THE OFFICIAL MAGAZINE OF THE OCEANOGRAPHY SOCIETY

Oceanography

CITATION

Buesseler, K.O. 2014. Fukushima and ocean radioactivity. *Oceanography* 27(1):92–105, http://dx.doi.org/10.5670/oceanog.2014.02.

DOI

http://dx.doi.org/10.5670/oceanog.2014.02

Fukushima and Ocean Radioactivity

BY KEN O. BUESSELER

ABSTRACT. The triple disaster of the March 11, 2011, earthquake, tsunami, and subsequent radiation releases from Japan's Fukushima Dai-ichi nuclear power plant were unprecedented events for the ocean and society. In this article, the radioactive releases from this event are compared to natural and prior human sources, with particular attention to cesium-137 and -134 radioisotopes. Total releases from Fukushima are not well constrained, with estimates from atmospheric fallout and direct ocean discharge spanning 4 to 90 peta Becquerels (PBq), but are most likely in the 15–30 PBq range. This source is smaller than any ^{137}Cs remaining in the North Pacific from global and close-in fallout from the 1960s. It is of similar magnitude to ^{137}Cs released to the ocean from the Sellafield nuclear reprocessing site on the Irish Sea, though of greater magnitude than fallout that reached the ocean from the 1986 Chernobyl nuclear power plant disaster in the Ukraine. The fate of Cs is largely determined by its soluble nature in seawater, though uptake in sediments does occur via cesium's association with both detrital particles and biological uptake and sedimentation. A mass balance of Cs supply from rivers and ongoing leakage from nuclear power plants suggests that sediments will remain contaminated for decades. This may be one reason why Cs concentrations in benthic fish stay elevated over predictions, causing fisheries to remain closed near Fukushima and ongoing concern to the public.

SOURCES OF RADIONUCLIDES TO THE OCEAN

We live in a radioactive world. Many of Earth's radioactive elements, or radionuclides, are contained in the solid body of the planet and are remnants of its formation. Because we live on an ocean planet, it should be no surprise that the waters of our ocean also contain many different radionuclides. Some of them are also natural byproducts of planetary and celestial processes, but some are the result of recent human activity, and many natural and anthropogenic radionuclides can be used to reveal crucial details about oceanic and planetary processes.

In the ocean, the most abundant natural radionuclide is potassium-40 (^{40}K), and it is relatively evenly distributed. It has a concentration, or radionuclide "activity," of 11,000 Bq m^{-3}, with one Becquerel (Bq) being a measure of one radioactive decay event per second. Although the number is large, this level of activity is quite small from a human health perspective. Even the total quantity of ^{40}K, about 15 million PBq, with one PBq equivalent to 10^{15} Bq, is of little concern when spread across the entire ocean (Figure 1).

One of the next most abundant natural radionuclides is uranium-238 (^{238}U), with a total ocean inventory of around 37,000 PBq. Both the potassium and the uranium radionuclides are derived ultimately from the weathering of rocks. ^{238}U in particular is a key element of interest to ocean scientists due to its long decay chain and several intermediate decay products, each of which has different chemical and radioactive properties that have propelled their use as tracers to study ocean currents, groundwater flow into the sea, particle settling rates, and burial rates on the seafloor (Cochran, 1982).

The scope of this article, however, will focus on what humans have added to the ocean in the nuclear era as a result of nuclear weapons, nuclear accidents, nuclear powered electric plants, reprocessing of nuclear fuels or burial of wastes, and, to a lesser extent, from nuclear-powered ships and submarines, as well as other applications in medicine and industry.

Of these anthropogenic sources, by far the largest was the release of a wide range of radionuclides as a consequence of the nuclear weapons testing programs that began in 1945 and peaked in the early 1960s. Globally, there have been at least 423 nuclear weapons tests between 1945 and 1980 (Hamilton et al., 1996), with about 90% of the yield coming from tests by the United States, United Kingdom, and former Soviet Union. Most of the large tests in the 1960s produced stratospheric fallout that was distributed worldwide. Three-quarters of that global fallout was deposited in the Northern Hemisphere due to the location of the major test sites, with higher deposition occurring in mid-latitudes because of the exchange between the stratosphere and troposphere in this latitudinal band.

The range of anthropogenic radionuclides released from weapons testing is quite large; these radionuclides are sometimes grouped into those of greatest radiological concern—cesium-137

(^{137}Cs), strontium-90 (^{90}Sr), the isotopes of plutonium (Pu) and americium (Am)—and those with lower health concern but wide use as ocean tracers—tritium, carbon-14 (^{14}C), technicium-99 (^{99}Tc), and iodine-129 (^{129}I) (Livingston and Povinec, 2000). Most of these radionuclides are highly soluble in seawater, so ocean currents and mixing processes determine their fate. However, isotopes of Pu and Am are more readily scavenged, or removed, from the water column via sinking of particles, a process that is especially intense near ocean margins and in high-productivity regions.

Recently, considerable attention has been paid to anthropogenic Cs, which is one of the more common fission products arising from nuclear weapons testing and reactor operations. Here, we focus on the status of oceanic sources of Cs both prior to, and resulting from, the Fukushima Dai-ichi nuclear power plant (NPP) meltdowns in spring 2011.

The largest source of ^{137}Cs is global fallout from nuclear weapons testing, with a cumulative delivery estimated to be around 950 PBq (Figure 1). Given the large surface area of the Pacific and maximum fallout in the Northern Hemisphere, the delivery of ^{137}Cs to the North Pacific alone is estimated to be 220 PBq (Aarkrog, 2003), which, with its 30.07 year half-life, would have been reduced by radioactive decay to 76 PBq in 2011. An additional source of fallout unique to the North Pacific was US nuclear testing at the Pacific Proving Grounds, with the largest tests conducted on the islands of Bikini and Eniwetok in the 1950s. Because these island tests were primarily surface-based, they produced significant local fallout, which, when decay is corrected to 2011, accounts for an additional 28 PBq ^{137}Cs in the North Pacific (Table 1).

