Radio-isotopic Analysis of Post-Fukushima Accident Japanese Soil Samples

By

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Abstract

Radioactive fallout from the triple reactor disaster at the Fukushima Daiichi Nuclear Power Station, following a magnitude 9.0 earthquake and subsequent tsunami, is readily observed in soil samples collected from Japan, well beyond the exclusion zone. Samples from three regions, approximately 210 km, 550 km, and 1060 km from the Fukushima Daiichi Nuclear Power Station, were tested for gross gamma activity and radio-isotopic composition. The primary isotopes of focus were the most commonly detected radio-isotopes of Cesium, $^{134}$Cs and $^{137}$Cs.

Note: Another individual should be listed as a co-contributor, given their important role in providing material from Japan for testing, but their name has been left out for purposes of anonymity. To that person, go my deepest thanks.
Introduction

Following the March 11 Tohoku earthquake of 2011 and the resulting Fukushima Daiichi Nuclear Power Station disaster, radioactive material was released into the air and water contaminating the area surrounding Fukushima Daiichi Nuclear Power Station, and by consequence Japan as a whole. The focus of this brief analysis is the detection of radioactive material from the accident with an emphasis upon detection of isotopes of Cesium, $^{134} \text{Cs}$ and $^{137} \text{Cs}$. Radio-cesium does not occur in nature in any significantly detectible quantity and represents an excellent marker for the presence of man-made nuclear fallout. Radio-cesium isotopes present a biological hazard as well, if introduced into the environment, given the similarities in biological and chemical behavior of cesium to that of potassium, an element necessary to the human body ("Cesium"). The dispersal of $^{134} \text{Cs}$ and $^{137} \text{Cs}$ is facilitated by their ease of transport via hydration and resulting precipitation, known as “washout”. A study concerning the release of $^{133} \text{Xe}$ and $^{137} \text{Cs}$ into the atmosphere (Stohl, et al, 2011) found that $^{137} \text{Cs}$ emissions from the damaged reactors at Fukushima may have reached between 23.3 Peta Bq and 50.1 Peta Bq (one Peta Becquerel equals $1 \times 10^{15}$ Becquerels), approximately 42% of the estimated emission from the Chernobyl disaster. It should be noted that these nuclear disasters were quite different in nature and that their comparison of total radio-cesium release purely concerns the magnitude of radioactive release. A significant portion of the radiation release was swept into the Pacific Ocean, though much of the Japanese countryside near the plant was contaminated, especially the immediate area surrounding the plant.

Soil samples from contacts living within Japan were gathered by a central contact who compiled their documentation and sent them to the United States for analysis. The samples were received in good order, each well documented and carefully packaged. Each set of samples had a chain of custody that had been maintained from source to destination. Soil from locations closer to Fukushima could not be obtained, due partially to the difficulty of finding a source to obtain samples, as well as difficulties in shipping soil with activities, which may exceed the standards and rules for transportation of radioactive material.

Methods

Soil samples taken from three different geographic locations in Japan were obtained and tested using gamma spectroscopy. From each location sampled, two small samples of soil were obtained at two different areas. The locations sampled were Kashiwa City, Chiba Prefecture, approximately 26 km north east of Tokyo and 210 km south west from the Fukushima plant, Shiga Prefecture, towards the center of Japan and east of Osaka, approximately 550 km south west from the Fukushima plant, and Saga City, Saga Prefecture, at the far southern end of Japan, being approximately 1060 km south west from the Fukushima plant. These locations display a relatively even distribution from southern, middle, and lower-
northern Japan.

Each sample was taken from soil far outside of the exclusion zone. Most samples were taken from public or residential areas in land implicitly deemed “safe”, insomuch as not being part of the exclusion zone surrounding the nuclear plant. The samples with the greatest measured activity were those taken from a playground and from soil under a water spout, in Kashiwa City. The only sample which was slightly askew of this logic was a sample, sample J-C, taken from under a house made before the disaster. The benefit of this sample was that it provided a snap-shot of per-disaster soil, though the possibility of contamination via rain water and atmosphere cannot be ruled out.

