Activity Density (Bq/m ²)	Soil Type	Percent of Airborne Survey Estimate
Tomioka hinoki	1.5×10^{6}	2.5×10^{6}	Allophanic black Soil	168%
Yamikiya	6.6×10^{5}	1.0×10^{6}	Allophanic black Soil	155%
Tomioka sugi	4.9×10^{5}	1.1×10^{6}	Brown forest soil	228%
Juman-yama	4.7×10^{5}	8. 4×10^5	Brown forest soil	179%
Tsushima Pine/Sugi	1.9×10^{6}	1.7×10^{6}	Allophanic black Soil	91%

 Table 2: Deposition Density Results Comparison

3.2 Slowly Available and Exchangeable Vertical Radiocesium Distribution in Soil

The sum of slowly available ¹³⁷Cs and exchangeable ¹³⁷Cs from top to bottom for each soil core is provided and displayed in the tables below. The soluble fraction of radiocesium resulted in less than detectable concentrations or below 4 Bq/ kg and we not measured.

The slowly available radiocesium refers to the activity remaining bound in soil following the two sequential extractions. Exchangeable radiocesium is represented by the activity of ¹³⁷Cs extracted by 1 M NH₄Ac.

Table 3 provides the average radiocesium activity found in the top 5-cm of soil layers at the sampling locations, and additional following tables provide the concentrations of radicoesium in each soil column collected and analyzed.

Location	Percent of Activity in Top 5-cm	Soil Type
Tomioka hinoki	85%	Allophanic black Soil
Yamikiya	80%	Allophanic black Soil
Tomioka sugi	90%	Brown forest soil
Juman-yama	86%	Brown forest soil
Tsushima Pine/Sugi	82%	Allophanic black Soil

Table 3: Percent of ¹³⁷Cs Activity in Top 5-cm of soil

	D1
Deptn BGS* (cm)	137Cs (Bq/kg)
0-2	1.6E+05
2-3	8.7E+04
3-4	2.5E+04
4-5	1.6E+04
5-6	1.7E+04
6-7	1.2E+04
7-9	8.6E+03
9-11	5.7E+03
11-13	3.4E+03
13-15	2.3E+03

Table 4: Total of Slowly Available and Exchangeable ¹³⁷Cs Soil Activity Concentration – Tomioka hinoki – Allophanic Black Soil

*Below Ground Surface

 Table 5: Total of Slowly Available and Exchangeable ¹³⁷Cs Soil Activity Concentration

 Yamakiya – Allophanic Black Soil

Depth BGS	D2
(cm)	¹³⁷ Cs (Bq/kg)
0-2	7.3E+04
2-3	3.4E+04
3-4	9.2E+02
4-5	6.5E+03
5-6	3.4E+03
6-7	2.5E+03
7-9	1.3E+03
9-11	1.4E+03
11-13	2.0E+04

Table 6: Total of Slowly Available and Exchangeable ¹³⁷Cs Soil Activity Concentration – Tomioka Sugi – Brown Forest Soil

Depth BGS*	D3	D4	D5	D6
(cm)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)
0-2	8.2E+04	5.5E+04	9.6E+04	9.3E+04
2-3	1.6E+04	3.0E+04	9.5E+04	4.4E+04
3-4	7.2E+03	1.2E+04	4.3E+04	1.5E+04
4-5	1.7E+03	2.5E+03	2.7E+04	8.9E+03
5-6	1.2E+03	8.7E+02	2.4E+04	2.9E+03
6-7	2.8E+03	7.2E+02	2.0E+04	9.8E+02
7-9	1.8E+03	6.4E+02	1.9E+04	6.6E+02
9-11	1.8E+03	4.0E+02	1.0E+04	5.4E+02
11-13	1.5E+03	3.0E+02	3.1E+03	4.2E+02
13-15	1.6E+03		9.6E+02	2.6E+02

Table 7: Total of Slowly Available and Exchangeable ¹³⁷Cs Soil Activity Concentration -Juman-yama – Brown Forest Soil

Depth BGS	D7	D8	D9
(cm)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)
0-2	1.3E+04	1.1E+05	2.9E+04
2-3	2.7E+03	5.2E+04	1.8E+04
3-4	2.5E+03	5.4E+04	2.2E+04
4-5	1.4E+03	3.6E+04	1.3E+04
5-6	1.3E+03	1.5E+04	7.3E+03
6-7	5.4E+02	3.4E+03	2.4E+03
7-9	3.6E+02	1.7E+03	1.7E+03
9-11	3.5E+02	1.3E+03	1.6E+03
11-13	4.6E+02	3.7E+03	
13-15	1.1E+03	2.3E+02	

Depth BGS	D10	D11	D12
(cm)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)
0-2	1.8E+05	1.2E+05	3.7E+04
2-3	1.4E+05	2.5E+04	2.7E+04
3-4	4.3E+04	8.1E+03	2.7E+04
4-5	1.5E+04	4.5E+03	2.1E+04
5-6	1.7E+04	1.8E+03	1.4E+04
6-7	8.3E+03	1.1E+03	6.6E+03
7-9	8.0E+03	1.1E+03	7.6E+02
9-11	5.8E+03	8.9E+02	3.2E+02
11-13	4.4E+03	4.6E+02	2.7E+02
13-15	2.9E+03	6.4E+02	4.2E+02

Table 8: Total of Slowly Available and Exchangeable 137Cs Soil Activity Concentration -Tsushima – Allophanic Black Soil

3.3 Exchangeable Radiocesium Vertical Distribution

The exchangeable ¹³⁷Cs activity concentrations of soil core intervals are provided in the following tables.

Table 9: Exchangeable ¹³⁷Cs Soil Activity Concentration - Tomioka hinoki – Allophanic Black Soil

Depth	D1	
BGS (cm)	¹³⁷ Cs (Bq/kg)	
0-2	6.78E+03	
2-3	3.70E+03	
3-4	2.20E+03	
4-5	1.51E+03	
5-6	1.80E+03	
6-7	1.53E+03	
7-9	1.21E+03	
9-11	9.85E+02	
11-13	4.75E+02	
13-15	3.16E+02	

Table 10: Exchangeable ¹³⁷Cs Soil Activity Concentration – Yamakiya – Allophanic Black Soil

Depth BGS	D2
(cm)	¹³⁷ Cs (Bq/kg)
0-2	6.28E+02
2-3	3.39E+02
3-4	2.45E+01
4-5	
5-6	
6-7	
7-9	
9-11	
11-13	

Table 11: Exchangeable ¹³⁷ Cs Soil Activity Concentration - Tomioka sugi – Brown F	orest
Soil	

Denth BGS	D3	D4	D5	D6		
(cm)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)		
0-2	9.48 <mark>E+02</mark>	1.48E+03	4.96E+02	4.27E+02		
2-3	1.75E+02	1.07E+03	6.17E+02	4.52E+02		
3-4	1.11E+02	2.85E+02	2.78E+02	3.59E+02		
4-5	2.83E+01		2.01E+02	2.73E+02		
5-6			2.86E+02	1.50E+02		
6-7			1.68E+02	6.64E+01		
7-9			1.51E+02			
9-11						
11-13						
13-15						

Table 12: Exchangeable ¹³⁷Cs Soil Activity Concentration – Juman-yama – Brown Forest Soil

Denth BGS	D7	D8	D9		
(cm)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)		
0-2	1.63E+02	3.53E+02	2.95E+02		
2-3	3.47E+01	3.24E+02	2.05E+02		
3-4	3.65E+01	5.01E+02	2.22E+02		
4-5	2.01E+01	4.33E+02			
5-6	1.81E+01	2.90E+02			
6-7	1.01E+01	5.75E+01			
7-9		2.93E+01			
9-11					
11-13					
13-15					

Table 13: Exchangeable ¹³⁷Cs Soil Activity Concentration – Tsushima

Depth BGS	D10	D11	D12
(cm)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)
0-2	3.11E+03	2.74E+03	1.75E+03
2-3	2.24E+03	6.13E+02	1.26E+03
3-4	1.01E+03	2.75E+02	1.20E+03
4-5	4.17E+02	1.51E+02	1.01E+03
5-6	3.23E+03	5.43E+01	5.78E+02
6-7	2.60E+02	4.23E+01	3.10E+02
7-9	2.69E+02	4.28E+01	4.18E+01
9-11			1.17E+01
11-13			
13-15			

3.5 Plant ¹³⁷Cs Activity Results

The following tables provide the activity concentrations of the sampled plants, reported by location. Reported is activity in dry weight plant material, the sample masses are provided in Appendix A. Activity concentration ranged from 640 Bq/kg to 7500 Bq/kg in dry plant material.

Sample ID	Plant ¹³⁷ Cs Activity Conc. (Bq/kg)	Soil Type
D1	5.E+03	Allophanic Black Soil
D2	2.E+03	
D3	2.E+04	
D4	1.E+04	
D5	1.E+04	
D6	1.E+04	Brown Forest Soil
D7	8.E+03	
D8	3.E+04	_
D9	2.E+04	
D10	8.E+02	
D11	5.E+03	Soil
D12	6.E+02	

 Table 14: Plant ¹³⁷Cs Activity Concentrations (dry weight)

3.6 Plant Concentration Ratios

Table 19 provides the ¹³⁷Cs concentration ratios of each analyzed plant sample set, the plant species, and sampling location. Concentration ratio, C_r is defined as the ratio of the dry ¹³⁷Cs activity concentration in dry plant to dry soil. (IAEA, 2010). The average ¹³⁷Cs soil activity concentration over the top 10-cm soil was used in calculating C_r .