The second largest source of anthropogenic radionuclides is related to the accidents at the nuclear power plants at Chernobyl in 1986 and Fukushima in 2011. The two events were quite different in their causes and consequences and, thus, in the quantities and range of radionuclides released to the ocean. Chernobyl was an explosive event that opened the reactor core and released gases, volatiles, and particulates during the explosion and the subsequent fires that lasted for about one week (Hohenemser et al., 1986; Pentreath, 1988). As a result, compared to Fukushima, it released more refractory elements, such as plutonium, ruthenium (Ru), and cerium (Ce) isotopes (Schwantes et al., 2012; Yoshida and Kanda, 2012).

By most estimates, Chernobyl is the largest accidental release of ^{137}Cs to the environment. Aarkrog (2003) estimates about 100 PBq released as local and tropospheric fallout, and other estimates range from 85 to 135 PBq (Davoine and Bocquet, 2007). Given the localized fallout over Europe, prevailing winds, and the many hundreds of kilometers between Chernobyl and the Baltic, North, Black, and Mediterranean Seas, the input of Chernobyl ^{137}Cs to the global ocean is estimated to be only 15% to 20% of the total released (Aarkrog, 2003), with negligible input to the Pacific. Thus, by 2011, total Chernobyl ^{137}Cs remaining in the environment would have been reduced to 56 PBq by decay, with only 8–11 PBq remaining in the ocean.

Less well resolved after two years of study is the total amount of ^{137}Cs and

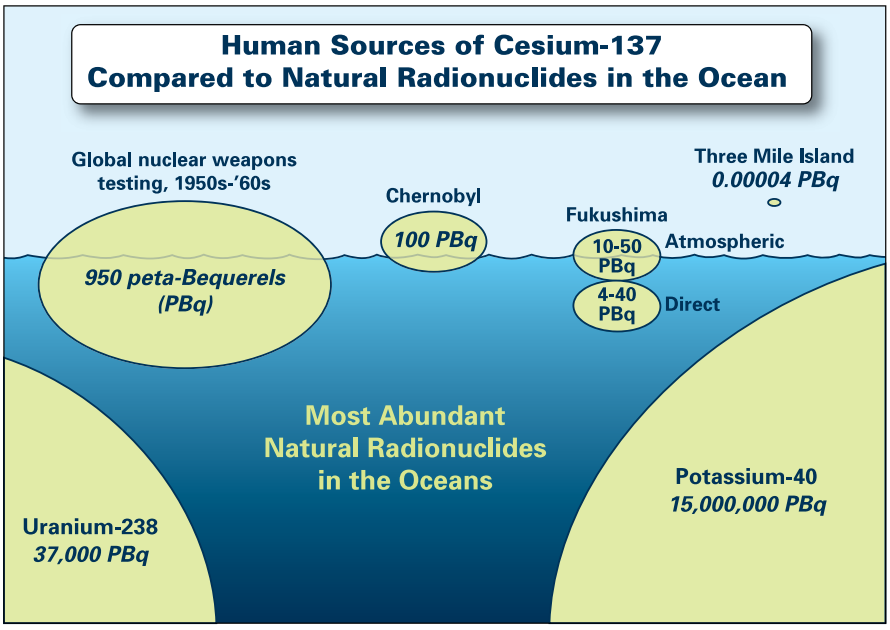

Figure 1. A comparison of the inventories of two common natural radionuclides, potassium-40 and uranium-238, with cesium-137 produced by various human sources. Inventories are calculated for the entire ocean for natural radionuclides, and for ^{137}Cs at time of delivery in units of PBq, or 10^{15} Becquerels (one Bq = one radioactive decay event per second). References in text and in Table 1.

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^{134}Cs released from Fukushima. There are two main sources of radionuclides from Fukushima. The first is the explosions and fires that led to the primary release of radioactive gases and volatiles to the atmosphere in the days immediately following the March 11 tsunami. The second source is the reactor cooling water along with direct discharge of radionuclides from the damaged buildings via surface run-off and groundwater transport from the site. These direct discharges peaked almost one month after the tsunami, in early April (Buesseler et al., 2011).

Given the prevailing winds off Japan, only 20% of the atmospheric fallout is estimated to have fallen on land, with the majority of the remainder thus deposited to the North Pacific (Morino et al., 2011). The magnitude of the Fukushima atmospheric source ranges from 10 to 15 PBq when based upon monitoring data collected in Japan and atmospheric dispersion modeling of radioactive fallout (Table 1; Chino et al., 2011; Morino et al., 2011). An estimate of 36 PBq comes from combining a larger global data set of aerosol monitoring stations and an inverse modeling approach that uses a global atmospheric transport model to deduce the source strength, though the full range by that method is rather large (23–50 PBq; Stohl et al., 2011).

The magnitude of direct ocean discharges from Fukushima is also difficult to constrain. Many of the estimates are based on the record of Cs discharge from Fukushima and limited early ocean measurements taken within 30 km of the site. These estimates range from 3.6 to 5.9 PBq (Kawamura et al., 2011; Estournel et al., 2012; Miyazawa et al., 2012; Tsumune et al., 2012). However, estimates of the source from analyses of Cs data in waters sampled in June 2011 further offshore are higher (11 to 18 PBq; Charette et al., 2013; Rypina et al., 2013).

The highest ocean discharge estimate is based on the evaluation of limited Cs time-series data in a 50 km area in the ocean around the NPP, resulting in extrapolation to a source term of 27 PBq (range 12 to 41 PBq; Bailly du Bois et al., 2012). So, as with the atmospheric delivery source term, the range in published estimates of the direct ocean discharge source is wide, 4 to 40 PBq, with most studies suggesting something more like 5 to 15 PBq (Table 1). This uncertainty may never be resolved, given limited ocean sampling in the weeks and months after the accident and minimal sampling further across the Pacific.

