I am currently a graduate student at UHM's School of Ocean and Earth Science and Technology where I am continuing my research in environmental geochemistry with plans to earn a PhD. I had the opportunity last year to present my research at the European Geosciences Union conference in Vienna, Austria. This work constituted part of a larger research project contained in my senior thesis, which quantifies Fukushima-derived fallout in Hawaiian soils.

Quantifying Atmospheric Fallout of Fukushima-Derived Radioactive Isotopes in Mushrooms in the Hawaiian Islands

Trista McKenzie

Independent research through Undergraduate Research Opportunities Program
Mentor: Dr. Henrietta Dulai

In March of 2011, the radioisotopes cesium-134 (^{134}Cs) and cesium-137 (^{137}Cs) were released into the atmosphere from the damaged Fukushima Dai-ichi Nuclear Power Plant. This study estimated the magnitude of atmospheric fallout of these isotopes on O'ahu and Hawai'i, and examined the patterns of cesium wet deposition with precipitation in mushroom samples. This study found that Fukushima-derived cesium was present in mushrooms collected in the Hawaiian islands and that Fukushima-derived cesium inventories in mushrooms were somewhat correlated with precipitation gradients. The activities detected were several orders of magnitude lower than fallout associated with the nuclear weapons testing in the Pacific.

Introduction

On March 11, 2011, the Fukushima Dai-ichi Power Plant experienced significant damage from a tsunami that followed the 9.0 magnitude Tohoku earthquake. The three reactors that were running during the earthquake were immediately put on an emergency shutdown (Thakur et al., 2013) and emergency generators managed the cooling systems. Approximately fifty minutes later, a subsequent tsunami hit the power plant. While the Fukushima Dai-ichi Power Plant had a ten-meter high sea wall, it was not enough to prevent the incoming tsunami of estimated height of thirteen to fifteen meters (Pararas-Carayannis, 2014). The seawater proceeded to flood the area that housed the emergency generators, disabling them. This resulted in the loss of cooling capability, overheating of the reactors, and the partial meltdown and damage of three of the six nuclear reactors.

Starting on March 12, 2011, significant amounts of radioactive isotopes were released into the atmosphere, including ^{134}Cs and ^{137}Cs. ^{134}Cs and ^{137}Cs were released at a near 1:1 ratio (Buesseler, et al., 2011). The majority of the Fukushima-derived atmospheric radioactivity then proceeded west toward the Pacific Ocean. The Fukushima Power Plant accident was classified as a Level 7 on the International Nuclear Event Scale (INES), indicating a major nuclear

Mānoa Horizons, Vol. 1, 2016, pp. 28–33

accident because of the high initial amount of radionuclides released into the environment (Thakur et al., 2013).

Between March 19 and March 24, 2011, the Environmental Protection Agency's (EPA) RadNet air filters detected these isotopes on the islands of O'ahu, Kaua'i, and Hawai'i along with other locations in the Pacific (Environmental Protection Agency, 2011; Figure 1). Gross beta radiation measured by the EPA includes all beta emitting radionuclides. The peak on Figure 1 shows that the majority of the plume passed above Hawai'i between March 19 and March 26, which is when most of the deposition was expected to happen. It should be noted that the activity levels of the radionuclides detected were very low and not remotely near action levels determined by the EPA.

On March 22, 2011, the EPA published a press release informing that Fukushima-derived radioisotopes had been detected in air masses over the western United States. EPA's RadNet air samplers had detected ^{137}Cs, ^{134}Cs, ^{132}Te, ^{132}I, and ^{131}I. All activity concentrations were hundreds of thousands to millions of times below the level of concern (Environmental Protection Agency, 2011).

The EPA's RadNet air filters and cartridges, which were analyzed for individual isotopes, detected the maximum activity for the radionuclides ^{137}Cs and ^{134}Cs for air around the islands of O'ahu and Kaua'i. For Kaua'i, the maximum values were 2.78 ± 0.17 x 10^{-3} Bq m^{-3}, 2.92 ± 0.23 x 10^{-3} Bq m^{-3}, for ^{137}Cs and ^{134}Cs, respectively. For O'ahu, the maximum values detected were 4.44 ± 0.56 x 10^{-3} Bq m^{-3} (^{137}Cs) and 3.44 ± 0.34 x 10^{-3} Bq m^{-3} (^{134}Cs).