Sample ID	Species	Location	Cr	Soil Type
		Tomioka		Allophanic
D1	Abies firma	cypress	0.12	Allophanic Dissis Sail
D2	Sasa palmate	Yamakiya	0.13	Black Soll
D3	Sasa palmate	_	1.47	
D4	Clerodendrum trichotomum		0.78	
	Elatostema umbellatum var.	Tomioka		
D5	majus	cedar	0.25	
	Polystichum ovatopaleaceum			Brown
D6	var. coraiense		0.55	Forest Soil
	Polystichum ovatopaleaceum			
D7	var. coraiense	Jumon	2.48	
D8	Sasa palmate	yama	0.67	
	Polystichum ovatopaleaceum	yunna		
D9	var. coraiense		1.37	
D10	Sasa palmate		0.01	Allophonic
D11	Sasa palmate	Tsushima	0.23	Black Soil
D12	Miscanthus sinensis		0.04	Diack Soli

Table 15: Species, Location, and Concentration Ratio of Sampled Plants

3.7 Correlations

The activity concentration of ¹³⁷Cs is plotted against total soil activity, exchangeable soil activity, and root surface area multiplied by exchangeable activity



Figure 10: Average 10-cm soil Activity versus Plant Activity

Figure 11: Exchangeable Soil activity versus Plant Activity (Log – Log)



A parameter of root area and exchangeable ¹³⁷Cs was calculated for each interval and summed for the entire soil core (Equation 1). A comparison of this Root Activity parameter and total plant activity is displayed on Figures 14.

Root Exchange Activity (Bq
$$\cdot$$
 cm²) = $\sum_{n=1}^{l} [2 - D \text{ Root Area}]_{i}$ (cm²) \cdot
[¹³⁷Cs_{ex} Soil Activity]_i (Bq) [1]



Figure 11: Root Exchange Activity Vs Plant Activity

DISCUSSION

4.1 Deposition Estimation

The results of ¹³⁷Cs activities in the soil at the sampling locations tend agree with the estimates produced via airborne survey (Nuclear Regulation Authority, 2013). The deposition estimates also agree with reported values from previous sampling years at the Yamakiya and Tsushima sites as reported by Yocheknko et al. (2017). The activity in soil and litter at Yamakiya in 2016 was reported as 0.78 ± 0.14 MBq m⁻² compared with the 1.0 MBq m⁻² presented here in only soil. At "Tsushima" in 2016 the soil and litter ¹³⁷Cs activity was reported as 1.6 ± 0.5 MBq m⁻². Soil samples collected in 2017 during this research resulted in soil activity of 1.6 ± 0.2 MBq m⁻². Litter containing uptake due to initial fallout interception has continually decomposed and leaching from the litter has contributed to the soil inventory of radiocesium. In 2016 the ¹³⁷Cs activity in litter was 4% of the total soil and litter activity compared with the 2014 sampling event when activity in litter approached 23% of total inventory.

The deposition estimates produced following the airborne survey were based on a correlation between measured air dose rate and the contribution to dose rate by ¹³⁷Cs activity as measured by an in-situ germanium semiconductor detector. The in-situ germanium semiconductor measured the ¹³⁷Cs activity at a 1-m height, and therefore the activity measured was influenced by soil and litter and biomass. This analysis resulted in an estimate of the total deposition of radiocesium. The agreement between deposition estimates produced during the airborne survey of 2013 and the soil sampling performed here in 2017, and previous sampling done at the sites, support a hypothesis that a majority of radiocesium is now existing in the soil

compartments within forest ecosystems. The inventory of radiocesium in tree biomass was reported at 6% in 2016 at the Yamakiya site (Yoschenko, 2017).

4.2 Fixed/Slowly Available and Exchangeable Radiocesium Distribution

More than 80% of the measured ¹³⁷Cs activity was found in the top 5-cm of the soil column (Table 2). These results are consistent with previously published values at the sites (Yoschenko, 2017). Similarly, data presented in NCRP 154 from locations around the world demonstrate that typically ~80 to 100% percent of ¹³⁷Cs activity resides in the top 10 cm of soil (NRCP 2006). The movement of radiocesium in many surface soils is limited and is mostly controlled by physical transport processes (wind, sediment movement, animal activity, soil cracking) rather than by water or groundwater transport.

The retention of radiocesium by surface layer soils is attributed to the binding of Cs⁺ to minerals comprising clay type soils. Micaceous clay minerals (such as illite or biotite) are often attributed as the primary minerals responsible for the sorption of radiocesium in soil (Absalom et al., 1999). For the soils encountered during sampling: allophanic black soil has a higher content of micaceous clay minerals than brown forest soil. Research groups have identified the physical locations within the clay mineral structures where binding takes place and which varies by clay mineral species. For the micaceous mineral illite the weathered edges of the particle or frayed edge sites (FES) provide locations where Cs⁺ bonding occurs.

Table 20 provides the average site exchangeable fraction of radiocesium extracted from soil aliquots with a 1 M NH₄Ac solution. The amount ranges from 1.2% to 5.5% of the total measured radiocesium content. The exchangeable activity represents the pool of radiocesium that provides radiocesium to the soluble or readily available pool of nutrients. The soluble fraction of

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radiocesium in the soil solution is assumed to be very low, though only two samples were measured they both contained activities less than 4 Bq/kg.

The designation also used in describing the forms of radiocesium found in soils is the same as soil potassium (K) content in soils. The amounts of soil potassium available to plants is vital to efficient agricultural crop production and as such it is routinely measured. Soil K content is commonly grouped into three forms: unavailable or fixed, slowly available, and exchangeable (NH₄Ac) or readily available (water soluble) (Kasier et al, 2016).

It should also be noted that ${}^{134}Cs$ activity was also quantified in the gamma spectrometry measurements of all samples. The ratio of ${}^{134}Cs$ to ${}^{137}Cs$ activity is consistent across nearly all samples and is characteristic of the Fukushima release at the time of measurement. At the time of measurement ${}^{134}Cs/{}^{137}Cs$ averaged 0.12 across all samples.

Location	Exchangeable ¹³⁷ Cs Fraction in Soil	Soil Type
Tomioka - hinoki	5.5%	Allophanic Black Soil
Yamakiya	1.9%	Allophanic Black Soil
Tomioka - Sugi	2.6%	Brown forest soil
Jumanyama	1.2%	Brown forest soil
Tsushima	3.2%	Allophanic Black Soil

Table 16: Exchangeable Fraction of ¹³⁷Cs in Top 10-cm of Soil

4.3 Concentration Ratios

Concentration ratios of the sampled understory plants ranged from 0.01 to 2.5.

Concentration ratios between the same species (*Sasa palmate* or *Polystichum ovatopaleaceum*) collected at different sampling sites demonstrated a very broad range. The range of concentration ratios of samples from a single sampling site sis narrower than the when comparing across all samples, as seen in Table 2.

To provide for consistency when developing soil to plant transfer factors the International Union of Radioecologists specifies the averaging of a radionuclide concentrations over a standardized soil layer of 10-cm for grasses and 20-cm for all other crops (IAEA, 2010). The understory species sampled for this research tend to have rooting depths more akin to grass species, and in some cases are grass (*Miscanthus sinensis, Sasa palmate*), as such the values presented in Table 2 use the average activity concentration of ¹³⁷Cs over the first 10-cm of soil in the calculation of concentration ratios. The standardized transfer factor method also calls for the sampling of plants at the end of their growth period. Plant samples presented here were collected within a four week period spanning the first week of July to the first week of August, 2017. It is possible that the plant activity concentration of sampled plants could change over the remainder of the growing season that extends into the month of October.

Plant concentration ratios are assumed to be governed by root uptake of a radionuclide, with the contribution of foliar uptake and uptake via resuspension of soil particles of minor significance. At seven years post-accident these assumptions are based on data indicating that the majority of radiocesium is now found in soil layers and plant uptake and return of radiocesium to soil is approaching a constant rate (Yoschenko 2017). The flow of radiocesium to plants is also assumed to be much smaller than the total pool radiocesium found within the soil. For conditions such as these, the IAEA and NCRP has reported values of concentration ratios which provide a basis for radiological risk assessments of both planned and accidental releases. Literature values of concentration ratios have significant ranges spanning orders of magnitude among the same species grown in different soils (NCRP, 2006).

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4.4 Correlations

In observing the relationship between soil activity, root surface area and total plant activity of ¹³⁷Cs possible trends appear, and it is likely that there exists some site-specific correlations that could be used in predicting plant uptake. Additional sampling would be required to develop a model.