Certainly, as an ocean source, Fukushima exceeds Chernobyl in terms of its contribution of anthropogenic Cs isotopes to the sea because of its location along the coast, rather than as a result of its total release. Given decay, the only significant source of ^{134}Cs

Table 1. Summary of cesium-137 sources to the environment and ocean in PBq (10^{15} Bq)

Source	Total[a]	Total Ocean[b]	2011 Ocean[c]	2011 North Pacific[d]
Global fallout	950	600	190	76
Close-in fallout	180	180	56	28[e]
Total fallout				104
Chernobyl	100	18[f]	10	n/a
Sellafield	39	39[g]	30	n/a
La Hague	0.96	0.96[h]	0.75	n/a
Fukushima				
Atmospheric				10–15[i]
				23–50[j]
Direct Ocean				3.6–5.9[k]
				11–18[l]
				12–41[m]
Total Fukushima				4–90

a. Cumulative total for given source at time of reference. From Aarkrog (2003) unless otherwise noted
b. Total deposited on the ocean
c. Total ocean decay corrected to 2011
d. Fraction of ocean input between 0 and 90°N in Pacific Ocean
e. Aarkrog (2003) assumed 50% of Pacific Proving Ground fallout deposited north of equator. Close-in fallout from other sources considered negligible (at least for North Pacific)
f. 10–20% of Chernobyl fallout fell into ocean, primarily North, Baltic, Black, and Mediterranean Seas in 1986
g. Sellafield discharges directly to Irish Sea. Peak in 1974
h. La Hague discharges to English Channel. Peak in 1971
i. Chino et al. (2011); Morino et al. (2011)
j. Stohl et al. (2011)
k. Miyazawa et al. (2012); Tsumune et al. (2012); Kawamura et al. (2011); Estournel et al. (2012)
l. Charette et al. (2013); Rypina et al. (2013)
m. Bally du Bois et al. (2012)

($t_{½}$ = 2.06 yrs) to the ocean in 2011 must be from Fukushima, and this is used as a diagnostic tool to quantify the fraction of Fukushima Cs in any given sample. Likewise, given time since the cessation of nuclear weapons testing, the total inventory of ^{137}Cs in the North Pacific in 2011 is no longer much greater than recent Fukushima sources (Table 1).

> "THE FUKUSHIMA DISASTER PROVIDES A CRITICAL TOOL FOR THE OCEAN SCIENCE COMMUNITY TO STUDY FUNDAMENTAL PHYSICAL, CHEMICAL, AND BIOLOGICAL PROCESSES IN THE PACIFIC BASIN FOR AS LONG AS WE CAN DETECT TRACES OF THE SUITE OF LONG-LIVED RADIONUCLIDES THAT ORIGINATED THERE."

In addition to the weapons tests and NPP accidents, the third largest source of anthropogenic radionuclides to the ocean is the intentional discharge of wastes associated with the reprocessing of spent fuel to recover U and Pu (Table 1). The major reprocessing plants are at Sellafield (UK), Cap de la Hague (France), and Tokai (Japan), with Sellafield being substantially greater than all the others combined (^{137}Cs discharges peaked in 1971 at 5 PBq yr^{-1} with a cumulative input totaled 39 PBq; Aarkrog, 2003). Interestingly, the isotope ratios of many radionuclides differ between sites and over time (Pentreath, 1988). Also, Cap de La Hague and Sellafield remain major sources to the ocean of ^{99}Tc and ^{129}I, two very long-lived radionuclides (213,000 and 1.6 million years, respectively). Given the generally north-flowing currents near both locations, these tracers, along with Cs, have proven valuable in studying transport in the Arctic Ocean (Kershaw and Baxter, 1995; Smith et al., 1998).

Finally, there are several minor ocean sources of radionuclides, including Cs isotopes from radioactive waste disposal, power plant operations, and smaller-scale accidents associated with satellites and nuclear submarines, but these will not be reviewed here as they are comparably quite small and often localized relative to these other source terms (for a review, see Livingston and Povinec, 2000).

In summary, in 2011, nuclear weapons testing remains the largest source of ^{137}Cs into the global ocean and the North Pacific (104 PBq). However, Fukushima is of similar magnitude, and it was delivered almost entirely to the North Pacific (15 to 30 PBq using the most common estimates for ^{137}Cs plus an equal amount of ^{134}Cs). It is similar in scale to the intentional releases of reprocessing waste into the Irish Sea (39 PBq), and greater than the fraction of the Chernobyl releases that entered the ocean (10 PBq; Table 1). As an accidental source of anthropogenic Cs, Fukushima is an unprecedented event for the ocean.

CYCLING OF CESIUM IN THE OCEAN

The fate of Fukushima Cs after delivery, whether by atmospheric fallout or direct discharge, is controlled by the same processes and largely determined by its highly soluble nature (< 0.1% of Fukushima Cs was caught on 1 µ filters; Buesseler et al., 2012). As a soluble element, ocean currents and mixing processes dilute Cs activities and transport it into the ocean interior. Off Japan, the Kuroshio current is a particularly strong western boundary current, and the Fukushima plant is located in an area where the east-flowing Kuroshio and southwest flowing Oyashio mix (Figure 2). The region is also characterized by complicated nearshore currents, tidally driven flows, and mesoscale eddies. The complicated pathways of these currents can be tracked by Lagrangian floats and are captured in models (Rypina et al., 2013).

We know from tracking the weapons testing fallout that surface ^{137}Cs decreases over decadal time scales, not only due to its decay but also due to continued downward mixing in the ocean. The net result of these processes is surface water effective removal half-lives for Cs of 10 to 20 years, with an average of 12 ± 1 years determined for the North Pacific (Povinec et al., 2005).

On shorter time scales, the levels of ^{137}Cs in the ocean immediately off the NPPs were reduced by three orders of magnitude one month following peak ocean discharge in April 2011 (Buesseler et al., 2011). More than two years later, ^{137}Cs activities in the ocean adjacent to the NPP are being monitored and

remain in the 1,000 Bq m^{-3} range, dropping to tens of Bq m^{-3} in the first few to tens of kilometers offshore (TEPCO, 2013). However, the persistence of high ^{134}Cs and ^{137}Cs in the ocean offshore Fukushima is one of the lines of evidence of a small but continued source from the NPP (Buesseler et al., 2011; Kanda, 2013).

