Fukushima: Science Behind the Scenes

by: Michael Apsokardu and Guotao Lu

Since the meltdown of three nuclear reactors following a tsunami and earthquake in 2011, seawater has been pumped into the Fukushima site to cool the nuclear reactors. Both water that has been intentionally pumped in and groundwater leeching to the reactors were carefully collected, and then went through an Advanced Liquid Processing System (ALPS) before being sent to storage tanks. The ALPS is a system that is in place to remove radiochemical species prior to the storage tank. According to the Tokyo Electric and Power Company (TEPCO), the system is designed to remove 62 different radiochemical species. More than 1.2 million tons of treated seawater have been stored as of today (*CNN source*).

Fukushima nuclear site (courtesy to *New York Times*)

Besides the ALPS, water in the storage tanks at the Fukushima site is continually monitored and tested for their radioactivity levels. For storage tanks exceeding the limits of release threshold, the water is filtered again by the ALPS until reaching the safety level. With the establishment of this firm testing policy and adherence to environmental regulations, the United States of America and the International Atomic Energy Agency (IAEA) and are in line with the World Health Organization regulation standards to support the proposal to release the stored water into the ocean within the next two years. The IAEA will continue to play an active role in this regulation and implementation plan at the Fukushima site moving forward (*Japan Today*).

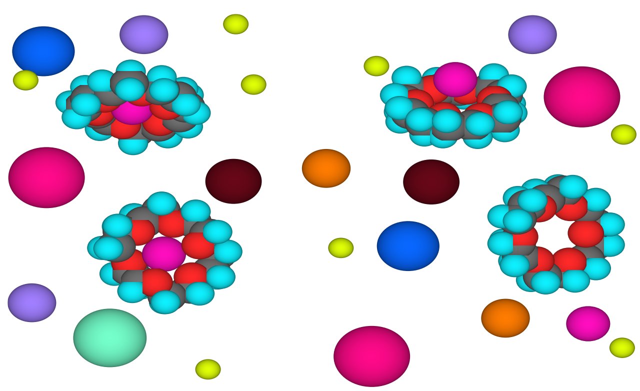
Some of the major species of interest are listed below along with their respective half-lives. The half-life is the time required for half of the isotope to decay and become stable. Tritium, an isotope of hydrogen, is the most abundant radionuclide present in the water with concentrations at least 3-4 orders of magnitude higher than the other radionuclides. Tritium is a low energy beta-emitter, and these levels of tritium are widely considered to be safe (Buesseler *Science* **2020)**.

Selected radionuclides at the Fukushima site and their half-lives. Adapted from Buesseler *Science* **2020**.

Radionuclide	Half-Life (years)
^{3}H	12.4
^{90}Sr	29.1
^{134}Cs	2.1
^{137}Cs	30.0

Cesium and strontium, on the other hand, are radiochemical species requiring more care in removal from the water. Cesium is highly soluble in water and easily transported in surface and ground water. Additionally, the length of the half-life is of a concern. Cesium is an emitter of both beta and gamma radiation and is harmful to humans when ingested. Biodistribution of cesium tends to favor tissues such as the muscle. These tissues then become subject to damage as a result of the high energy gamma radiation (*CDC source*). Strontium, similar to cesium, is difficult to remove from water and has an equally long half-life. Strontium is a beta emitting species. Upon ingestion, strontium tends to deposit in the bones, increasing the likelihood of the formation of bone tumors (Uesugi, M.; et. al.; *Talanta* **2018**).

The testing methods for these radionuclides are widely developed and accepted. The list of 62 radionuclides removed by the ALPS contains a mixture of both beta and gamma emitting radiation. Concentrations of gamma radiation in water samples can be measured by gamma ray spectrometry, while beta radiation is often measured using liquid scintillation. In terms of sample preparation, tritium is a special case in the group in that it is difficult to separate from water for measurement but is already present in high enough concentrations to be directly measured from water samples. Other radionuclides often require extraction and concentration steps prior to measurements of radioactivity.



Empore™ radiation SPE disks use molecular recognition technology to selectivity extract and concentrate radioactive species such cesium and strontium.

Empore™, a legendary solid phase extraction (SPE) sampling membrane invented by 3M and now manufactured by CDS Analytical (Oxford, Pennsylvania, USA), is producing novel SPE disks to selectively extract radioactive cesium and strontium. The Empore™ SPE technology functions by pulling, by vacuum, up to one liter of water through the disk trapping target chemical species on sorbent particles embedded in an inert polymer matrix. Target chemical species are then extracted from the disk with an appropriate solvent and concentrated. In the case of cesium and strontium, these unique Empore™ products become trapped with the use of molecular recognition technology.