There is a marked difference in the plant concentration ratios between the two soil groups sampled from: Allophanic black soil (Average C_r : 0.1, n= 5) and Brown forest soil (Average C_r : 1.0, n =7). Soil typing was performed using an online mapping software (NARO, 2018). Allophanic soil is marked by its high clay content and minerals of allophane and imogolite, (NARO, 2018) both aluminum silicate clay minerals. While brown forest soils tend to have higher organic matter content they also contain clays but in smaller fraction compared with allophanic soils. These soil typings are broad and additional soil typing would be required to confirm the soil types to include mineral species, soils pH, particle size distribution and cation exchange capacity.

No statistically significant correlation between root surface area, radiocesium in soil and plant uptake was developed from the data analyzed. Possibly, additional soil characteristics masks the effect of root distribution on uptake. Also, the root surface area and radiocesium are distributed primarily in the top 5-cm of soil, making a correlation difficult to establish. The method employed to determine root surface area distribution assumes that a significant majority of the sampled roots belong to the sampled plant and as there are many roots in the forest soils and certainly some fraction of sampled roots does not belong to the sampled plant. Similarly, roots of interest certainly extend beyond the sampled portion yet sampled roots are assumed to represent the distribution of plant roots.

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Additionally, Konoplev et al. (1996) demonstrated that in using only two extractions (water and NH₄Ac) data from ion concentrations could represent the frayed edge site concentrations and the equilibrium between exchangeable and soluble ions in soil solution. The movement of plant nutrients from soil into plant cell walls is understood to be governed primarily by diffusion and so the concentrations of competing ions play a significant role in the quantity of radiocesium incorporated into a plant (Bujtas et al., 2000).

CONCLUSION

Concentration ratios presented here provide evidence of a wide range of $C_{\rm r}$ for plant uptake at some locations in the contaminated forests of Fukushima. The range of concentration ratios (d.w. plant activity/d.w. soil activity) of 0.01 to 2.5 falls within the range of values observed for herbs and pasture crops grown in clay soils recommended by the IAEA Parameter handbook. With the pool of radiocesium found in the soil now approaching a maximum and in an equilibrium state, the ability to predict the uptake into plants and animals for the long term can be developed. However, the hypothesis that root surface area and exchangeable soil concentrations of radiocesium in soils describe plant uptake of radiocesium was not proven.

Concentration ratio appears to depend more on soil type than other parameters measured here (root area, fixed or exchangeable radiocesium distribution). Two soil types were represented by the samples: Allophanic black soil, and brown forest soil. Both soils appear to retain radiocesium in upper layers but different ranges of plant concentration ratios were observed within the soil types. Plants sampled in brown forest soils had an average C_r of 1.0 while plants sampled in allophanic black soils had an average C_r of 0.1. The physiochemical parameters of the soil that influence nutrient exchange including soil pH, particle sizes, cation exchange capacity and nutrient availability may mask the effect of the parameters measured here. Greater than 80% of all measured soil radiocesium was found within the top 5 cm of surface soils. The retention of radiocesium by surface soils is such that even in the conditions of the wet montane slopes of Fukushima migration of radiocesium downward by infiltration is limited.

REFERENCES

Absalom, J.; Young, S.; Crout, N.; Nisbet, A.; Woodman, R.; Smolders, E.; & Gillett, A. (1999). Predicting Soil to Plant Transfer of Radiocesium Using Soil Characteristics. *Environmental Science and Technology*, 1999, *33* (8), pp 1218–1223 DOI: 10.1021/es9808853

Aoyama, M., Kajino, M., Tanaka, T.Y., Sekiyama, T.T., Tsumune, D., Tsubono, T., Hamajima, Y., Inomata, Y., & Gamo, T. (2016). ¹³⁴Cs and ¹³⁷Cs in the North Pacific Ocean derived from the March 2011 TEPCO Fukushima Dai-ichi Nuclear Power Plant accident, Japan. Part two: estimation of ¹³⁴Cs and ¹³⁷Cs inventories in the North Pacific Ocean. *Journal of Oceanography*, 72, 67–76. DOI: 10.1007/s10872-015-0332-2

Bujtas, K., & Szerbin, P. (2000). Soil-to-Plant Transfer of Radionuclides. *The Environmental Challenges of Nuclear Disarmament*, 205-214. Kluwer Academic Publishers.

Hashimoto, S., Ugawa, S., Kanko, K., & Shichi, K. (2012) The total amounts of radioactively contaminated materials in forests in Fukushima, Japan. *Scientific Reports*, 2, 416. DOI:10.1038/srep00416

International Atomic Energy Agency (IAEA). (2010). Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments. Technical Report Series No. 472. Vienna.

Johnson, T. (2017). Introduction to Health Physics. New York. McGraw Hill Education.

Kaiser, D., Rosen, & John A. Lamb. (2016) Potassium for Crop Production. University of Minnesota Extension. FO-6794-D.

Konoplev, A. V., Drissner, J., Klemt, E., Konopleva, I. V., & Zibold, G. (1996). Parameterization of radiocaesium soil-plant transfer using soil characteristics. 26th Annual ESNA/IUR Meeting: Soil-Plant Relationships, (p. 197). Austria

Nakahara, S., & Ichikawa, M. (2013). Mortality in the 2011 Tsunami in Japan. *Journal of Epidemiology*, 23(1), 70–73. http://doi.org/10.2188/jea.JE20120114

NARO. (2018). Soil Mapping of Japan. April 4, 2018. Retrieved from: https://soilinventory.dc.affrc.go.jp/figure.html

National Council on Radiation Protection and Measurements (NCRP). (2006). NCRP Report No. 154. Cesium-137 in the Environment: Radioecology and Approaches to Assessment and Management. Bethesda, MD.

Nuclear Regulation Authority, 2013. Results of the Airborne Monitoring in the evacuation directed zones. http://radioactivity.nsr.go.jp/en/contents/7000/6936/24/270_0513.pdf (Accessed 2018, March 15).

QGIS Development Team (2018). QGIS Geographic Information System. Open Source Geospatial Foundation Project. Retrieved from: http://qgis.osgeo.org.

Schindelin, J.; Arganda-Carreras, I. & Frise, E. et al. (2012), "Fiji: an open-source platform for biological-image analysis", *Nature methods* **9**(**7**): 676-682, PMID 22743772, DOI:10.1038/nmeth.2019

Steinhauser, G., Brandl, A., & Johnson, T. (2014) Comparison of the Chernobyl and Fukushima nuclear accidents: A review of the environmental impacts. *Science of The Total Environment*, Vol. 470–471, 800-817. DOI: 10.1016/j.scitotenv.2013.10.029.

United States Geological Survey. (2016). M 9.1 - near the east coast of Honshu, Japan. Retrieved from:

https://earthquake.usgs.gov/earthquakes/eventpage/official20110311054624120_30#executive. November 7, 2016.

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). (2014) VOLUME I Scientific Annex A: Levels and effects of radiation exposure due to the nuclear accident after the 2011 great east-Japan earthquake and tsunami. UNSCEAR 2013: Report to the General Assembly with Scientific Annexes. Retrieved from http://www.unscear.org/docs/reports/2013/13-85418_Report_2013_Annex_A.pdf

Yoschenko, V., Takase, T., Hinton, T.G., Nanba, K., Onda, Y., Konoplev, A., Goto, A., Yokoyama, A., & Keitoku, K. (2017). Radioactive and stable cesium isotope distributions and dynamics in Japanese cedar forests. *Journal of Environmental Radioactivity*, 186, 34-44. DOI: 10.1016/j.jenvrad.2017.09.026.

Zare, M. & Afrouz, S. (2012) Crisis Management of Tohoku; Japan Earthquake and Tsunami 11 March 2011. *Iranian Journal of Public Health*, Vol. 41, 12-20.

APPENDIX A

Results Tables

						S	ample: D1								
	Japanese Fir (Abies Firma). Location: Tomioka														
Sample name			Cs-134 (Bq/kg)		Cs-137 (Bq/kg)			err/Activity		Aliquot	T. (.) 0.11 0		Root Surface Area		
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	Cs134/Cs137	Cs-134	Cs-137	Sample dry mass (g)	Dry mass (g)	Live time (s)	Pixel count (600 dpi image)		
170623	leaves	D1Leaves	2.21E+02	8.20E+00	1.71E+03	2.96E+01	0.13	3.7	1.7	2.71		31500			
170623	wood	D1Wood	5.82E+02	1.73E+01	3.65E+03	4.15E+01	0.16	3.0	1.1	1.11		54000			
170623	soil 0-3 cm	D1 0-3	1.87E+04	2.42E+02	1.55E+05	1.03E+03	0.12	1.3	0.7	1.7	32.2	3600	3.30E+06		
170623	soil 3-4 cm	D1 3-4	1.02E+04	1.13E+02	8.67E+04	4.93E+02	0.12	1.1	0.6	2.6	27.2	5800	1.38E+06		
170623	soil 4-5 cm	D1 4-5	3.07E+03	4.65E+01	2.52E+04	1.91E+02	0.12	1.5	0.8	3.25	26.0	9000	1.86E+06		
170623	soil 5-6 cm	D1 5-6	1.92E+03	3.71E+01	1.63E+04	1.58E+02	0.12	1.9	1.0	3.09	43.5	9000	2.29E+06		
170623	soil 6-7 cm	D1 6-7	2.04E+03	2.88E+01	1.71E+04	1.22E+02	0.12	1.4	0.7	3.24	30.3	15000	1.27E+06		
170623	soil 7-8 cm	D1 7-8	1.44E+03	2.35E+01	1.23E+04	9.92E+01	0.12	1.6	0.8	2.91	36.1	18000	1.43E+06		
170623	soil 8-10 cm	D1 8-10	1.04E+03	1.38E+01	8.48E+03	5.81E+01	0.12	1.3	0.7	4.64	51.6	24000	1.80E+06		
170623	soil 10-11 cm	D1 10-11	6.82E+02	1.70E+01	5.64E+03	6.99E+01	0.12	2.5	1.2	3.91	32.1	12600	5.16E+05		
170623	soil 11-13 cm	D1 11-13	4.01E+02	6.56E+00	3.38E+03	2.76E+01	0.12	1.6	0.8	5.52	99.6	36000	1.33E+06		
170623	soil 13-15 cm	D1 13-15	2.83E+02	5.44E+00	2.31E+03	2.22E+01	0.12	1.9	1.0	3.88	41.2	54000	3.10E+05		