^{134}Cs and ^{137}Cs readily attach to particles in the atmosphere (Thakur et al., 2013) and are therefore relatively easy to quantify on air filters that capture suspended particles. The Nuclear and Industrial Safety Agency (NISA) of Japan initially estimated that 15 PBq (Petabecquerel: = 10^{15} Becquerel) of ^{137}Cs was released into the atmosphere. Several months after the nuclear disaster, this result was lowered by NISA to 11 PBq of ^{137}Cs (Thakur et al., 2013). Additionally, ^{134}Cs and ^{137}Cs derived from Fukushima were released at a near one to one uniform ratio (Buesseler et al., 2011).

Atmospheric fallout is primarily deposited by means of precipitation (Wetherbee et al., 2012). Since cesium isotopes were detected in air and precipitation it is fair to assume that the Hawaiian Islands experienced wet deposition of these nuclides. The Hawaiian Islands experience unique rain patterns due to the prevailing trade wind patterns and thus one hypothesis is that wet deposition was driven by rain distribution in March 2011. Indeed, it has been previously determined by measuring soil samples in Hawai'i that there is a positive linear relationship between precipitation and cesium fallout from nuclear weapons tests (Cox & Fankhauser, 1984). Atmospheric nuclear weapons' testing was most prevalent from 1945–1963. During this time an estimated 950 PBq of cesium was released (UNESCO, 2000) that left a positive fingerprint on the islands. This study investigates how Fukushima releases compare to those inputs. During atmospheric releases the radionuclides are not homogeneously distributed in the atmosphere, in addition their

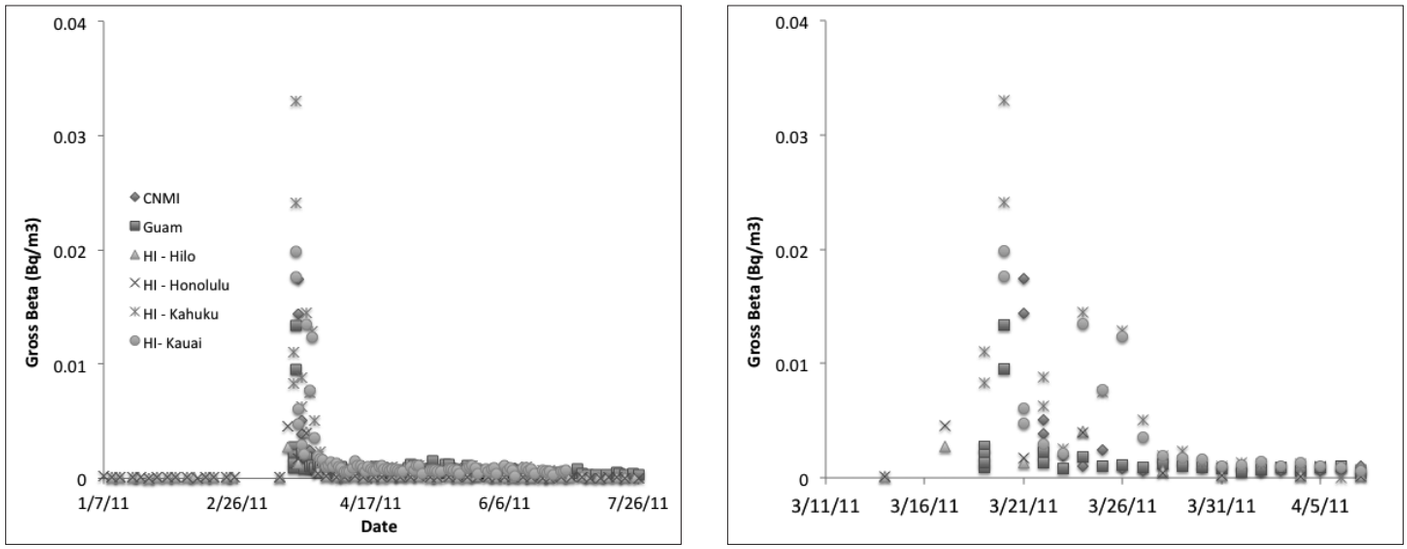

Figure 1 Gross beta radiation detected in the air in 2011 before, during, and after the FPP accident by EPA's RadNet system (Environmental Protection Agency, 2011).