Given the natural trace radioactive composition of soil, e.g., thorium, potassium, uranium, etc., the need to discriminate between Naturally Occurring Radioactive Material, herein NORM, and those artificially produced, Gamma spectroscopy was chosen. Gamma spectroscopy captures gamma rays and X-rays, producing a map of energy detected vs. numbers of detected events. Concentrations of detected events create what amounts to a unique fingerprint of a radioisotope. The decay scheme of a radioactive element may include the emission of gamma rays. The decay schemes of $^{137}$Cs and $^{134}$Cs result in the formation of stable barium isotopes. $^{137}$Cs decays via beta minus decay into radio-barium, either $^{137}$Ba or $^{137}$Ba $m$, a metastable state of $^{137}$Ba lasting 153.12 seconds and resulting in a photon of 661.66 keV (one kilo electron volt equals $10^3$ electron volts) being released to reduce the metastable state to a stable state, $^{137}$Ba. $^{134}$Cs decays via beta minus decay, becoming $^{134}$Ba. $^{134}$Ba emits several different gamma rays at several different energies to achieve a stable state, most notably 604.72 keV.

A full determination of all detectible isotopes was performed, but the primary focus was set upon radio-cesium, due to the presence of radio-cesium in half of the samples. For radio-cesium analysis, the two most intense gamma energies from each of the isotopes, $^{137}$Ba and $^{134}$Ba, where chosen for isotopic analysis. From $^{134}$Ba, the two most common energies are 604.72 keV with intensity of 0.9762, and 795.86 keV, with an intensity of 0.8553. Additionally, a third gamma energy was chosen from $^{134}$Ba given that the first two most intense energies from $^{134}$Ba come from a similar energy region, thus 569.33 keV, with an intensity of 0.1538. From $^{137}$Ba, the two most common energies are 661.66 keV with an intensity of 0.851, and 32.19 keV, (as X-Rays) with an intensity of 0.036, the latter being the result of internal conversion rather than a direct gamma. It should be noted that X-rays are photons which are emitted from the electrons surrounding an atom while gamma rays are photons emitted from the nucleus of atoms.

Gamma spectroscopy was performed using a gamma scintillation counter and a multichannel spectrum analyzer. The scintillation counter used was a Radiation Sensors LLC. model 6S6P1.5VDC2 38mm NaI(Tl) (NaI(Tl) refers to the detection crystal used, a Sodium Iodide Thallium doped), which typically provides a 7% resolution, and the multichannel
analyzer was a Spectrum Techniques LLC. model UCS30 Universal Computer Spectrometer (Fig. 1).

The gamma scintillation counter was held vertically in a testing fixture with the detector crystal pointed downward. The entire detector crystal region of the scintillation counter was wrapped in a toroidal mass of lead, having a radius of

**Gamma Spectrometer**

a) Radiation Sensors LLC. Model 656F1.5VDC2 38mm Na(Tl)
b) Testing chamber with graduated tray slots
c) Lead blocks 38.1 mm x 19.05 mm x 76.2 mm in size.
d) Toroidal mass of lead, having a radius of 57.15 mm and an outer band thickness of 38.1 mm.
e) High voltage power and data cables come from and return to Spectrum Techniques LLC. model UCS30 Universal Computer Spectrometer.
f) Four layers of 3mm high density lead sheeting, wrapped in aluminum foil.
g) Plastic testing tray for specimen.
h) 0.5 mm aluminum beta shield.
i) Borosilicate glass sample canister containing sample.

![Diagram of gamma spectrometer used.](image)

57.15 mm and an outer band thickness of 38.1 mm. The photomultiplier tube comprising the upper two thirds of the scintillation counter was shielded within four layers of 3mm each high density lead sheeting, wrapped in aluminum foil. The sides of the detection chamber were shielded by stacks of tiny interlocking lead blocks. Each lead block measures 38.1 mm x 19.05 mm x 76.2 mm in size. Surrounding the entire scintillation counter were a stack of greater than 40 such blocks of lead forming an interlocked mass. The purpose of the lead was to reduce the incidence of background radiation as much as possible, providing a clearer spectrum.