	Sample: D1: NH4Ac Extraction														
Sa	Sample name		Cs-134 (Bq/kg)		Cs-137	' (Bq/kg)	Ca124/Ca127	err/A	ctivity	NH4Ac Mass	Live time (c)				
Date	Part		Activity	Act. err	Activity	Act. err	US134/US137	Cs-134	Cs-137	(g)	Live time (s)				
170623	0-3 cm NH4Ac	D1 AC0-3	3.47E+01	1.09E+00	2.73E+02	2.82E+00	0.13	3.1	1.0	42.2	54000				
170623	3-4 cm NH4Ac	D1 AC3-4	3.14E+01	1.28E+00	2.49E+02	3.51E+00	0.13	4.1	1.4	38.7	34000				
170623	4-5 cm NH4Ac	D1 AC4-5	2.48E+01	1.13E+00	1.95E+02	3.05E+00	0.13	4.5	1.6	36.8	36000				
170623	5-6 cm NH4Ac	D1 AC5-6	1.48E+01	8.49E-01	1.18E+02	2.27E+00	0.13	5.8	1.9	39.5	42000				
170623	6-7 cm NH4Ac	D1 AC6-7	1.79E+01	8.13E-01	1.47E+02	2.17E+00	0.12	4.5	1.5	39.6	54000				
170623	7-8 cm NH4Ac	D1 AC7-8	1.56E+01	4.90E-01	1.12E+02	1.49E+00	0.14	3.1	1.3	39.7	64700				
170623	8-10 cm NH4Ac	D1 AC8-10	1.82E+01	9.16E-01	1.42E+02	3.26E+00	0.13	5.0	2.3	39.4	22600				
170623	10-11 cm NH4Ac	D1 AC10-11	1.26E+01	5.60E-01	9.84E+01	1.93E+00	0.13	4.4	2.0	39.2	44000				
170623	11-13 cm NH4Ac	D1 AC11-13	8.37E+00	4.41E-01	6.81E+01	1.48E+00	0.12	5.3	2.2	38.5	54000				
170623	13-15 cm NH4Ac	D1 AC13-15	3.16E+00	2.90E-01	2.53E+01	8.93E-01	0.12	9.2	3.5	48.5	54000				

							Sample: D2								
	Sasa Palmate. Location: Tomioka														
Sa	Sample name		Cs-134 (Bq/kg)		Cs-137	(Bq/kg)		err/Activity		Aliquot	Total Soil Sample	Line Correctory	Root Surface Area		
Date	Part	- Sample ID	Activity	Act. err	Activity	Act. err	US134/US137	Cs-134	Cs-137	mass (g)	Dry mass (g)	Live time (s)	Pixel count (600 dpi image)		
170707	sasa plant	D2	3.28E+02	1.32E+01	2.19E+03	2.92E+01	0.15	4.0	1.3	1.43					
170707	soil 0-7 cm	D2 S0-7	8.84E+03	1.23E+02	7.29E+04	5.15E+02	0.12	1.4	0.7	3.1	36.9	3700	6.65E+05		
170707	soil 7-8 cm	D2 S7-8	4.08E+03	6.03E+01	3.40E+04	2.56E+02	0.12	1.5	0.8	3.6	26.4	6000	3.77E+05		
170707	soil 8-9 cm	D2 S8-9	1.24E+02	8.44E+00	8.93E+02	2.88E+01	0.14	6.8	3.2	4.1	33.2	12000	4.45E+05		
170707	soil 9-10 cm	D2 S9-10	8.17E+02	1.49E+01	6.49E+03	6.02E+01	0.13	1.8	0.9	4.3	29.9	18000	2.12E+05		
170707	soil 10-11 cm	D2 S10-11	3.87E+02	1.07E+01	3.39E+03	4.38E+01	0.11	2.8	1.3	4.2	38.9	18000	3.56E+05		
170707	soil 11-12 cm	D2 S11-12	3.02E+02	6.49E+00	2.46E+03	2.60E+01	0.12	2.2	1.1	4.5	28.1	36000	2.69E+05		
170707	soil 12-13 cm	D2 S12-13	1.51E+02	5.29E+00	1.26E+03	1.90E+01	0.12	3.5	1.5	4.3	38.4	36000	4.50E+05		
170707	soil 13-14 cm	D2 S13-14	1.74E+02	5.49E+00	1.40E+03	2.03E+01	0.12	3.2	1.5	3.9	42.7	36000	1.40E+05		
170707	soil 14-15 cm	D2 S14-15	2.37E+03	1.81E+01	1.96E+04	7.64E+01	0.12	0.8	0.4	4.1	26.9	36000	1.33E+05		

	Sample: D2 NH4Ac Extraction													
Sa	Sample name		Cs-134 (Bq/kg)		Cs-137 (Bq/kg)		00124/00127	err/Activity		NH4Ac Mass				
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	05134/05137	Cs-134	Cs-137	(g)	Live time (s)			
170707	NH4 acetate 7-8 cm	D2 AC7-8	1.87E+01	7.40E-01	1.39E+02	1.93E+00	0.13	4.0	1.4	35.99	54000			
170707	NH4 acetate 8-9 cm	D2 AC8-9	ND		1.12E+01	7.23E-01			6.5	37.14	64000			
170707	NH4 acetate 9-10 cm	D2 AC9-10	ND		1.0.20	6.50E-01				39.67	64000			
170707	NH4 acetate 10-11 cm	D2 AC10-11								38.89				
170707	NH4 acetate 11-12 cm	D2 AC11-12								38.3				
170707	NH4 acetate 12-13 cm	D2 AC12-13								36.36				
170707	NH4 acetate 13-14 cm	D2 AC13-14								36.33				
170707	NH4 acetate 14-15 cm	D2 AC14-15								38.38				

					Sasa	Palmate.	Location: Ton	nioka					
Sar	Sample name		Cs-134	(Bq/kg)	Cs-137	′ (Bq/kg)	Cs134/Cs137	err/A	ctivity	Aliquot Sample	Total Soil Sample Dry	Live time (s)	Root Surface Area
Date	Part		Activity	Act. err	Activity	Act. err		Cs-134	Cs-137	dry mass (g)	mass (g)		Pixel count (600 dpi image)
170707	plant	D3 plant	2984	54.08	20800	172.4	0.14	1.8	0.8	9.5		4600	
170707	soil	D3 S0-6	9.83E+03	8.81E+01	8.10E+04	3.74E+02	0.12	0.9	0.5	2.7	25.9	9000	3.91E+06
170707	soil	D3 S6-7	1.97E+03	2.32E+01	1.61E+04	9.70E+01	0.12	1.2	0.6	4.1	14.4	18000	7.91E+05
170707	soil	D3 S7-8	8.30E+02	1.51E+01	7.10E+03	6.27E+01	0.12	1.8	0.9	4.3	33.4	18000	8.35E+05
170707	soil	D3 S8-9	2.08E+02	7.23E+00	1.66E+03	2.78E+01	0.13	3.5	1.7	5.4	33.8	18000	5.86E+05
170707	soil	D3 S9-10	1.36E+02	5.33E+00	1.16E+03	1.99E+01	0.12	3.9	1.7	5.5	38.8	24000	6.68E+05
170707	soil	D3 S10-11	3.40E+02	6.22E+00	2.75E+03	2.52E+01	0.12	1.8	0.9	5.5	33.6	36000	5.64E+05
170707	soil	D3 S11-12	2.15E+02	5.35E+00	1.78E+03	2.11E+01	0.12	2.5	1.2	5.1	37.9	36000	5.50E+05
170707	soil	D3 S12-13	2.20E+02	5.44E+00	1.76E+03	2.02E+01	0.12	2.5	1.1	5.3	41.7	36000	2.99E+05
170707	soil	D3 S13-14	1.79E+02	4.99E+00	1.49E+03	1.87E+01	0.12	2.8	1.3	5.5	45.0	36000	4.01E+05
170707	soil	D3 S14-15	1.90E+02	4.58E+00	1.56E+03	1.79E+01	0.12	2.4	1.1	6.1	44.5	36000	3.21E+05
				Samr	ole: D3 NH	44Ac Extr	action						