fallout is driven by meteorological conditions, especially precipitation. The concentrations are generally higher in mountainous areas or oceanic environments where precipitation is largest (Cox & Fankhauser, 1984). Because precipitation rates on the Hawaiian Islands are higher on the windward side of the islands this is where higher radionuclide concentrations are anticipated as well. Cox & Fankhauser demonstrated that after the nuclear weapons tests fallout there was a predictable pattern in ^{137}Cs activities in soil and rock samples that suggested that the higher rainfall deposited higher activities of cesium.

Certain species of mushroom have previously been determined to be good bioindicators of radionuclide deposition from studies following the Chernobyl nuclear disaster (Duff et al., 2008). Mushroom species that have gills, pores, or spines and have stalks have been shown to have a stronger affinity for radiocesium and are less dependent on growing in soil that encourages radiocesium uptake (Duff et al., 2008). Mushrooms with these characteristics typically have a higher mycelium concentration and have a longer average lifespan (Castro et al., 2012). Additionally, it has been shown that the greatest amount of cesium accumulates in the cap of the mushroom (Oolbekkink, 1989).

In addition to differences between mushroom species, certain soil characteristics have been shown to positively influence radiocesium uptake by mushrooms. The bioavailability of radiocesium in soil is strongly correlated to the concentration of clays, specifically exchangeable potassium (Seeger, 1978). Stable cesium concentration in the soil has shown to be inversely related to radiocesium uptake (Oolbekkink, 1989). Additionally, it has been shown that soils with a pH of 6 or greater will allow for maximum radiocesium uptake. Using this information, one can assume that clays or alkaline soils should have higher bioavailable radiocesium concentrations than an acidic or organic soil. It has also been shown that mushrooms growing in decaying wood uptake significantly less radiocesium than all other substrates (Oolbekkink, 1989). Moreover, given the large number of factors that influence cesium uptake by mushrooms, the transfer factor (TF), which is the ratio of radiocesium activity concentrations (Bq/kg) in mushroom to the radiocesium concentration per soil area (Bq/m^2), may differ by up to 200 times (Kaduka et al., 2006).

Following the Fukushima nuclear disaster, studies have been conducted on mushrooms within close proximity to the FDNPP. One study found that 81.2% of wild mushrooms collected within 30 km of the FDNPP contained over 100 Bq/kg of radiocesium and 36.4% of those contained over 1000 Bq/kg (Nakashima, et al., 2015).

Experiments following the Fukushima Dai-ichi nuclear disaster have shown that a majority of the ^{137}Cs deposited in soil tends to remain in the top five centimeters of the vertical soil profile (Tanaka et al., 2012). The authors suggest this is because ^{137}Cs has a strong affinity for soil compounds. In the case of ^{137}Cs, the authors demonstrate that ^{137}Cs tends to bind with clay minerals in the soil and greater than 87% of Fukushima derived ^{137}Cs was found in the top five centimeters of soil. Further studies on the migration of pre-Fukushima ^{137}Cs in soils in forests near the FDNNP have further shown a very slow downward migration averaging at 0.46 mm/year with Fukushima-derived ^{137}Cs having negligible downward displacement (Teramage, et al., 2016). This is consistent with experiments conducted after Chernobyl, which demonstrated that ^{137}Cs deposited in the topsoil layer is adsorbed into that layer and can be bioavailable for long periods of time (Filipovic-Vincekovic et al., 1991).

Other evidence speaks for low cesium migration in soils and sediments. The presence of ^{137}Cs from nuclear fallout from the 1960's is used in sedimentation rate studies (Ritchie et al., 1990). In these studies sediment cores are collected and analyzed for excess ^{210}Pb and the presence of ^{137}Cs. The layer containing ^{137}Cs is assumed to have formed in the 1960's. This application further supports the general assumption that cesium is stabilized in soils and sediments for many decades.