In the months following the accident, researchers reported a wide range of variability in Cs activity in the ocean off Japan. This variability was due to the changing source strength and variations in prevailing currents and eddies. For example, in June 2011, there was more than a three-orders-of-magnitude variability in surface ocean ^{134}Cs and ^{137}Cs activities in the waters 30 to 600 km off the coast of Fukushima (Figure 2). This variability can be explained by the association of the highest activity waters with a semipermanent nearshore eddy that retained the Fukushima Cs, and by the Kuroshio current acting as both a major transport conduit into the Pacific and a southward boundary to transport. Modeling of these data suggested that by June 2011 more than 85% to 90% of the fallout had already moved further east into the Pacific and that the local waters were dominated by the direct ocean discharge source at that time (Rypina et al., 2013).

While most of the attention focused on the higher activities closer to Japan, Aoyama et al. (2013) used ships of opportunity to track the progression of the Fukushima-derived ^{137}Cs and ^{134}Cs plume across the Pacific. Using 10 Bq m^{-3} as the frontal boundary, their data showed that the front had reached 180° longitude after one year.

On the basin scale, global ocean circulation models of the spread of Fukushima Cs are generally consistent (Behrens et al., 2012; Nakano and Povinec, 2012; Rossi et al., 2013), but they do differ in important ways. These differences include the timing that Fukushima-tagged surface waters will reach the US West Coast, which range from two to four years, and with an even wider range of predicted activities, < 2 Bq m^{-3} in Behrens et al. (2012) to 20 to 30 Bq m^{-3} in Rossi et al. (2013). The large difference in predicted ^{137}Cs activities off the US West Coast is in part due to the two-times higher source input used in Rossi et al. (22 PBq) vs. Behrens et al. (10 PBq), but that alone does not explain the order of magnitude difference in maximum surface activities, which must be due to the velocity fields and additional dispersion and mixing in Behrens et al., resulting in more dilution.

Both Behrens et al. (2012) and Rossi et al. (2013) models are eddy-resolving, and this is likely an important difference from predictions by Nakano and Povinec (2012), who use a coarser two-degree resolution model. All three studies predict that subsurface ^{137}Cs activities in intermediate and mode waters will rapidly become higher than surface waters, which after two to three years will result in a Fukushima Cs maximum at depth and penetration of the Fukushima signal of several hundred meters (Figure 3).

To determine which model predictions are more accurate would require more extensive vertical sampling across the Pacific than is currently available. Some information will be forthcoming

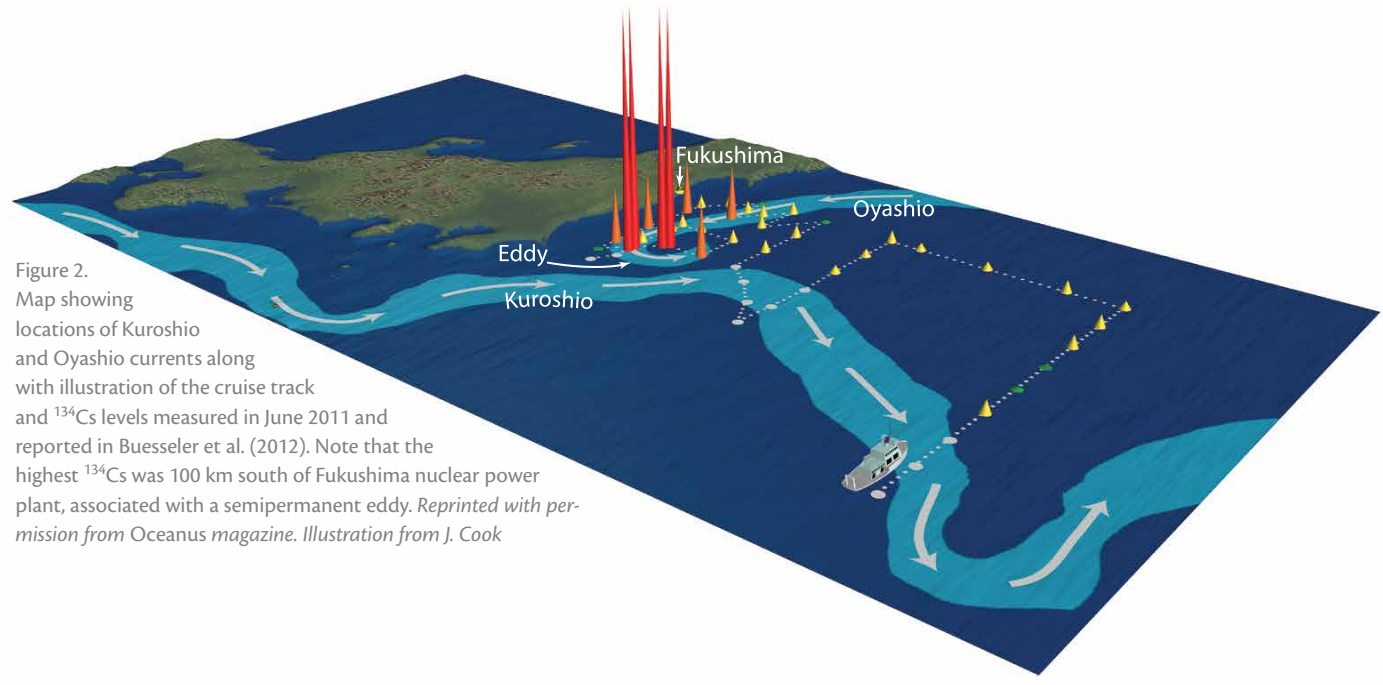

Figure 2. Map showing locations of Kuroshio and Oyashio currents along with illustration of the cruise track and ^{134}Cs levels measured in June 2011 and reported in Buesseler et al. (2012). Note that the highest ^{134}Cs was 100 km south of Fukushima nuclear power plant, associated with a semipermanent eddy. *Reprinted with permission from* Oceanus *magazine. Illustration from J. Cook*

from analyses underway in Japan and the United States, and monitoring of coastal activities along the United States can easily distinguish Fukushima ^{137}Cs and ^{134}Cs over background ^{137}Cs activities of only 1 to 2 Bq m^{-3}, especially if the higher predictions for > 10 Bq m^{-3} prove true.