				Samp	ole: D3 N⊦	HAC Extra	action				
Sar	nple name		Cs-134	(Bq/kg)	Cs-137	' (Bq/kg)		err/A	ctivity	NH4Ac	Live time (s)
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	Cs134/Cs137	Cs-134	Cs-137	Mass (g)	
170707	NH4 acetate	D3 AC0-6	1.05E+01	7.48E-01	8.65E+01	1.67E+00	0.12	7.1	1.9	36.8	54000
170707	NH4 acetate	D3 AC6-7	3.84E+00	5.63E-01	2.65E+01	9.63E-01	0.15	14.7	3.6	38.26	60000
170707	NH4 acetate	D3 AC7-8	ND		2.05E+01	8.43E-01			4.1	37.38	64000
170707	NH4 acetate	D3 AC8-9			9.94E+00	6.83E-01			6.9	36.86	64000
170707	NH4 acetate	D3 AC9-10								35.88	
170707	NH4 acetate	D3 AC10-11								36.78	
170707	NH4 acetate	D3 AC11-12								38.65	
170707	NH4 acetate	D3 AC12-13								37.56	
170707	NH4 acetate	D3 AC13-14								37.5	
170707	NH4 acetate	D3 AC14-15								35.18	

						Samp	le: D4						
		ŀ	larleguin	Glory-bov	ver (Clero	dendrum	trichotomur	nasa). L	ocation:	Tomioka			
San	nple name	Sample ID	Cs-134	(Bq/kg)	Cs-137	(Bq/kg)	Cs134/Cs137	err/A	ctivity	Aliquot Sample	Total Soil Sample	Live time	Root Surface Area
Date	Part		Activity	Act. err	Activity	Act. err		Cs-134	Cs-137	dry mass (g)	Dry mass (g)	(s)	Pixel count (600 dpi image)
170707	plant	D4 plant	1401	49.11	10998	182.6	0.13	3.5	1.7	0.8		18000	
170707	soil	D4 S0-7	6.29E+03	4.62E+01	5.30E+04	1.99E+02	0.12	0.7	0.4	3.10	16.1	18000	6.44E+05
170707	soil	D4 S7-8	3.41E+03	2.95E+01	2.89E+04	1.27E+02	0.12	0.9	0.4	4.25	22.8	18000	4.69E+05
170707	soil	D4 S8-9	1.40E+03	1.85E+01	1.15E+04	7.82E+01	0.12	1.3	0.7	4.55	23.4	18000	1.82E+05
170707	soil	D4 S9-10	3.05E+02	8.47E+00	2.47E+03	3.30E+01	0.12	2.8	1.3	5.59	52.8	18000	2.22E+05
170707	soil	D4 S10-11	1.08E+02	3.96E+00	8.75E+02	1.37E+01	0.12	3.7	1.6	5.83	44.2	36000	1.60E+05
170707	soil	D4 S11-12	8.75E+01	3.23E+00	7.16E+02	1.19E+01	0.12	3.7	1.7	6.41	46.9	36000	2.62E+05
170707	soil	D4 S12-13	8.37E+01	3.22E+00	6.36E+02	1.15E+01	0.13	3.8	1.8	6.30	33.0	36000	1.22E+05
170707	soil	D4 S13-14	4.72E+01	2.60E+00	3.96E+02	9.35E+00	0.12	5.5	2.4	6.03	49.7	36000	6.08E+04
170707	soil	D4 S14-15	3.78E+01	2.72E+00	2.98E+02	9.18E+00	0.13	7.2	3.1	5.83	42.2	30000	4.28E+04

				Samp	ole: D4 NH	4Ac Extra	action				
San	mple name	Sample ID	Cs-134	(Bq/kg)	Cs-137	(Bq/kg)	Co124/Co127	err/A	ctivity	NH4Ac	Live time (a)
Date	Part	Sample ib	Activity	Act. err	Activity	Act. err	03134/03137	Cs-134	Cs-137	Mass (g)	Live time (S)
170707	NH4 acetate	D4 AC0-7	2.01E+01	6.19E-01	1.61E+02	2.22E+00	0.12	3.1	1.4	39.03	54000
170707	NH4 acetate	D4 AC7-8	1.51E+01	5.75E-01	1.28E+02	2.07E+00	0.12	3.8	1.6	37.92	54000
170707	NH4 acetate	D4 AC8-9	5.30E+00	3.76E-01	4.16E+01	1.26E+00	0.13	7.1	3.0	38.27	54000
170707	NH4 acetate	D4 AC9-10									
170707	NH4 acetate	D4 AC10-11									
170707	NH4 acetate	D4 AC11-12									
170707	NH4 acetate	D4 AC12-13									
170707	NH4 acetate	D4 AC13-14									
170707	NH4 acetate	D4 AC14-15									

						Sam	ple: D5						
			Vipe	r grass (<i>E</i>	latostema	a umbella	tum var. maji	us). Loca	ation: To	mioka			
Sa	mple name		Cs-134	(Bq/kg)	Cs-137	′ (Bq/kg)		err/A	ctivity	Aliquot Sample	Total Soil Sample		Root Surface Area
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	CS134/CS137	Cs-134	Cs-137	dry mass (g)	Dry mass (g)	Live time (s)	Pixel count (600 dpi image)
170713	soil	D5 S0-2	1.16E+04	9.31E+02	9.56E+04	3.96E+02	0.12	8.0	0.4	5.02	21.7	5300	1.20E+05
170713	soil D5 S2-3 1.15E+04 6.33E+01 9.47E+04 2.70E+02 0.12 0.6 0.3 4.70 23.5											12000	1.31E+05
170713	soil	D5 S3-4	5.18E+03	3.16E+01	4.29E+04	1.33E+02	0.12	0.6	0.3	5.96	24.3	18000	2.93E+04
170713	soil	D5 S4-5	3.20E+03	2.41E+01	2.65E+04	1.02E+02	0.12	0.8	0.4	6.26	23.4	18000	1.98E+04
170713	soil	D5 S5-6	2.88E+03	2.87E+01	2.40E+04	1.22E+02	0.12	1.0	0.5	5.90	26.3	12000	1.77E+04
170713	soil	D5 S6-7	2.46E+03	4.57E+01	2.02E+04	1.90E+02	0.12	1.9	0.9	6.52	34.1	3800	9.19E+03
170713	soil	D5 S7-9	2.33E+03	2.11E+01	1.91E+04	8.83E+01	0.12	0.9	0.5	6.83	57.5	16000	4.25E+03
170713	soil	D5 S9-11	1.27E+03	1.46E+01	1.05E+04	6.17E+03	0.12	1.1	58.9	6.92	67.1	18000	
170713	soil	D5 S11-13	3.73E+02	7.41E+00	3.14E+03	3.01E+01	0.12	2.0	1.0	6.46	52.7	24000	
170713	soil	D5 S13-15	1.22E+02	3.66E+00	9.63E+02	1.35E+01	0.13	3.0	1.4	6.68	89.2	36000	

				Sampl	e: D5 NH	4Ac Extra	ction				
Sa	mple name	Sampla ID	Cs-134	(Bq/kg)	Cs-137	′ (Bq/kg)	Co124/Co127	err/A	ctivity	NH4Ac	Livo timo (c)
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	05134/05137	Cs-134	Cs-137	Mass (g)	Live time (S)
170713	NH4 acetate	D5 AC0-2	2.68E+01	8.41E-01	1.88E+02	2.41E+00	0.14	3.1	1.3	36.29	54000
170713	NH4 acetate	D5 AC2-3	3.18E+01	9.40E-01	2.41E+02	2.67E+00	0.13	3.0	1.1	38.03	54000
170713	NH4 acetate	D5 AC3-4	2.34E+01	7.97E-01	1.69E+02	2.27E+00	0.14	3.4	1.3	36.90	54000
170713	NH4 acetate	D5 AC4-5	1.37E+01	7.29E-01	1.04E+02	1.84E+00	0.13	5.3	1.8	35.18	54000
170713	NH4 acetate	D5 AC5-6	1.44E+01	6.68E-01	9.87E+01	1.67E+00	0.15	4.6	1.7	37.92	60000
170713	NH4 acetate	D5 AC6-7	1.35E+01	6.91E-01	9.39E+01	1.63E+00	0.14	5.1	1.7	37.64	60000
170713	NH4 acetate	D5 AC7-9	1.29E+01	7.30E-01	8.56E+01	1.58E+00	0.15	5.7	1.8	32.83	64000
170713	NH4 acetate	D5 AC9-11			-					36.23	
170713	NH4 acetate	D5 AC11-13								35.70	
170713	NH4 acetate	D5 AC13-15								36.47	