Building on Cox & Fankhauser's conclusions and the RADNET data that showed the presence of Fukushima-derived radionuclides in the atmosphere above the islands in March 2011, the purpose of this study was to confirm the presence of Fukushima-derived fallout by analyzing mushrooms in order to test the hypothesis that 1) the atmospheric fallout in the Hawaiian Islands was present and 2) wet deposition was the major driver of radionuclide deposition and that there is a positive correlation between rain and deposited amounts of cesium. Because the newly deposited cesium levels were expected to be low and suspected to be near detection limits, we took advantage of bioaccumulation of cesium in organisms and analyzed mushroom samples to confirm the presence of fallout. Mushrooms have been shown to be good indicators of the presence or absence of cesium fallout in Europe (Duff et al., 2008) and Asia (Nakashima, et al., 2015). For our study mushrooms were collected along precipitation gradients as locational indicators for wet cesium deposition.

Methods

Mushroom samples on O'ahu and Hawai'i were collected in areas of differing rainfall amounts. Each sample was air dried, ashed, and homogenized prior to being placed in the crucible. The radioactivity in each sample was measured by gamma spectroscopy with an Ortec GEM-40 coaxial HPGe detector. Samples on average were counted for four to seven days. Rainfall amounts in March 2011 were determined using the Hawai'i Rainfall Atlas (Giambelluca, et al., 2013) and then all results were decay corrected to March 20, 2011.

This study could not be restricted to any one specific species of mushrooms so any species available at the study sites was collected and identified if possible. The presented results therefore are from different species with different bioaccumulation rates.

Results

Mushroom samples were collected between 2012 and 2013. ^{134}Cs and ^{137}Cs activities were decay corrected to March 2011 and compared to rainfall in the same month. Detectable ^{134}Cs activities ranged from 0.2–7.5 Bq/kg and detectable ^{137}Cs activities ranged from 0.4–89.0 Bq/kg. Our results are given in Table 1.

Table 1 Locations of sampling, species, ^{134}Cs and ^{137}Cs activities (Bq/kg), and March 2011 rainfall (mm). <CL means activities were below critical level of detection.

ISLAND	SAMPLE LOCATION	SPECIES	LAT LON	^{134}CS (BQ/KG)	^{137}CS (BQ/KG)	MAR 2011 RAIN(MM)
O'ahu	Mānoa Falls	Polyporales sp.	21.34, -157.80	1.56 ± 0.15	2.80 ± 0.23	304 ± 42.0
O'ahu	Kahana Dam	Microporus affinis	21.54, -157.88	0.72 ± 0.10	1.25 ± 0.12	290 ± 56.6
O'ahu	UHM	Unidentified	21.30, -157.82	<CL	<CL	118.9 ± 25.2
O'ahu	UHM	Unidentified	21.98, -157.82	<CL	<CL	118.9 ± 25.2
O'ahu	Dole St	Chlorophyllum molybdites	21.30, -157.82	<CL	0.39 ± 0.08	107.5 ± 19.7
O'ahu	Makiki Valley	Unidentified	21.32, -157.83	1.33 ± 0.12	1.93 ± 0.16	178.2 ± 31.9
O'ahu	Makiki Valley	Unidentified	21.320, -157.83	0.83 ± 0.08	1.67 ± 0.14	178.2 ± 31.9
O'ahu	Kahana Valley	Earliella scabrosa	21.54, -157.89	<CL	4.79 ± 0.39	335.0 ± 57.7
O'ahu	Kahana Valley	Microporus affinis	21.54, -157.89	0.60 ± 0.07	3.34 ± 0.26	335.0 ± 57.7
O'ahu	Kahana Dam	Microporus affinis	21.54, -157.88	0.70 ± 0.09	1.43 ± 0.12	335.0 ± 57.7
Hawai'i	MK Acc. Rd	Sullius brevipes	19.72, -155.45	1.12 ± 0.19	88.99 ± 6.87	161.8 ± 29.6
Hawai'i	Saddle Rd	Gomphidium oregonensis	19.69, -155.45	1.12 ± 0.19	30.05 ± 2.34	189.5 ± 32.3
Hawai'i	Waiakea Exp St	Unidentified	19.65, -155.08	0.96 ± 0.11	19.52 ± 1.51	418.0 ± 35.3
Hawai'i	Waiakea Exp St	Earliella scabrosa	19.65 -155.08	<CL	23.87 ± 1.84	418.4 ± 35.7
Hawai'i	Waiakea Exp St	Marasmiellus inoderma	19.65, -155.08	7.51 ± 0.69	18.78 ± 1.49	419.1 ± 36.0
Hawai'i	Stainbeck Hwy	Unidentified	19.59, -155.15	1.17 ± 0.14	4.20 ± 0.34	571.1 ± 55.7
Hawai'i	Stainbeck Hwy	Unidentified	19.59, -155.15	0.48 ± 0.06	5.96 ± 0.47	571.1 ± 55.7
Hawai'i	Kaiwiki Rd	Microporus flabelliformis	19.76, -155.16	6.80 ± 0.67	20.27 ± 1.61	655.1 ± 42.1
Hawai'i	ChinChuck Rd	Microporus flabelliformis	19.87, -155.16	5.96 ± 0.54	9.43 ± 0.78	673.2 ± 38.2
Hawai'i	MacKenzie Park	Auricularia sp.	19.44, -154.86	0.24 ± 0.04	4.83 ± 0.38	259.7 ± 12.9
Kaua'i	Princeville	Lepiota besseyi	22.22, -159.49	1.48 ± 0.21	2.87 ± 0.26	195.2 ± 25.9
Kaua'i	Wainiha	Merulius tremollosus	22.20, -159.56	1.13 ± 0.11	1.64 ± 0.15	319.6 ± 52.2
Kaua'i	Kōke'e St Park	Suillus brevipes	22.12, -159.71	1.97 ± 0.20	17.04 ± 1.33	125.4 ± 31.4
Kaua'i	Waipa	Macrocybe spectabilis	22.20, -159.51	<CL	8.14 ± 0.65	236.1 ± 30.9