In the long term, there is the prospect of monitoring Fukushima-tagged waters in the Pacific using $^{137}Cs/^{90}Sr$ and $^{137}Cs/^{129}I$ ratios, which differ in Fukushima waters from the earlier weapons testing fallout (Casacuberta et al., 2013; Hou et al., 2013; Tumey et al., 2013). Because most of the atmospheric deposition from Fukushima is thought to have occurred within a few 100 km of Japan, over time it will matter less in the models if the source was predominantly atmospheric (Cs and I) or direct discharge (Cs, I, and Sr), as both sources are proximal to Japan and differ in their timing by less than one month.

SEDIMENTATION PROCESSES

Although the focus on Cs in ocean sciences is most often on its use as a conservative, soluble tracer, there is still a small association of Cs with marine sediments and biota that results in its accumulation in the food web (see following section) and sedimentation of Cs to the seafloor (Honda et al., 2013; Kusakabe et al., 2013). The pathway for Cs to the sediments is via sinking particles, which can be biogenic and/or lithogenic. Honda et al. (2013) report on two time series sediment traps in the western North Pacific that collected sinking particles before and after the Fukushima releases (Figure 4). They measured the first Fukushima ^{134}Cs arriving in late March 2011 in a 500 m trap at the K2 site, some 1,100 km from the Fukushima site, south of Kamchatka in the Subarctic Gyre. Because of the timing and distance, the only likely source was atmospheric fallout from the early Fukushima explosions. Similar arrival times, but 5 to 10 times lower ^{137}Cs and ^{134}Cs concentrations, were found at a time-series trap site S1 800 km from Fukushima and south of the Kuroshio current in the subtropical gyre.

From such data, Honda et al. (2013) make several important conclusions regarding vertical transport of Cs associated with sinking particles. First, using the time between deposition and arrival at the 500 m trap, they derive sinking velocities of 20 to 70 meters per day from the surface ocean. Similarly, by comparing the arrival time of Cs measured at 500 m compared to the deeper 4,800 m traps, they derive a deep ocean sinking rate >180 meters per day. These sinking velocities were comparable between sites even though the chemical composition at K2 (46% opal and 32% calcium carbonate) was different than at S1 (9% opal and 72% calcium carbonate). These sinking rates are comparable to prior studies of particle fluxes in the ocean (Fowler and Knauer, 1986) and suggest that the seafloor off Japan would have seen the Fukushima radionuclides within days following the arrival of either atmospheric fallout or contaminated waters.

Second, using measured Cs data in surface waters and some assumptions about its vertical distribution, Honda et al. (2013) use the Cs-flux/Cs-inventory ratio to calculate a residence time for Cs that ranges from 70 to 310 years. They suggest that this might be an underestimate because the removal rates may be higher in the early deposition stages. It is important to remember that these long residence times support the widespread use of Cs as a soluble conservative tracer over the next several decades, similar to conclusions drawn from Cs trap studies post-Chernobyl in the Pacific Ocean (Kusakabe et al., 1988) and the Mediterranean (Fowler et al., 1987) and Black Seas (Buesseler et al., 1987).

Finally, Honda et al. (2013) do not find a significant correlation between Cs in sinking particles and the fraction of clay minerals, and these data thus leave open the question of whether lithogenic or biogenic sediment fractions transport Cs to the seafloor. Certainly, there are likely associations with both, as marine

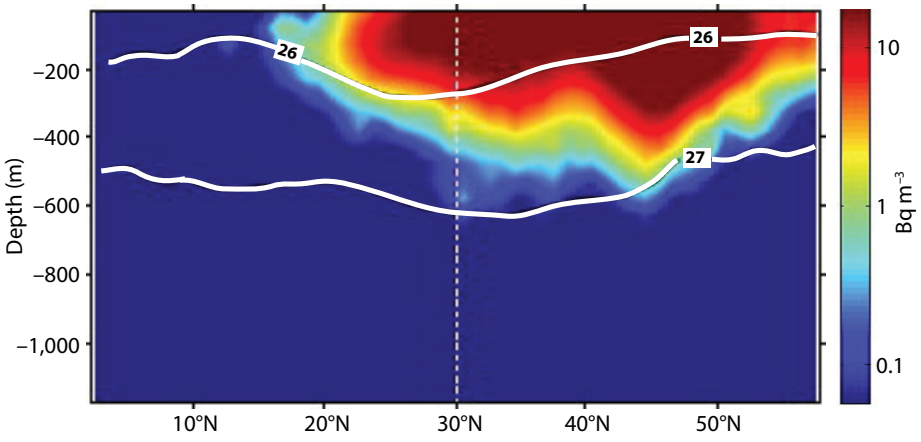

Figure 3. Numerically simulated north/south vertical section predicting ^{137}Cs at 140°W in April 2014. Isopycnals contoured in white represent the mode water layer. *Adapted from Rossi et al. (2013)*

plankton take up Cs, and aluminosilicates adsorb it, and all of these materials are components of sinking particles in the ocean.

Once deposited to the seafloor, the fate of Fukushima Cs is determined by a combination of deeper mixing by bioturbation—the biological mixing at the seafloor via organisms—and the re-release of Cs either through remobilization back to the solution phase or, more likely, transport in turbidity layers along the bottom out to the ocean interior, where they can be deposited more than 100 km from their original sources (Oguri et al., 2013).

Thus far, there are fewer studies of the distributions of Fukushima Cs in sediments than in waters off Japan. This is in part because it is more time-consuming and difficult to collect sediment than water samples, given the need to ensure that the uppermost layers of the core top are not lost during sampling. Thus, early data reported from grab samples are fine for determining order-of-magnitude Cs concentrations for monitoring, but not as useful for looking at spatial and temporal changes. Sediment types vary widely as does Cs distribution on the seafloor. Even using carefully collected samples from a multi-corer, Kusakabe et al. (2013) show how even at a single sampling location, replicate cores can differ by a factor of three in the amount of Fukushima Cs they contain. They urge caution in interpreting sediment variability, and saw, for example, no simple trends in Cs content versus distance from the Fukushima NPP or obvious decrease with time (Figure 5). Their data suggest that < 1% to 2% of the total Fukushima Cs released ended up in marine sediments off Japan.