Reduction of natural background radiation within the 1 keV to 1030 keV energy spectrum was required to facilitate isotope detection. Given a Half-value layer (The Half-value layer of a material is the amount of a material require to reduce the radiation intensity it shields by half) for lead for gamma energies of 1 MeV (one mega electron volt equals $10^6$ electron volts), of 8.61 mm (Wolfram|Alpha, 2012), a rewritten exponential decay equation revealed that the lead blocks, having a thickness of 38.1 mm, would ideally prevent $95.3450\%$ of gamma energies at or below 1 MeV (Fig 2).

$$100e^{\left(\frac{\ln 2}{8.61 \text{ mm}}\right)38.1 \text{ mm}} = 4.65496$$

*Fig 2. Calculation of lead maximum shielding.*

The remaining 4.655% of gamma energy at or under 1 MeV was deemed acceptable background. Tests with use of the lead blocks indicated that the background count was considerably reduced. Originally, a ten minute multichannel scaling analysis
provided an average background of 3,546 detections per minute (59.1 detections per second), whereas the same analysis, with the lead fully encapsulating the unit, as was used during the subsequent tests, showed a more acceptable average background of 244.5 detections per minute (4.075 detections per second).

Calibration of the gamma spectrometer was initially performed after a shielded baseline was established. Initial calibration was performed using a 37,000 Bq $^{137}$Cs isotope source and a two point energy calibration for energies 661.66 keV and 32.19 keV. Settings for the scintillator were chosen to allow focus on a single 1 MeV segment of gamma energies from the samples. The minima and maxima discriminators for the test were set at channels (13,1024) and the energy calibration range was calculated to encompass 10 keV to 1030 keV, the full energy aperture, discriminated, of 1,020 keV. Subsequent tests were performed over the course of several days and at several time periods for optimal thermal calibration to account for thermal drift. The energy calibration analysis and subsequent recalibration, for the purposes of fine tuning, was performed using standard commercial gamma sources with known primary energies. The following isotopes were used to correct the spectrum calibration: $^{137}$Cs, $^{241}$Am, $^{152}$Eu, $^{40}$K, $^{238}$U, and $^{232}$Th. Uranium and Thorium samples, and their related progeny, were used for proper identification of NORM, the focus being on their more gamma intense daughters, while $^{40}$K was merely used to ensure the proper termination of the high end of the spectrum energy aperture.

Final calibration was initially via a three-point linear approximation derived from a plot of each known gamma energy from the afore mentioned sources. The change in detected energy with respect to each channel was not a linear relationship, but rather a quadratic relationship; a curve bending slowly upwards in magnitude as it progressed upwards in channel number. The rate of change was too low for a successful three point linear approximation to be within a tolerance of +/- 5 keV/channel. This problem was likely the result of the 1 MeV energy aperture used, which was confirmed during a Delaunay triangulation (fig. 3, Wolfram|Alpha, Delaunay triangulation, 2012) of the plotted data. Use of a basic three-point calibration was relegated to general energy determination, with an approximate tolerance of +/- 10 keV, being too inaccurate for the fine accuracy require for isotope detection. During actual isotopic analysis, a Riemann sum-based method was

![Graph showing channel vs. energy detected in keV](image)

**Fig 3.** x axis represents channels, y axis represents energy detected in keV.
chosen, using small 100 or 200 keV segments, each containing a quadratic curvature which more closely approximated a line. The segments were individually calibrated using intersecting gamma lines from known sources, primarily $^{152}$Eu. The result was a much more thorough and accurate analysis. For simplification, an average calibration constant of 1.05 (0.05), describing change in energy with respect to channel such that $\text{Energy keV} = 1.05(0.05) \times \text{Channel}$, can generally described the channel/energy relationship, though the actual $\Delta E/\Delta \text{channel}$ progression was not linear, but quadratic.

Sample activity was determined using a 37,296.6532 Bq $^{152}$Eu source which was fully calibrated against a NIST traceable mixed gamma source (source ID SRS:80899-854). The calibrated source was tested using a HPGe detector and determined to have an activity of 37370 Bq with an accuracy of +/- 5%. The $^{152}$Eu source was calibrated February 14, 2012 and had a calculated activity of approximately 37,296.6532 Bq on February 28, 2012 during the final activity analysis. The variance between stated activity and calculated activity was less than 0.19 % difference. $^{152}$Eu was chosen over a calibrated $^{137}$Cs because of the more widely distributed photon energy range emitted.