						Sample: I	D6						
		F	ern (<i>Polys</i>	tichum o	vatopalea	iceum var	. corai	ense). Lo	ocation:	Tomioka			
Sa	mple name		Cs-134	(Bq/kg)	Cs-137	(Bq/kg)	Ce134/	err/A	ctivity	Aliquot	Total Soil	l ivo timo	Root Surface Area
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	Cs137	Cs-134	Cs-137	Sample dry mass (g)	Sample Dry mass (g)	(s)	Pixel count (600 dpi image)
170713	plant	D6 plant	1604	18.2	12820	73.8	0.13	1.1	0.6	2.87		54000	
170713	soil	D6 S0-1	1.30E+04	5.26E+01	9.21E+04	1.95E+02	0.14	0.4	0.2	5.60	35.2	18000	3.66E+06
170713	soil	D6 S1-2	6.17E+03	3.72E+01	4.34E+04	1.36E+02	0.14	0.6	0.3	5.34	19.2	18000	9.57E+05
170713	soil	D6 S2-3	2.11E+03	1.91E+01	1.46E+04	6.86E+01	0.14	0.9	0.5	7.22	32.2	18000	4.06E+05
170713	soil	D6 S3-4	1.22E+03	1.42E+01	8.63E+03	5.15E+01	0.14	1.2	0.6	7.61	49.7	18000	2.43E+05
170713	soil	D6 S4-5	4.01E+02	8.41E+00	2.76E+03	2.87E+01	0.15	2.1	1.0	7.92	50.1	18000	1.46E+05
170713	soil	D6 S5-7	1.33E+02	4.03E+00	9.10E+02	1.23E+01	0.15	3.0	1.4	7.23	83.2	36000	2.92E+05
170713	soil	D6 S7-9	9.08E+01	3.26E+00	6.65E+02	1.02E+01	0.14	3.6	1.5	7.73	79.2	36000	1.68E+05
170713	soil	D6 S9-11	7.59E+02	3.26E+00	5.44E+02	9.79E+00	1.39	0.4	1.8	7.02	74.0	36000	
170713	soil	D6 S11-13	6.13E+01	3.04E+00	4.20E+02	8.75E+00	0.15	5.0	2.1	7.02	95.5	36000	
170713	soil	D6 S13-15	3.38E+01	3.27E+00	2.65E+02	9.37E+00	0.13	9.7	3.5	7.86	67.8	18000	

170713	soil	D6 S11-13	6.13E+01	3.04E+00	4.20E+02	8.75E+00	0.15	5.0	2.1	7.02	95.5	3600
170713	soil	D6 S13-15	3.38E+01	3.27E+00	2.65E+02	9.37E+00	0.13	9.7	3.5	7.86	67.8	1800
				Sample:	D6 NH4Ao	Extraction	on					
Sai	mple name	Comple ID	Cs-134	(Bq/kg)	Cs-137	' (Bq/kg)	Cs134/	err/A	ctivity	NH4Ac Mass	Live time (a)	
Date	Part		Activity	Act. err	Activity	Act. err	Cs137	Cs-134	Cs-137	(g)	Live time (s)	
170713	NH4 acetate	D6 AC0-1	7.94E+00	4.66E-01	6.61E+01	1.50E+00	0.12	5.9	2.3	36.2	54000	
170713	NH4 acetate	D6 AC1-2	8.44E+00	5.41E-01	6.53E+01	1.74E+00	0.13	6.4	2.7	37.0	40200	
170713	NH4 acetate	D6 AC2-3	8.64E+00	5.08E-01	6.97E+01	1.52E+00	0.12	5.9	2.2	37.2	54000	
170713	NH4 acetate	D6 AC3-4	6.67E+00	4.20E-01	5.51E+01	1.36E+00	0.12	6.3	2.5	37.7	54000	
170713	NH4 acetate	D6 AC4-5	3.68E+00	3.17E-01	3.21E+01	1.08E+00	0.11	8.6	3.4	37.2	54000	
170713	NH4 acetate	D6 AC5-7	2.11E+00	3.70E-01	1.27E+01	7.26E-01	0.17	17.5	5.7	38.0	54000	
170713	NH4 acetate	D6 AC7-9								34.0		
170713	NH4 acetate	D6 AC9-11								38.1		
170713	NH4 acetate	D6 AC11-13								37.9		
170713	NH4 acetate	D6 AC13-15								36.0		

					Sá	ample: D7							
		Fern	(Polystich	um ovato	paleaceu	m var. co	raiense	e). Loca	ation: J	luman-yan	na		
Sa	mple name	Sample ID	Cs-134	(Bq/kg)	Cs-137	′ (Bq/kg)	Cs134/	err/A	ctivity	Aliquot	Total Soil	Live time	Root Surface Area
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	Cs137	Cs-134	Cs-137	mass (g)	mass (g)	(s)	Pixel count (600 dpi image)
170726	plant	D7 plant	8.26E+02	5.90E+01	7.52E+03	2.38E+02	0.11	7.1	3.2	0.54		10900	
170726	soil	D7 S0-2	1.35E+03	2.14E+01	1.25E+04	9.03E+01	0.11	1.6	0.7	7.62	93.6	9000	1.32E+06
170726	soil	D7 S2-3	3.21E+02	1.03E+01	2.65E+03	4.07E+01	0.12	3.2	1.5	8.23	29.5	9000	4.07E+05
170726	soil	D7 S3-4	2.95E+02	8.20E+00	2.47E+03	3.29E+01	0.12	2.8	1.3	8.76	45.8	12000	5.09E+05
170726	soil	D7 S4-5	1.67E+02	6.50E+00	1.40E+03	2.54E+01	0.12	3.9	1.8	8.46	45.8	12000	4.17E+05
170726	soil	D7 S5-6	1.47E+02	6.19E+00	1.25E+03	2.40E+01	0.12	4.2	1.9	8.51	25.8	12000	1.53E+05
170726	soil	D7 S6-7	6.52E+01	3.50E+00	5.32E+02	1.27E+01	0.12	5.4	2.4	8.74	52.2	18000	2.18E+05
170726	soil	D7 S7-8	4.09E+01	3.05E+00	3.63E+02	1.09E+01	0.11	7.5	3.0	8.47	43.3	18000	4.06E+04
170726	soil	D7 S8-9	3.65E+02	2.89E+00	3.53E+02	1.08E+01	1.03	0.8	3.1	8.26	47.6	18000	4.58E+03
170726	soil	D7 S9-10	5.47E+01	3.40E+00	4.58E+02	1.25E+01	0.12	6.2	2.7	8.01	39.6	18000	1.95E+04
170726	soil	D7 S10-15	1.44E+02	3.63E+00	1.13E+03	1.39E+01	0.13	2.5	1.2	7.80	76.8	36000	3.92E+04

			Sa	mple: D7 l	NH4Ac Ex	traction					
Sa	mple name	Sample ID	Cs-134	(Bq/kg)	Cs-137	′ (Bq/kg)	Cs134/	err/A	ctivity	NH4Ac	Live time (a)
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	Cs137	Cs-134	Cs-137	Mass (g)	Live time (S)
170726	NH4 acetate	D7 AC0-2	4.38E+00	4.46E-01	3.32E+01	1.22E+00	0.13	10.2	3.7	37.5	40900
170726	NH4 acetate	D7 AC2-3	ND		7.70E+00	6.03E-01			7.8	37.1	54000
170726	NH4 acetate	D7 AC3-4	ND		8.52E+00	6.23E-01			7.3	37.5	54000
170726	NH4 acetate	D7 AC4-5	ND		4.60E+00	5.47E-01			11.9	37.0	54000
170726	NH4 acetate	D7 AC5-6	ND		4.32E+00	5.44E-01			12.6	35.6	54000
170726	NH4 acetate	D7 AC6-7	ND		2.37E+00	5.00E-01			21.1	37.3	54000
170726	NH4 acetate	D7 AC7-8								36.0	
170726	NH4 acetate	D7 AC8-9								36.7	
170726	NH4 acetate	D7 AC9-10								35.6	
170726	NH4 acetate	D7 AC10-15								37.5	