It is illustrative to look at a mass balance of Cs sources and sinks in the coastal ocean off Japan one to two years after the peak releases. In late 2012, the largest Cs inventory is associated with seafloor sediments (about 100 TBq; TBq = 10^{12} Bq) because concentrations and, hence, inventories in the water are now much lower (15 TBq) (Figure 6).

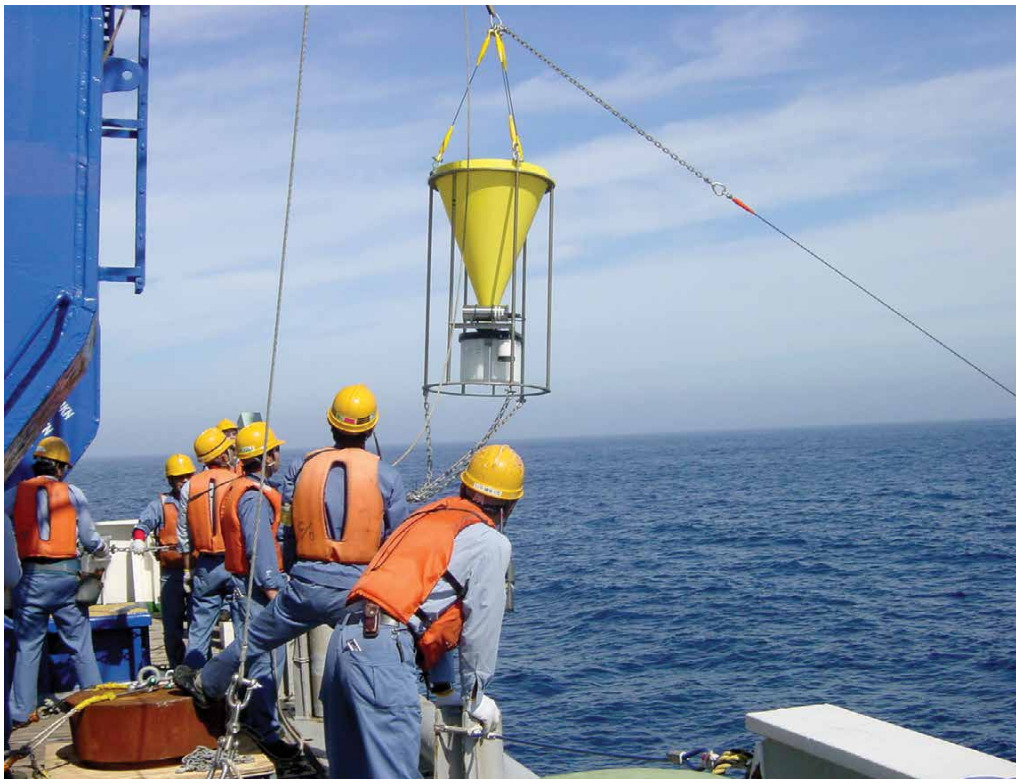

Figure 4. Time-series sediment trap being deployed off Japan on JAMSTEC ship R/V *Mirai*. *Photo credit: M. Honda*

Two long-term sources of Fukushima Cs are leakage from the NPP site and river-borne Cs. One of the best estimates of continued leakage uses the exchange rate of harbor water with seawater at the NPP and the time history of Cs in these waters to estimate about 0.2–0.3 TBq per month of Cs entering the ocean at the end of 2012 (Kanda, 2013). Although significant, this rate is, of course, more than four orders of magnitude smaller than the direct discharge of Cs in 2011 (Table 1), but still of great concern to ongoing cleanup efforts at the site (and to contamination of fish—see below).

Some of the continued Cs source is likely to be previously contaminated sediments and groundwater at the NPPs. In 2011, it was already noted that continued elevation of Cs isotopes in the ocean suggested a small but continued NPP source (Buesseler et al., 2011). News in July 2013 of increases in radionuclides, particularly ^{90}Sr and tritium, in groundwater at the NPP site well above regulatory levels suggests that input to the ocean may be changing. More work is needed to assess these sources.

The second long-term source of ^{137}Cs to the ocean is river-borne waters and sediments (Nagao et al., 2013, Chartin et al., 2013). Nagao et al. (2013) found that up to half of the annual Cs flux from the Japanese rivers is from heavy rain events. Because Cs is largely insoluble in freshwater, its transport and delivery via rivers is much more tied to the

delivery of high sediment loads associated with heavy rains and flood events. The Cs being carried directly to the ocean was suggested to be associated with Cs bound by clay minerals, some of which may be released upon entering the saltwater, where Cs is more soluble. A riverine ^{137}Cs input of about 1 TBq per month in late 2012 can be estimated (Figure 6; J. Kanda, Tokyo University of Marine Science and Technology, *pers. comm.*, 2013), with more than half associated with the river-borne particles.

Not yet discussed in this budget is potential release of Cs from the seafloor. Cs can be released back into bottom waters either with resuspension of the bottom sediments or remobilization in pore water. In several earlier studies, the effective sedimentary half-life of Cs derived from the Sellafield discharges has been shown to be about 10 years in the Irish Sea, significantly shorter than its 30 year half-life (e.g., Mitchell et al., 1999). Using this sedimentary half-life of Cs, there would be a loss of half of the total sediment inventory in 10 years or, using the 100 TBq inventory, about 0.5 TBq per month.

There are arguably large errors in all of these continued Cs source and sink terms, but it is evident that the inputs from land via leakage at the NPP and rivers are on of the same magnitude as the amount of Cs that might leave the system via sediments and transport offshore. Thus, we should not expect to see a clear and rapid decrease in Cs in coastal sediments. There is a great need for more careful study of the continued Cs sources, sinks, and transport rates to make a reliable long-term prediction of when the water and sediments will return to pre-Fukushima levels (see Box 1).