The UCS30 gamma spectrometer automatically calculates the approximate dead time for each interaction, maintaining a real-time timer, which counts in real seconds, as well as an adjusted time, called “live time”, which adjusts for each period of time that the detector is unable to count due to having just detected an event. The variance between real and live time typically amounts to a few seconds lost to events per many thousands of seconds of real time, for most low activity samples. The calibrated $^{152}$Eu source was tested for 300 live-time seconds with a control soil sample placed between the source and the detector to simulate the soil absorption of the actual samples (fig. 4). The activity of each sample was measured over 300 seconds and the results were divided into the total counts of the calibrated source to produce a ratio of the counts of the sample with respect to the counts of the calibrated source (fig. 5). For each sample, ratio of the counts of sample over the calibrated source was multiplied by the known activity of the calibrated source to determine the inferred activity of the sample (Fig 6). The final results were divided by the mass of each sample, producing activity per gram, and

![Fig 4. Initial activity calibration using calibrated source and control soil.](image-url)
then multiplied by 1,000 to produce activity per kilogram.

The detector was unshielded for the entire activity test to reduce Compton scattering (Compton scattering occurs when gamma rays are deflected by atomic interactions, loosing energy and often causing secondary detections) and other detector artifacts caused by shielding. Background count was not removed from the source because activity was based upon a ratio and not on the actual count rate.

Samples were labeled and cataloged using the prefix, “J” and a single letter or letter/number pair to distinguish each from the other samples. The samples were received in small plastic bags or in small plastic canisters. Most samples remained in their original container, unless removed for testing. Testing was performed using borosilicate laboratory glassware, e.g., a 100 ml beaker or 300 ml Florence flask, in order to accurately determine volume and mass, but without changing activity. To ensure the latter, a test was performed in which a fully calibrated $^{137}$Cs source of 3733.2282 Bq was tested at an exact geometry from the scintillation detector both with and without a borosilicate glass piece above it. The change in counts was far less than 1 detection per second, falling well within acceptable tolerances. The most likely result of boronsilicate interactions with gammas would be a minor deflection with a slight change in energy, thus a broadening of the photo-peaks, though this predicted effect was never observed. The source used was fully calibrated against a NIST traceable mixed gamma source (source ID SRS:80899-854). The calibrated source was tested using a HPGe detector and determined to have a accuracy of +/- 5%. The $^{137}$Cs source was calibrated January 4, 2012 at 3737 Bq and had a calculated activity of approximately 3733.2282 Bq on January 20, 2012 during the final activity analysis. Careful attention was given to handling samples which might contain loose radioactive material, such as radio-strontium or radio-cesium. Latex gloves, a paper face mask, goggles, and a lab coat were used during the transfer of some of the samples to long-term storage containers. An SE International Inspector EXP+ Geiger counter was present and powered on for all tests and material handling.
Results and Discussion

Samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>Weight (g)</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>J-A</td>
<td>20</td>
<td>Soil from residential yard under rain spout. Kashiwa City, Chiba Prefecture, Japan.</td>
</tr>
<tr>
<td>J-B</td>
<td>19</td>
<td>Soil from a playground. Kashiwa City, Chiba Prefecture, Japan.</td>
</tr>
<tr>
<td>J-C</td>
<td>85</td>
<td>Soil from under a house built before March 2011. Shiga Prefecture, Japan.</td>
</tr>
<tr>
<td>J-D</td>
<td>57</td>
<td>Soil from a public park. Shiga Prefecture, Japan.</td>
</tr>
<tr>
<td>J-S1</td>
<td>94</td>
<td>Gravel from the surface of a baseball field beside Saga prefectural offices, Saga City, Saga Prefecture, Japan.</td>
</tr>
<tr>
<td>J-S2</td>
<td>24</td>
<td>Soil from a drainage ditch in Saga City, Saga Prefecture, Japan.</td>
</tr>
</tbody>
</table>

Sample Activities (Gross Gamma Only)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Activity (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>J-A</td>
<td>97,590.3</td>
</tr>
<tr>
<td>J-B</td>
<td>41,668.7</td>
</tr>
<tr>
<td>J-C</td>
<td>9,664.0          **</td>
</tr>
<tr>
<td>J-D</td>
<td>7,225.1</td>
</tr>
<tr>
<td>J-S1</td>
<td>8,683.6          **</td>
</tr>
<tr>
<td>J-S2</td>
<td>16,959.6         *</td>
</tr>
</tbody>
</table>

* Increased activity is likely the result of primary decay activity from NORM via $^{232}$Th and progeny.
** An average of the two sample containers was used to make this determination.