						Sample:	D8						
				Sa	sa Palma	te. Locati	on: Ju	man-yam	na				
Sa	mple name	Comala ID	Cs-134	(Bq/kg)	Cs-137	' (Bq/kg)	Cs134/	err/A	ctivity	Aliquot	Total Soil		Root Surface Area
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	Cs137	Cs-134	Cs-137	mass (g)	mass (g)	Live time (s)	Pixel count (600 dpi image)
170713	plant	D8 plant	3313	34.99	25730	143.1	0.13	1.1	0.6	4.80		18300	
170713	soil	D8 S0-3	1.30E+04	5.79E+01	1.07E+05	2.47E+02	0.12	0.4	0.2	4.17	19.1	5.06E+06	
170713	soil	D8 S3-4	6.18E+03	3.92E+01	5.21E+04	1.68E+02	0.12	0.6	0.3	4.36	21.2	18000	1.93E+06
170713	soil	D8 S4-5	6.39E+03	3.93E+01	5.30E+04	1.66E+02	0.12	0.6	0.3	4.56	17.2	18000	1.14E+06
170713	soil	D8 S5-6	4.30E+03	3.17E+01	3.52E+04	1.34E+02	0.12	0.7	0.4	4.65	23.6	18000	5.61E+05
170713	soil	D8 S6-7	1.81E+03	2.06E+01	1.51E+04	8.62E+01	0.12	1.1	0.6	4.90	27.3	18000	3.47E+05
170713	soil	D8 S7-8	3.98E+02	6.57E+00	3.31E+03	2.63E+01	0.12	1.7	0.8	6.03	22.0	36000	7.70E+04
170713	soil	D8 S8-9	2.08E+02	4.60E+00	1.67E+03	1.82E+01	0.12	2.2	1.1	6.40	35.6	36000	9.50E+04
170713	soil	D8 S9-11	1.64E+02	4.97E+00	1.32E+03	1.90E+01	0.12	3.0	1.4	6.76	61.7	24300	
170713	soil	D8 S11-13	5.02E+01	1.99E+00	3.74E+03	6.57E+00	0.01	4.0	0.2	7.54	119.6	54000	
170713	soil	D8 S13-15	2.87E+01	1.58E+00	2.31E+02	5.31E+00	0.12	5.5	2.3	7.42	85.1	54000	
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			;	Sample: D	8 NH4Ac	Extractio	n				
Sa	mple name	Sample ID	Cs-134	(Bq/kg)	Cs-137	(Bq/kg)	Cs134/	err/A	ctivity	NH4Ac Mass	Live time (c)
Date	Part	- Sample ID	Activity	Act. err	Activity	Act. err	Cs137	Cs-134	Cs-137	(g)	Live time (s)
170713	NH4 acetate	D8 AC0-3	5.86E+00	4.43E-01	4.20E+01	1.35E+00	0.14	7.6	3.2	35.0	46000
170713	NH4 acetate	D8 AC3-4	4.53E+00	4.22E-01	3.78E+01	1.16E+00	0.12	9.3	3.1	37.4	54000
170713	NH4 acetate	D8 AC4-5	7.84E+00	4.48E-01	6.24E+01	1.45E+00	0.13	5.7	2.3	36.6	54000
170713	NH4 acetate	D8 AC5-6	6.88E+00	4.59E-01	5.42E+01	1.37E+00	0.13	6.7	2.5	37.2	54000
170713	NH4 acetate	D8 AC6-7	5.07E+00	4.86E-01	3.94E+01	1.42E+00	0.13	9.6	3.6	36.0	38500
170713	NH4 acetate	D8 AC7-8	1.58E+00	3.28E-01	9.24E+00	6.41E-01	0.17	20.8	6.9	37.6	54000
170713	NH4 acetate	D8 AC8-9	ND		5.25E+00	6.02E-01			11.5	35.7	43000
170713	NH4 acetate	D8 AC9-11								36.6	
170713	NH4 acetate	D8 AC11-13								39.4	
170713	NH4 acetate	D8 AC13-15								37.0	

						Sample	e: D9						
		F	Fern <i>(Poly</i>	stichum c	ovatopale	aceum va	r. coraiense).	Locatio	n: Juma	n-yama			
Sa	mple name	Sample ID	Cs-134	(Bq/kg)	Cs-137	′ (Bq/kg)	Cs134/Cs137	err/A	ctivity	Aliquot Sample dry	Total Soil Sample Dry	Live time	Root Surface Area
Date	Part		Activity	Act. err	Activity	Act. err		Cs-134	Cs-137	mass (g)	mass (g)		Pixel count (600 dpi image)
170726	plant	D9 plant	3.45E+03	4.27E+01	2.43E+04	1.46E+02	0.14	1.2	0.6	0.82		64000	
170726	soil	D9 S0-7	3.55E+03	4.45E+01	2.87E+04	1.82E+02	0.12	1.3	0.6	4.09	19.0	9000	2.12E+05
170726	soil	D9 S7-8	2.16E+03	2.18E+01	1.79E+04	9.05E+01	0.12	1.0	0.5	5.40	30.6	18000	4.23E+05
170726	soil	D9 S8-9	2.66E+03	2.45E+01	2.18E+04	1.02E+02	0.12	0.9	0.5	5.07	19.9	18000	5.92E+05
170726	soil	D9 S9-10	1.58E+03	1.85E+01	1.28E+04	7.64E+01	0.12	1.2	0.6	5.40	30.3	18000	6.45E+05
170726	soil	D9 S10-11	8.94E+03	1.28E+01	7.31E+03	5.21E+01	1.22	0.1	0.7	5.80	39.8	20400	4.30E+05
170726	soil	D9 S11-12	3.06E+02	6.86E+00	2.41E+03	2.65E+01	0.13	2.2	1.1	6.35	44.6	24000	1.42E+05
170726	soil	D9 S12-13	2.05E+02	4.35E+00	1.71E+03	1.73E+01	0.12	2.1	1.0	7.24	46.9	36000	1.06E+05
170726	soil	D9 S13-14	1.96E+02	5.03E+00	1.58E+03	1.92E+01	0.12	2.6	1.2	6.63	48.9	30000	7.73E+04
170726	soil	D9 S14-15								6.66	88.8		7.39E+04

	Sample: D9 NH4Ac Extraction													
Sample name		Sample ID	Cs-134 (Bq/kg)		Cs-137 (Bq/kg)		Co424/Co427	err/Activity		NH4Ac	Live time (s)			
Date	Part	Jampie ID	Activity	Act. err	Activity	Act. err	05134/05137	Cs-134	Cs-137	Mass (g)	Live time (5)			
170726	NH4 acetate	D9 AC0-7	4.43E+00	3.49E-01	3.12E+01	1.04E+00	0.14	7.9	3.3	38.7	54000			
170726	NH4 acetate	D9 AC7-8	3.20E+00	2.77E-01	2.87E+01	1.01E+00	0.11	8.7	3.5	38.5	54000			
170726	NH4 acetate	D9 AC8-9	3.50E+00	3.62E-01	2.98E+01	1.04E+00	0.12	10.4	3.5	37.8	54000			
170726	NH4 acetate	D9 AC9-10								38.7				
170726	NH4 acetate	D9 AC10-11								38.5				
170726	NH4 acetate	D9 AC11-12							-	38.3				
170726	NH4 acetate	D9 AC12-13								35.7				
170726	NH4 acetate	D9 AC13-14								39.4				
170726	NH4 acetate	D9 AC14-15								35.6				

	Sample: D10												
Sasa Palmate. Location: Tsushima													
Sa	mple name		Cs-134 (Bq/kg)		Cs-137 (Bq/kg)			err/Activity					Root Surface Area
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	Cs134/ Cs137	Cs-134	Cs-137	Aliquot Sample dry mass (g)	Sample Dry mass (g)	Live time (s)	Pixel count (600 dpi image)
170807	plant	D10 plant	116.54	9.1	796.5	21.19	0.15	7.8	2.7	1.50		64000	
170807	soil	D10 S0-2	2.52E+04	1.33E+02	1.77E+05	4.84E+02	0.14	0.5	0.3	3.82	21.4	8000	1.32E+06
170807	soil	D10 S2-3	1.94E+04	1.08E+02	1.35E+05	3.91E+02	0.14	0.6	0.3	4.01	18.5	9000	6.09E+05
170807	soil	D10 S3-4	6.03E+03	5.44E+01	4.19E+04	1.95E+02	0.14	0.9	0.5	5.02	20.3	9000	6.96E+05
170807	soil	D10 S4-5	2.03E+03	3.34E+01	1.41E+04	1.18E+02	0.14	1.7	0.8	4.68	22.3	9000	4.85E+05
170807	soil	D10 S5-6	1.90E+03	2.16E+01	1.33E+04	7.69E+01	0.14	1.1	0.6	5.14	23.0	18000	3.52E+05
170807	soil	D10 S6-7	1.14E+03	1.77E+01	8.00E+03	6.09E+01	0.14	1.6	0.8	4.95	21.8	18000	2.96E+05
170807	soil	D10 S7-9	1.12E+03	1.68E+01	7.74E+03	5.78E+01	0.14	1.5	0.7	5.41	44.4	18000	8.03E+05
170807	soil	D10 S9-11	8.35E+02	1.27E+01	5.84E+03	4.34E+01	0.14	1.5	0.7	5.46	43.3	24000	1.83E+05
170807	soil	D10 S11-13	6.32E+02	9.08E+00	4.38E+03	3.12E+01	0.14	1.4	0.7	5.19	24.5	36000	1.90E+05
170807	soil	D10 S13-15	4.10E+02	9.30E+00	2.86E+03	3.06E+01	0.14	2.3	1.1	5.33	55.7	36000	2.15E+05
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Sample: D10 NH4Ac Extraction												
Sa	mple name	Sample ID	Cs-134 (Bq/kg)		Cs-137	(Bq/kg)	Cs134/	err/A	NH4Ac			
Date	Part	Gample ID	Activity Act. err		Activity	Act. err	Cs137	Cs-134	Cs-137	Mass (g)		
170807	NH4 acetate	D10 AC0-2	3.97E+01	9.78E-01	3.15E+02	3.73E+00	0.13	2.5	1.2	37.71		
170807	NH4 acetate	D10 AC2-3	2.88E+01	9.59E-01	2.40E+02	3.36E+00	0.12	3.3	1.4	37.51		
170807	NH4 acetate	D10 AC3-4	1.60E+01	5.80E-01	1.31E+02	2.02E+00	0.12	3.6	1.5	38.86		
170807	NH4 acetate	D10 AC4-5	6.53E+00	4.28E-01	5.08E+01	1.31E+00	0.13	6.6	2.6	38.45		
170807	NH4 acetate	D10 AC5-6	5.19E+01	3.75E+00	4.45E+02	1.27E+01	0.12	7.2	2.8	37.31		
170807	NH4 acetate	D10 AC6-7	4.54E+00	4.45E-01	3.35E+01	1.09E+00	0.14	9.8	3.3	38.39		
170807	NH4 acetate	D10 AC7-9	5.54E+00	2.41E-01	3.76E+01	6.16E-01	0.15	4.3	1.6	38.73		
170807	NH4 acetate	D10 AC9-11					-		-	37.27		
170807	NH4 acetate	D10 AC11-13								35.81		
170807	NH4 acetate	D10 AC13-15							-	36.42		