Discussion

A majority of our samples had detectable Fukushima-derived cesium because mushrooms bioaccumulate and thus concentrate cesium. Measured cesium activities in mushrooms from the islands of O'ahu, Hawai'i, and Kaua'i are shown in Figure 2.

On O'ahu, we found that cesium activities generally were correlated with higher precipitation rates. We found detectable ^{134}Cs inventories ranging from 0.3 Bq/kg to 1.7 Bq/kg and ^{137}Cs inventories ranged from 0.4 Bq/kg to 4.8 Bq/kg.

On the island of Hawai'i, samples were only collected on the windward side of the island and primarily in the Hilo area. With some exceptions, mushrooms collected on the island of Hawai'i generally followed the expected trend of increased cesium deposition with increased rainfall. Detectable ^{134}Cs and ^{137}Cs inventories ranged from 0.2 Bq/kg to 7.5 Bq/kg and 4.8 Bq/kg to 89.0 Bq/kg, respectively.

^{134}Cs represents exclusively Fukushima-derived fallout whereas ^{137}Cs represents both Fukushima-derived and historical fallout. Given that ^{134}Cs and ^{137}Cs were released from the Fukushima Nuclear Power Plant at a near one to one ratio (Buesseler, et al., 2011), we can assume the total cesium inventory in mushrooms from Fukushima ranged from 0.6 to 3.4 Bq/kg on O'ahu and 0.4 Bq/kg to 15 Bq/kg on Hawai'i. These levels are well under the Derived Intervention Limit (DIL) of 1200 Bq/kg total cesium and thus are far below a level which concerns human health (FDA, 2005).

Additionally, we evaluated the fraction of "old" pre-Fukushima cesium and "new" Fukushima-derived cesium content in mushrooms (Figure 3). It is apparent that all mushrooms that had detectable cesium had some old cesium in them. In fact, most cesium in mushrooms was from pre-Fukushima fallout as shown by the large number of mushrooms with 60–100% old cesium in them.

While bioaccumulation happens at different rates within species, mushrooms still show a generally positive trend of precipitation driven cesium deposition that is also heavily influenced by mushroom species, canopy cover, and substrate characteristics.