Isotopes Identified

<table>
<thead>
<tr>
<th>Sample</th>
<th>Identified Isotopes</th>
</tr>
</thead>
<tbody>
<tr>
<td>J-A</td>
<td>Cs137, Cs134 †</td>
</tr>
<tr>
<td>J-B</td>
<td>Cs137, Cs134 †</td>
</tr>
<tr>
<td>J-C</td>
<td>NORM: Thorium series indicated by a strong Pb-212*, 137 Cs</td>
</tr>
<tr>
<td>J-D</td>
<td>NORM: Thorium series indicated by a strong Pb-212*, 137 Cs</td>
</tr>
<tr>
<td>J-S1</td>
<td>NORM: Thorium series indicated by a strong Pb-212*</td>
</tr>
<tr>
<td>J-S2</td>
<td>NORM: Thorium series indicated by a strong Pb-212*, Trace 137 Cs</td>
</tr>
</tbody>
</table>

† Cesium isotopes were of such activity that they effectively hid any other isotopes.
* Uranium and Thorium decay progeny are often found in soil. These isotopes are naturally occurring and permeative.

Sample J-A

Sample J-A, a mass of 20g of soil, came from under a rain spout in a residential area of Kashiwa City, Chiba Prefecture, Japan. Sample J-A contained both the soil from the area as well as residue from water raining down upon it. The distance from the rain spout to Fukushima is approximately 210 km, potentially receiving some of the fallout. The sample was tested with a Geiger counter and determined to have a contact reading of 300 (20) CPM above background. The specimen was removed from its plastic casing and carefully tested with a Geiger counter, using a binary search method, to determine if the detected radioactivity was the result of large particulate contaminants or a generally diffuse contamination. The results were a normal distribution of the activity throughout the sample. The sample was placed within a laboratory glass sample container for spectroscopic testing.
Only 300 seconds into the spectroscopic testing of the sample, the results were already clearly evident. Peaks for $^{134}$Cs and $^{137}$Cs were extremely prominent, overriding all over data. The activity of the radio-cesium caused Compton scattering covering half of the entire spectrum effectively hiding any NORM or other potentially detectable radio-isotopes. Further tests will need to be performed using extended testing times, perhaps 48 hours, and no lead shielding, reducing the Compton scatter. As a secondary precaution, given the levels of radio-cesium found, a sample of $^{137}$Cs with an approximate activity of 37,000 Bq was compared to the J-A peaks to fully validate the results.

Sample J-A came from an area of north eastern Tokyo, merely 26 km north east of the center of Tokyo. The area where the sample came from is highly populated and home to thousands of Japanese citizens. The soil gross gamma activity was determined to be 97,590.3 Bq/kg. NORM activity calculation cannot be determined without either waiting perhaps a few decades for the decay of the radio-cesium isotopes or without chemical separation of radio-cesium. Ostensibly, other fissile products may be contained within the sample, but this cannot be confirmed. There concentrations would be based upon availability of transport and fissile probabilities. $^{134}$Cs and $^{137}$Cs are absolutely present in the sample in quantities far beyond what could be considered trace. Indications for NORM are unobtainable using an NaI(Tl) detector using standard configurations, given the radio-cesium. A Fukushima fallout hypothesis is clearly evident.

Sample J-B

Sample J-B demonstrated nearly identical isotopic properties to sample J-A, though lower in overall activity, 41,668.7 Bq/kg. Sample J-B contained a mass of 19g of soil from a children's playground in Kashiwa City, Chiba Prefecture, Japan. The location of the playground can be found at Google Map coordinates, 35.86618, 139.965311. Using a
Geiger counter, the sample measured 120 (20) CPM above background. The specimen was removed from its plastic casing and carefully tested with a Geiger counter, using a binary search method, to determine if the detected activity was the result of large particulate contaminants or a generally diffuse contamination. The results were a normal distribution of the activity. The sample was placed back within its original container for spectroscopic testing.

After only 300 seconds into the spectroscopic testing, just as with sample J-A, the results were clearly evident. Peaks for $^{134}$Cs and $^{137}$Cs were prominent, overriding all over data. The activity of the radio-cesium caused Compton scattering covering half of the entire spectrum effectively hiding any NORM or other potentially detectible radio-isotopes. Further tests will need to be performed using extended testing times, perhaps 48 hours, and no lead shielding, reducing the Compton scatter. As a secondary precaution, given the levels of radio-cesium found, a sample of $^{137}$Cs at 37,000 Bq was compared to the J-B radio-cesium peaks to fully validate the results. Sample J-B was compared to sample J-A, with matching peaks and relative intensities, confirming the results.