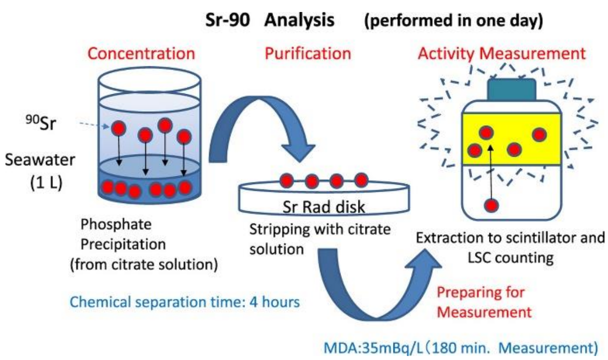


Table 9 Determination of ^{90}Sr in seawater samples near the Fukushima Daiichi NPP

Method	Sample (1 L)	Recovery (%)	Measured ^{90}Sr (Bq/L)
(a) Direct measurements of ^{90}Sr (New method)	Outside of FDNPP Harbor (2013.11.10)	85*	0.11 ± 0.02
	Ukedo Harbor (2014.8.7)	85*	0.10 ± 0.02
	Sr-90 (spiked with 0.166 ± 0.005 Bq)	85*	0.16 ± 0.02
	Sr-90 (spiked with 33.2 ± 0.5 Bq)	85*	33.9 ± 3.1
	IAEA-TEL-2015-03 No.01 (30.1 ± 0.9 Bq)***	85*	27.9 ± 2.8
(b) Measurements of separated ^{90}Y (GDA extraction method)**	Outside of FDNPP Harbor (2013.11.10)	80	0.13 ± 0.02
	Ukedo Harbor (2014.8.7)	83	0.11 ± 0.02
	Y-90 (spiked with 0.166 ± 0.005 Bq)	83	0.23 ± 0.03
	Y-90 (spiked with 33.2 ± 0.5 Bq)	75	27.2 ± 2.8

Process for extraction of strontium using the Empore radiation SPE disk (left) and the measured recovery and radioactivity of strontium in numerous locations near the Fukushima site (right). Hirayama, Y.; et. al.; *J. Environ. Radioact.* **2020**.

The methods using both cesium and strontium radiation disks have been well-established in recent water sampling applications near the Fukushima site. In one example, scientists are measuring ^{90}Sr in bodies of water near the Fukushima site and multiple surrounding bays in harbors. In this method, ^{90}Sr is first selectively extracted from the water samples using the Empore™ ^{90}Sr disk and is then measured using an extraction scintillator. Empore™ disks are a critical component to developing methods providing results faster with high accuracy and reproducibility. The recoveries and radioactivity at these sites can be seen in the figure above. (Uesugi, M.; et. al.; *Talanta* **2018**). In a second example, scientists are measuring ^{134}Cs and ^{137}Cs near the Fukushima site. Using the Empore™ Cs radiation SPE disk, ^{134}Cs and ^{137}Cs are selectively extracted from water samples and then measured with the use of gamma ray spectrometry. The results in the figure below show the decrease in cesium radioactivity over a four-year period in an isolated lake. Researchers were able to determine that isotopic cesium was transported with through the bed of the lake to other areas surrounding the Fukushima site. With these methods scientists are demonstrating the ability track the movement of water, and radiochemical species, through underground water systems (Hirayama, Y.; et. al.; *J. Environ. Radioact.* **2020**). These methods demonstrate a need for reliable and reproducible sample preparation procedures that scientists will be able to call on for water sampling of radiochemical species.

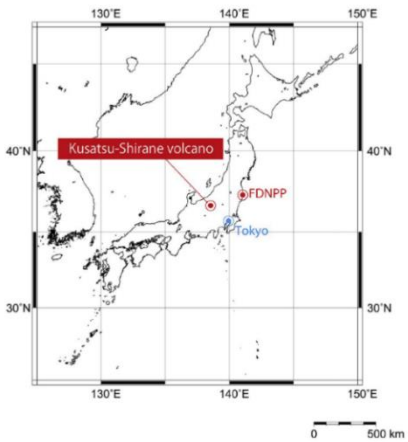


Fig. 1. Locations of Kusatsu-Shirane volcano and the Fukushima Dai-ichi Nuclear Power Plant (FDNPP).

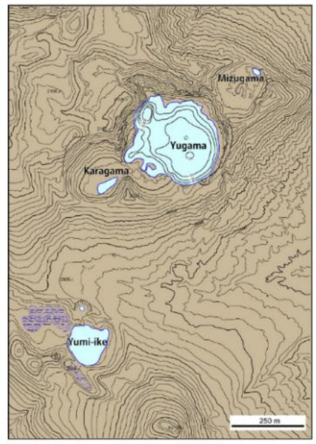


Fig. 2. A map around the summit area of Kusatsu-Shirane volcano. Contours are in meters. There are four crater lakes named Yugama, Mizugama, Karagama and Yumi-ike in the summit area.

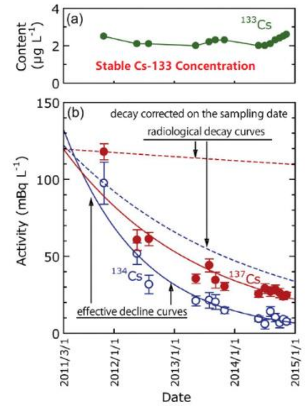


Fig. 5. The time series data of the contents of cesium in Yugama. The data for radioactive cesium were decay-corrected on the sampling date. Error bars in (b) represent errors in gamma-ray counting. Dotted lines and solid lines in (b) show the radiological decay curves and the effective decline curves, respectively.

Location of the Fukushima site (left), topography of the land (middle), and decay of isotopic cesium from 2011 to 2015 (right). Taken from Uesugi, M.; et. al.; *Talanta* **2018**.

Our world will be stronger when we collaborate and use science to develop cohesive strategy to fight against natural diseases and disasters. This goal requires global efforts from constantly observing, monitoring and analyzing scientific data to drive decisions that offer the greatest social, economic and environmental benefits. The efforts paid at Fukushima is one of such classic examples.