BIOLOGICAL UPTAKE

Many factors determine how much radioactivity is assimilated by marine biota, including the species-specific uptake and excretion rates of different organisms. The uptake of radionuclides in algae and fish is most often described by a concentration factor, CF, which is the ratio, in this case, of Cs in the organism (Bq kg^{-1} wet weight) divided by the concentration in the water (Bq L^{-1}). CFs for Cs are not high, with values in the range of 50–100 (IAEA, 2004) and only modest increases as one moves up the food chain (Doi et al., 2012). Post-Fukushima, for example, a CF of six to eight was found for zooplankton and small fish in waters within 600 km off Japan (converted using dry to wet ratio of 0.15–0.20; Buesseler et al., 2012). However, there was wide variability in CFs, which may be influenced to some extent by the size of the organism for the plankton and concentrations in the food supply for the larger animals.

Also significant for Cs distributions in fish is that the biological loss rate is quite fast, on the order of one to a few percent per day, leading to a presumed steady-state biological half-life of about 50 days for fish (MAFF, 2012). Thus, a fish living and feeding in a contaminated area that moves to waters with no further source of contamination should show a rapid decrease in Cs. This is exactly what Madigan et al. (2012) report for Pacific

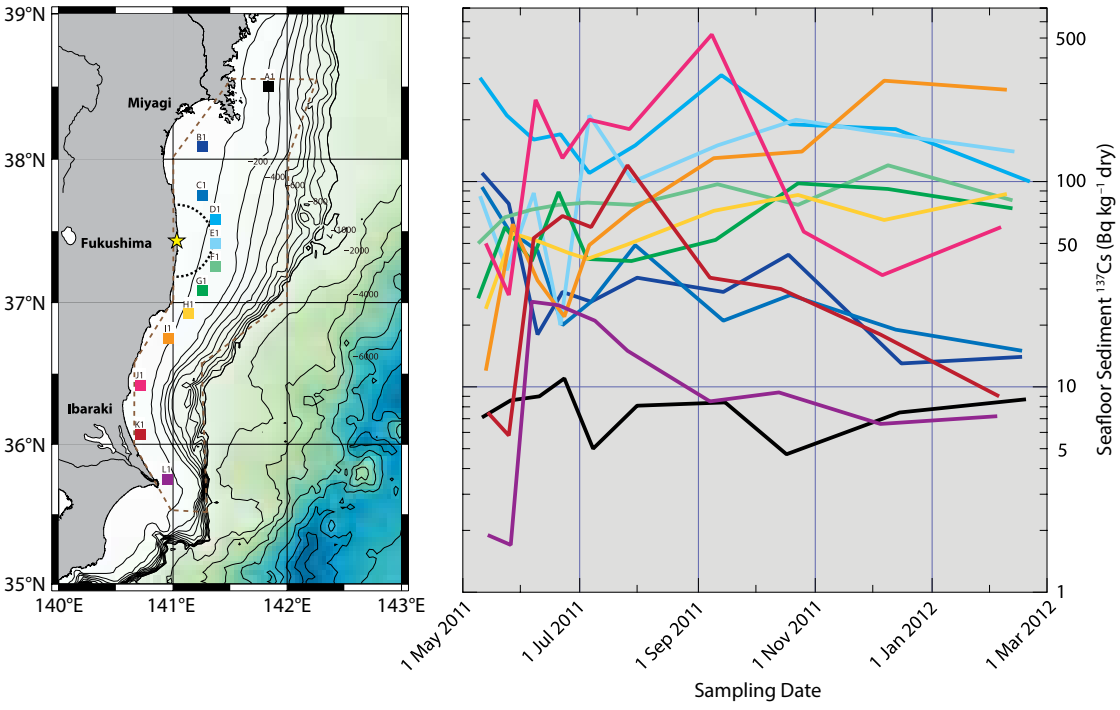

Figure 5. Cesium-137 in top 3 cm of seafloor sediments off Japan. Core locations and colors on map correspond to time series of ^{137}Cs on right. *Adapted from Kusakabe et al. (2013)*

bluefin tuna caught in August 2011 off San Diego that had been feeding off Japan in the spring. Fukushima-derived ^{134}Cs was measured in these fish, but at levels 15 to 30 times lower than what would have existed off Japan. In fact, this decrease in ^{134}Cs was proposed as a new "tag" to monitor the speed of fish migration across the Pacific.

It was somewhat surprising, then, that after re-analysis of data from Japan's Ministry of Fisheries (MAFF, 2012), the levels of total Cs (^{137}Cs and ^{134}Cs) are found to remain elevated one year after the accident in many fish collected in the coastal waters off Japan (Buesseler, 2012). In this first year, the demersal, or bottom-dwelling, fish off Fukushima were particularly elevated, with over 40% exceeding the current Japanese regulatory limit of 100 Bq kg^{-1} (wet weight)