Sample J-B came from an area of north eastern Tokyo, merely 26 km north east of the center of Tokyo, like sample J-A. The sample came from a playground where children play. The soil for the playground is a packed dirt where the children can run free. Behind the playground is a full pool and additional recreation equipment. Beyond radio-cesium, the presence of other fallout radio-isotopes cannot be determined. The soil is simply too high in radio-cesium activity to be tested for other trace fallout products using an NaI(Tl) detector. $^{134}$Cesium and $^{137}$Cesium are absolutely present in the sample in quantities far beyond what could be considered trace. Indications for NORM are unobtainable using an NaI(Tl) detector in my configuration, given the radio-cesium activity. A Fukushima fallout hypothesis is clearly evident.
Sample J-C

Sample J-C contained a total mass of 85g of soil within two sample containers. Sample J-C came from beneath a house, built before March 2011, located in Shiga Prefecture, Japan. The samples produced no noticeable activity from a Geiger counter. NORM was detected in the form of Thorium 232, via $^{232}$Th progeny. Energy peaks from isotopes commonly associated with the Thorium 232 decay chain were found in equilibrium, including gamma energy lines for $^{212}$Pb. Indications of minor peaks, which supported equilibrium ratios where: $^{228}$Th at 215.99 keV, 131.61 keV, and $^{212}$Pb at 238.63 keV.

![Gamma Spectrum of sample J-C](image)

A minor increase in counts was detected in both the [650, 670] keV and [30, 33] keV ranges. The lower energy X-rays at [30, 33] keV were barely distinguishable from the background, but the gammas at [651, 667] keV produced a small peak, clearly evident above other peaks in the same energy region. The total counts for the entire run, with background subtracted, were 602, suggesting a detection of merely $6.97\times10^{-3}$ CPS for the entire peak, fully double the 331 counts of 8 channels before the peak and the 8 channels after the peak. Though small in activity, this peak does hint at the presence of $^{137}$Cs, at least in trace quantities. The total activity of sample J-C was 9,664.0 Bq/kg. The afore mentioned $^{228}$Th and $^{212}$Pb daughter isotopes support a hypothesis of NORM via the thorium 232 decay chain. $^{137}$Cs can be considered due to the small indications of activity, yet a lack of detectible $^{134}$Cs prevents a definite determination of origin.

Sample J-D

Sample J-D contained a total mass of 57g of soil. The soil came from a public park in Shiga Prefecture, Japan. The samples produced no noticeable activity from a Geiger counter. The spectrum displayed common peaks from the Thorium 232 decay chain. Equilibrium of thorium decay chain daughters was detected and common gamma energy lines for $^{232}$Th
progeny were identified: $^{228}$Th at 215.99 keV, $^{220}$Rn at 549.73 keV, $^{212}$Pb at 238.63 keV, and $^{212}$Bi at 727.33 keV.

![Gamma Spectrum of sample J-D](image)

**Fig 10. Gamma Spectrum of sample J-D**

A minor increase in counts was detected in both the [657, 676] keV and [28, 34] keV ranges. The lower energy X-rays at [28,34] keV were barely distinguishable while gammas at [657, 676] keV produced a small and clear photo-peak. The peak was well formed and normal, coming to an apex at 657.9 keV, clearly disguisable for a probable $^{137}$Cs detection on an NaI(Tl) detector. The similarities of samples J-C and J-D, from a NORM perspective, are attributable to their geographic proximity. The total calculated activity of sample J-D was 7,225.1 Bq/kg. The occurrence of radio-cesium is attributable to the transport mechanism of radio-cesium via water. The thorium decay series isotopes, $^{228}$Th, $^{220}$Rn, $^{212}$Pb, and $^{212}$Bi, support a hypothesis of NORM via the thorium 232 decay chain. $^{137}$Cs can be considered due to the small indications of activity, yet a lack of detectible $^{134}$Cs prevents a definite determination of origin.