Conclusion

The presence of Fukushima-derived fallout on the islands of O'ahu and Hawai'i has been confirmed. The level of radioactivity detected, however, is many magnitudes lower than a level of health concern using the FDA's Derived Intervention Limit of 1200 Bq/kg radiocesium. We found a monotonically increasing trend between Fukushima-derived cesium fallout and rainfall in March 2011. Factors such as mushroom species and canopy cover were determined to be important factors in Fukushima-derived cesium deposition and mushroom uptake.

References

Buesseler, K., Aoyama, M., & Fukasawa, M. (2011). Impacts of the Fukushima Nuclear Power Plants on Marine Radioactivity. Environmental Science & Technology Environ. Sci. Technol., 45(23), 9931–9935.

Castro, L. D., Maihara, V., Silva, P., & Figueira, R. (2012).

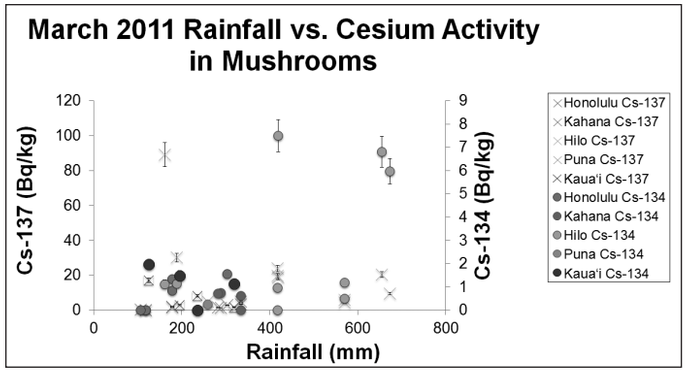

Figure 2 March 2011 rainfall in mm vs. radiocesium activity in Bq/kg for samples collected on the islands of O'ahu and Hawai'i (both ^{137}Cs and ^{134}Cs). Activities have been decay corrected to March 2011, the month of the Fukushima disaster.

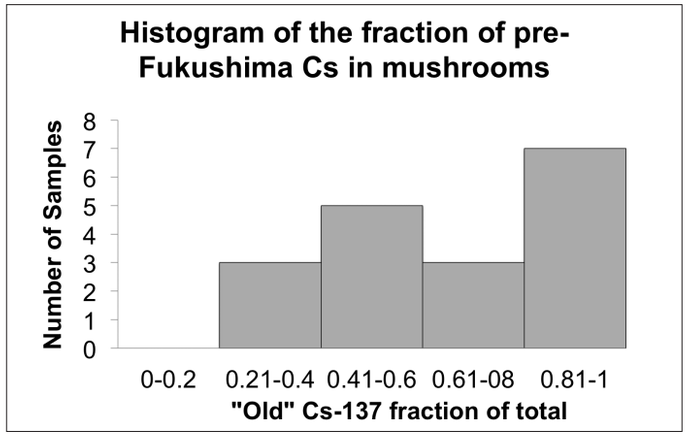

Figure 3 Old" pre-Fukushima cesium as a fraction of total cesium measured in mushrooms.

Artificial and natural radioactivity in edible mushrooms from Sao Paulo, Brazil. Journal of Environmental Radioactivity, 113, 150–154.

Cox, M. E., & Fankhauser, B. L. (1984). Distribution of Fallout Cesium-137 in Hawaii. Health Physics, 46(1), 65–71.

Department of Health (DOH) (2011). Milk Sample Shows Trace Amounts of Radiation in Milk from Big Island Dairy.

Duff, M. C., & Ramsey M. L. (2008). Accumulation of radiocesium by mushrooms in the environment: a literature review. Journal of Environmental Radioactivity, 99(6): 912–23, doi:10.1016/j.jenvrad.2007.11.017

Environmental Protection Agency (EPA) (2011). RADNET Database.

Food and Drug Administration (FDA) (2005). "Compliance Policy Guide: Radionuclides in Imported Foods—Levels of Concern." Silver Spring, MD. Web.

Filipović-Vinceković, N., Barišić, D., Mašić, N., & Lulić, S. (1991). Distribution of fallout radionuclides through soil surface layer. Journal of Radioanalytical and Nuclear Chemistry, Articles Journal of Radioanalytical and Nuclear Chemistry Articles, 148(1), 53–62.