	Sample: D11													
Sasa Palmate. Location: Tsushima														
Sample name		Sample ID	Cs-134 (Bq/kg)		Cs-137 (Bq/kg)		Cs134/	err/Activity		Aliquot Sample dry	Total Soil Sample Dry	Live time	Root Surface Area	
Date	Part		Activity	Act. err	Activity	Act. err	05157	Cs-134	Cs-137	mass (g)	mass (g)	(3)	Pixel count (600 dpi image)	
170807	plant	D11 plant	8.58E+02	2.81E+01	5.31E+03	7.24E+01	0.16	3.3	1.4	1.66				
170807	soil	D11 S0-2	1.42E+04	5.29E+01	1.18E+05	2.26E+02	0.12	0.4	0.2	5.86	56.3	18000	1.98E+06	
170807	soil	D11 S2-3	3.00E+03	2.68E+01	2.48E+04	1.12E+02	0.12	0.9	0.5	4.89	26.8	18000	7.18E+05	
170807	soil	D11 S3-4	9.58E+02	1.53E+01	7.81E+03	6.23E+01	0.12	1.6	0.8	4.99	23.6	18000	2.27E+05	
170807	soil	D11 S4-5	5.22E+02	1.14E+01	4.37E+03	4.60E+01	0.12	2.2	1.1	5.17	24.4	18000	1.20E+05	
170807	soil	D11 S5-6	2.06E+02	7.70E+00	1.74E+03	2.86E+01	0.12	3.7	1.6	5.36	26.6	18000	8.48E+04	
170807	soil	D11 S6-7	1.36E+02	5.61E+00	1.08E+03	2.03E+01	0.13	4.1	1.9	5.00	28.4	24000	9.54E+04	
170807	soil	D11 S7-9	1.36E+02	5.82E+00	1.09E+03	2.14E+01	0.12	4.3	2.0	5.13	54.3	22000	2.06E+05	
170807	soil	D11 S9-11	1.13E+02	4.12E+00	8.87E+02	1.45E+01	0.13	3.7	1.6	5.38	38.0	36000	6.77E+04	
170807	soil	D11 S11-13	5.66E+01	3.15E+00	4.56E+02	1.06E+01	0.12	5.6	2.3	5.43	32.2	36000	1.16E+05	
170807	soil	D11 S13-15	7.95E+01	4.12E+00	6.37E+02	1.44E+01	0.12	5.2	2.3	5.40	44.6	27000	4.66E+04	

Sample: D11 NH4Ac Extraction													
Sample name		Sample ID	Cs-134	(Bq/kg)	Cs-137	′ (Bq/kg)	Cs134/	err/Activity		NH4Ac Mass			
Date	Part	Sample ID	Activity	Act. err	Activity	Act. err	Cs137	Cs-134	Cs-137	(g)			
170807	NH4 acetate	D11 AC0-2	4.51E+01	1.06E+00	3.81E+02	4.05E+00	0.12	2.4	1.1	42.15			
170807	NH4 acetate	D11 AC2-3	8.96E+00	6.25E-01	8.02E+01	2.00E+00	0.11	7.0	2.5	37.4			
170807	NH4 acetate	D11 AC3-4	4.92E+00	4.93E-01	3.61E+01	1.39E+00	0.14	10.0	3.8	38.03			
170807	NH4 acetate	D11 AC4-5	2.73E+00	3.83E-01	2.02E+01	1.09E+00	0.13	14.0	5.4	38.65			
170807	NH4 acetate	D11 AC5-6	ND		7.45E+00	5.94E-01			8.0	39.06			
170807	NH4 acetate	D11 AC6-7	ND		5.54E+00	5.37E-01			9.7	38.12			
170807	NH4 acetate	D11 AC7-9	ND		5.91E+00	5.51E-01			9.3	37.13			
170807	NH4 acetate	D11 AC9-11								36.73			
170807	NH4 acetate	D11 AC11-13								37.72			
170807	NH4 acetate	D11 AC13-15								42.15			

	Sample: D12														
	Suzuki grass (Miscanthus sinensis). Location: Tsushima														
Sample name		Samula ID	Cs-134 (Bq/kg)		Cs-137	7 (Bq/kg)	Ce134/Ce137	err/A	ctivity	Aliquot	Total Soil	Live time	Root Surface Area		
Date	Part		Activity	Act. err	Activity	Act. err		Cs-134	Cs-137	mass (g)	mass (g)	(s)	Pixel count (600 dpi image)		
170807	plant	D12 plant	97.1	3.338	637.7	9.63	0.15	3.4	1.5	8.61		64000			
170807	soil	D12 S0-2	5.00E+03	3.93E+01	3.56E+04	1.44E+02	0.14	0.8	0.4	8.36	110.3597051	9000	2.25E+06		
170807	soil	D12 S2-3	3.66E+03	2.52E+01	2.58E+04	9.07E+01	0.14	0.7	0.4	7.58	54.4	18000	8.02E+05		
170807	soil	D12 S3-4	3.56E+03	2.42E+01	2.57E+04	8.85E+01	0.14	0.7	0.3	8.09	31.1	18000	5.29E+05		
170807	soil	D12 S4-5	2.85E+03	2.18E+01	2.03E+04	7.98E+01	0.14	0.8	0.4	7.73	61.1	18000	7.66E+05		
170807	soil	D12 S5-6	1.85E+03	1.69E+01	1.31E+04	6.14E+01	0.14	0.9	0.5	8.77	55.4	18000	5.28E+05		
170807	soil	D12 S6-7	9.06E+02	1.07E+01	6.29E+03	3.79E+01	0.14	1.2	0.6	8.00	53.1	24000	5.98E+05		
170807	soil	D12 S7-9	9.48E+01	3.99E+00	7.14E+02	1.25E+01	0.13	4.2	1.7	8.70	117.9	24000	4.39E+05		
170807	soil	D12 S9-11	3.99E+01	3.51E+00	3.03E+02	9.24E+00	0.13	8.8	3.0	7.47	81.1	24000	2.90E+05		
170807	soil	D12 S11-13	3.99E+01	2.61E+00	2.71E+02	6.73E+00	0.15	6.5	2.5	8.09	77.3	36000	7.19E+04		
170807	soil	D12 S13-15	5.75E+01	3.45E+00	4.19E+02	9.51E+00	0.14	6.0	2.3	6.24	48.3	36000	6.97E+04		
					-										
				Samp	ole: D12 N	H4Ac Ext	raction					1			

	Sample: D12 NH4Ac Extraction													
Sa	mple name	Sample ID	Cs-134	(Bq/kg)	Cs-137	′ (Bq/kg)	Co124/Co127	err/Activity		NH4Ac Mass	Live time (s)			
Date	Part	oumpie ib	Activity	Act. err	Activity	Act. err	05134/05137	Cs-134	Cs-137	(g)	Live time (3)			
170807	NH4 acetate	D12 AC0-2	6.90E+01	1.23E+00	3.86E+02	4.69E+00	0.18	1.8	1.2	37.8	30000			
170807	NH4 acetate	D12 AC2-3	3.23E+01	9.66E-01	2.57E+02	3.49E+00	0.13	3.0	1.4	37.2	36000			
170807	NH4 acetate	D12 AC3-4	3.08E+01	8.32E-01	2.51E+02	3.13E+00	0.12	2.7	1.2	38.8	44000			
170807	NH4 acetate	D12 AC4-5	2.46E+01	6.70E-01	1.98E+02	2.52E+00	0.12	2.7	1.3	39.2	54000			
170807	NH4 acetate	D12 AC5-6	1.87E+01	6.47E-01	1.46E+02	2.20E+00	0.13	3.5	1.5	34.8	54000			
170807	NH4 acetate	D12 AC6-7	8.44E+00	5.13E-01	6.70E+01	1.48E+00	0.13	6.1	2.2	37.1	54000			
170807	NH4 acetate	D12 AC7-9	ND		9.70E+00	8.08E-01			8.3	37.5	36000			
170807	NH4 acetate	D12 AC9-11	ND		2.22E+00	2.28E-01			10.3	39.6	120000			
170807	NH4 acetate	D12 AC11-13								38.1				
170807	NH4 acetate	D12 AC13-15								38.4				