**Sample J-S1**

Sample J-S1 contained the aggregate rocks and sand. The sample had a total mass of 94g and contained gravel from the surface of a baseball field beside Saga prefectural offices, Saga City, Saga Prefecture, Japan. The canisters produced no noticeable count change using a Geiger counter. The calculated activity of J-S1 was 8,683.6 Bq/kg. The spectrum displayed common peaks for the Thorium 232 decay chain. Equilibrium of daughters was evident in minor irregularities in the spectrum around common primary intensity gamma energy lines for $^{232}$Th progeny. Equilibrium of thorium decay chain daughters was detected and common gamma energy lines for $^{232}$Th progeny were identified: $^{228}$Th at 215.99 keV, 131.61 keV, $^{220}$Rn at 549.73 keV, $^{212}$Pb at 238.63 keV, and $^{212}$Bi at 727.33 keV. The thorium decay series isotopes, $^{228}$Th, $^{220}$Rn, $^{212}$Pb, and $^{212}$Bi, support a hypothesis of NORM via the thorium 232 decay chain. No unnatural isotopic indications were found.
Sample J-S2

Sample J-S2 contained soil and sediment from a drainage ditch in Saga City, Saga Prefecture, Japan, with a mass of 24g. The sample produced no noticeable count change from a Geiger counter. A minor increase in counts was detected in both the [600, 700] keV and [30, 34] keV ranges. Though these ranges are seen clearly in samples J-A and J-B, they were insubstantial to prove radio-cesium activity in sample J-S2, but given the proximity to Fukushima of 1160km, the slight variance cannot be merely ignored. The overall sample activity, 16,959.6 Bq/kg, was 1.953 times greater than sample J-S,
from NORM washout. Radio-cesium can readily bond with water to form cesium hydroxide and hydrogen gas, allowing transport via precipitation. The sample origin, being from a water drainage ditch, provides a reasonably likely mechanism for both NORM and radio-cesium hydroxide washout. Pronounced peaks in the lower energy ranges support a NORM hypothesis via the thorium decay chain. $^{232}$Th progeny were identified: $^{228}$Th at 215.99 keV, $^{220}$Rn at 549.73 keV, $^{212}$Pb at 238.63 keV, and $^{212}$Bi at 727.33 keV. The thorium decay series isotopes, $^{228}$Th, $^{220}$Rn, $^{212}$Pb, and $^{212}$Bi, support a hypothesis of NORM via the thorium 232 decay chain. $^{137}$Cs can be considered due to the small indications of activity, yet a lack of detectible $^{134}$Cs prevents a definite determination of origin for sample J-S2.

**Conclusion**

The presence of high concentrations of radio-cesium in soil samples from Japan is not an unexpected outcome, given current events. Radio-cesium is a very common fallout product from nuclear fission and easily transported by water. Though man-made and never naturally occurring in any detectible quantity, $^{137}$Cs can have several possible points of origin, such as legacy fallout from nuclear testing, or even residue from the Hiroshima or Nagasaki bombs. Given the trace levels found in one of the two most southern samples, J-S2, and without an equilibrium of $^{134}$Cs detected, the origin of this man-made isotope cannot be verified. The origin of radio-cesium in samples J-A and J-B, however, are absolutely linkable to Fukushima reactor release, given the short half-life of $^{134}$Cs, found in quantity with $^{137}$Cs, in these samples. Had these same samples been tested March or April of 2011, a significant presence of radio-iodine would more then likely been found in samples J-A,B,C, and D, and perhaps S1 and S2. The distribution of the fallout was a stark reminder of the influences of weather and precipitation on the overall distribution of radioactive fallout from nuclear disasters. The resulting fallout contamination zones that would result from a radiological attack or nuclear disaster are as predictable as the weather, and given to a series of probabilities.

The radioactive contamination of parts of northern Japan, though terrible, provides the world with a rare chance to study the results of what can go wrong. Much like any accident, lessons learned will decide the full weight and impact of events. With any luck, we will not be doomed to repeat our unfortunate mistakes. The most unfortunate results of the contamination of Japan will probably not be felt now, but in twenty or more years by the grown children who play in the playground in Kashiwa city, or the park in Shiga Prefecture. It is these children who may absorb radio-cesium into their blood and radio-strontium into their bones and teeth. Their legacy may be similar to the extended victims of Chernobyl, Hiroshima, and Nagasaki.
References