Giambelluca, T. W., Q. Chen, A.G. Frazier, J. P. Price, Y.-L. Chen, P.-S. Chu, J. K. Eischeid, and D. M. Delparte, 2013: Online Rainfall Atlas of Hawai'i. *Bull. Amer. Meteor. Soc.* 94, 313–316, doi: 10.1175/BAMS-D-11-00228.1.

Hawaii Soil Atlas. (2014). http://gis.ctahr.hawaii.edu/SoilAtlas

Kaduka, M., Shutov, V., Bruk, G., Balonov, M., Brown, J., & Strand, P. (2006). Soil-dependent uptake of 137Cs by mushrooms: Experimental study in the Chernobyl accident areas. Journal of Environmental Radioactivity, 89(3), 199–211.

Kato, H., Onda, Y., and Teramage M. (2012). Depth distribution of 137Cs, 134Cs, and 131I in soil profile after Fukushima Dai-ichi Nuclear Power Plant Accident. Journal of Environmental Radioactivity, 111, 59–64.

Mangano, J. J., & Sherman, J. D. (2013). Elevated airborne beta levels in Pacific/West Coast US States and trends in hypothyroidism among newborns after the Fukushima nuclear meltdown. OJPed Open Journal of Pediatrics, 03(01), 1–9.

Nakashima, K., Orita, M., Fukuda, N., Taira, Y., Hayashida, N., Matsuda, N., & Takamura, N. (2015). Radiocesium concentrations in wild mushrooms collected in Kawauchi Village after the accident at the Fukushima Daiichi Nuclear Power Plant. PeerJ, 3.

Oolbekkink, G. T., & Kuyper, T. W. (1989). Radioactive caesium from Chernobyl in fungi. *Mycologist, 3*(1), 3–6.

Pararas-Carayannis, G. (2013). The Great Tohoku-Oki Earthquake and Tsunami of March 11, 2011 in Japan: A Critical Review and Evaluation of the Tsunami Source Mechanism. Pure and Applied Geophysics Pure Appl. Geophys., 171(12), 3257–3278.

Révay, Zs., Belgya, T., Ember, P. P., Molnár, G. L..(2001). Recent Developments in HYPERMET PC, Journal of Radioanalytical Nuclear Chemistry, 248,401–405.

Ritchie, J. C., & Mchenry, J. R. (1990). Application of Radioactive Fallout Cesium-137 for Measuring Soil Erosion and Sediment Accumulation Rates and Patterns: A Review. Journal of Environment Quality, 19(2), 215.

Sanders, C. J., Smoak, J. M., Cable, P. H., Patchineelam, S. R., & Sanders, L. M. (2011). Lead-210 and Beryllium-7 fallout rates on the southeastern coast of Brazil. Journal of Environmental Radioactivity, 102(12), 1122–1125.

Seeger, R. (1978). Potassium Content Of Wild Mushrooms. Abstracts, 583.

Siegel, D. I. (1989). Geochemistry of the Cambrian-Ordovician aquifer system in the northern Midwest, United States, 40–42.

Tanaka, S., & Kado, S. (2014). Analysis of Radioactive Release from the Fukushima Daiichi Nuclear Power Station. Reflections on the Fukushima Daiichi Nuclear Accident, 51–83.

Teramage, M. T., Onda, Y., & Kato, H. (2016). Small scale temporal distribution of radiocesium in undisturbed coniferous forest soil: Radiocesium depth distribution profiles. Journal of Environmental Management, 170, 97–104.

Thakur, P., Ballard, S., & Nelson, R. (2013). An overview of Fukushima radionuclides measured in the northern hemisphere. Science of The Total Environment, 458–460, 577–613.

United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 2000 report to the general assembly with scientific annexes. New York: United Nations; Publication E 00:IX.3; 2000.

Wetherbee, G. A., Gay, D. A., Debey, T. M., Lehmann, C. M., & Nilles, M. A. (2012). Wet Deposition of Fission-Product Isotopes to North America from the Fukushima Dai-ichi Incident, March 2011. Environmental Science & Technology Environ. Sci. Technol., 46(5), 2574–2582.