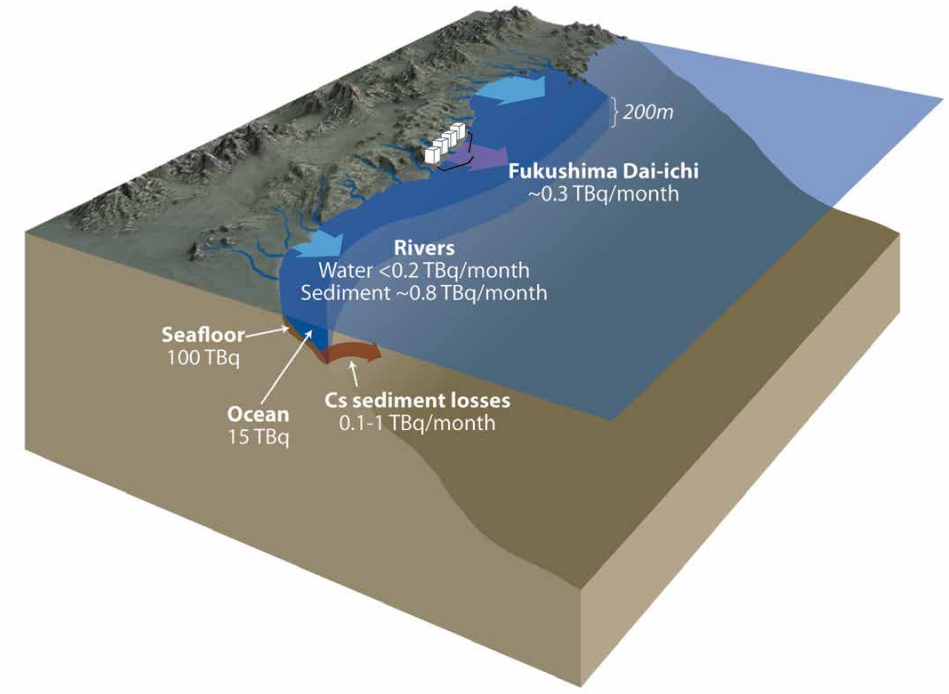

Figure 6. Estimates of ^{137}Cs inventories in seafloor sediments and ocean waters out to 200 m off the coast of Fukushima for late 2012. These inventories are compared to fluxes of ^{137}Cs from rivers and the Fukushima Dai-ichi nuclear power plant site, and estimates for Cs sediment losses via remobilization. Units for inventories are TBq and for fluxes TBq per month (one TBq = 10^{12} Bq). *Adapted from a presentation given by J. Kanda, November 2012, at the University of Tokyo, http://www.whoi.edu/fileserver.do?id=138570&pt=2&p=141589). Illustration by J. Cook*

BOX 1. THE FUTURE OF MARINE RADIOCHEMISTRY

Research, Training, and Communication Challenges

In the aftermath of Fukushima—after years of relative complacency—the public and policymakers have renewed concerns about radioactive contamination. In addition, radioactive wastes have piled up without enough permanent safe places to store them. Nuclear-fueled ships and submarines ply our ocean. Some are now being decommissioned because they are beyond their useful ship lives. Worldwide, the number of nuclear power plants (> 400) is expanding in many countries, while older nuclear power plants are being shut down. There are concerns about the spread of nuclear weapons and non-nuclear "dirty" bombs. Yet, at the same time, in the United States and other countries, nuclear scientists and radiochemists who studied fallout from the 1960s and after Chernobyl are retiring, creating a workforce gap that needs to be filled. In addition, at least in the United States, there are fewer academic research studies being supported. This means far less training of graduate students and postdocs in the demanding analyses of environmental radionuclides, which yield important data such as those in this paper.

One response to these concerns has been the launch of a new Center for Marine and Environmental Radioactivity (CMER) at the Woods Hole Oceanographic Institution. The mission of CMER is to increase scientific and public understanding of the sources, fates, and consequences of natural and human-made radioactive elements in the environment, in particular the ocean. This endeavour will be accomplished by providing research and education opportunities in environmental radioactivity and, in partnership with other institutions worldwide, by supporting and sustaining a critical mass of scientific capability to propel breakthroughs and generate valuable knowledge that can be used to inform the public and policymakers about the risks and uses of ionizing radiation in the environment.

CMER
Center for Marine & Environmental Radioactivity

For more information see
http://www.whoi.edu/CMER

in seafood, and no statistical decrease in Cs was seen. This figure suggested to us a continuing source of Cs from the NPP and/or marine sediments. When we extend that analysis to the end of 2012 (Figure 7), there is a slow decrease in Cs levels in these bottom fish off Fukushima, equating to an ecological half-life of 330 days, much slower than predicted initially.

Regarding the slow decline in Cs in fish, Tateda et al. (2013) question the validity of using steady-state concentration factors and, in fact, present a dynamic biological compartment model that can fit most of the longer time course of Cs in algae and fish. Their model captures the transfer of Cs through the food chain, so it includes a delay in Cs increase in fish and continued transfer to fish through feeding even after Cs concentrations have decreased in seawater. Most of the fish fit their model and show a decrease with an ecological half-life of two to nine months, depending on specific feeding habits and, thus, Cs sources in the food chain. However, consistent with the persistence of Cs in bottom fish, Tateda et al. (2013) conclude that there must be an additional source of Cs associated with the bottom sediment or near-bottom suspended matter, as the Cs in these benthic fish did not decrease as fast as predicted by their model.

Taking samples even closer to the reactor has resulted in some of the highest values to date of over 25,000 Bq kg^{-1} in greenling (*Hexagrammos otakii*) close to shore in late 2012 and 740,000 Bq kg^{-1} in greenling caught inside the NPP harbor in early 2013. These headline stories only add to Japanese anxieties over the prospects of re-opening fisheries off Northeast Japan. In hindsight, lowering the legal limit in Japan for total ^{137}Cs and ^{134}Cs in seafood from 500 to 100 Bq kg^{-1} wet weight in April 2012 may have also added to the anxiety and mistrust in Japan, as consumers don't know why the change was needed. In fact, this limit is the most restrictive level of any country, with, for example, levels of up to 1,200 Bq kg^{-1} allowed in the United States (US FDA, 2005). In the end, the coastal fisheries off Fukushima remain closed, which is a great economic (billions of dollars) and cultural loss for Japan.

In considering potential doses from consuming fish, Fisher et al. (2013) remind us that the naturally occurring alpha-emitter polonium-210 (^{210}Po) is a greater source of radiation to consumers than Cs. For example, in the United States, which has lower ^{137}Cs levels in fish and lower seafood consumption rates than Japan, the dose to consumers from Cs in fish would be 0.9 µSv (Sv = Sievert; assuming consumption of 24.1 kg yr^{-1} and 2.5 Bq total Cs kg^{-1} wet weight). This is 600 times less than the dose from ^{210}Po in those same fish and five times less than obtained from one dental x-ray (5 µSv).

In Japan, seafood consumption rates are higher (57 kg yr^{-1}), but even using the legal limit of 100 Bq kg^{-1} Cs in fish results in a dose of 75 µSv or more than 10 times lower than the international dose limit of 1 mSv yr^{-1} set for members of the public. At a Cs level of 100 Bq kg^{-1}, the dose from ^{210}Po is almost 20 times greater. As such, none of these potential doses from Cs would be expected to result in observable increases in cancer

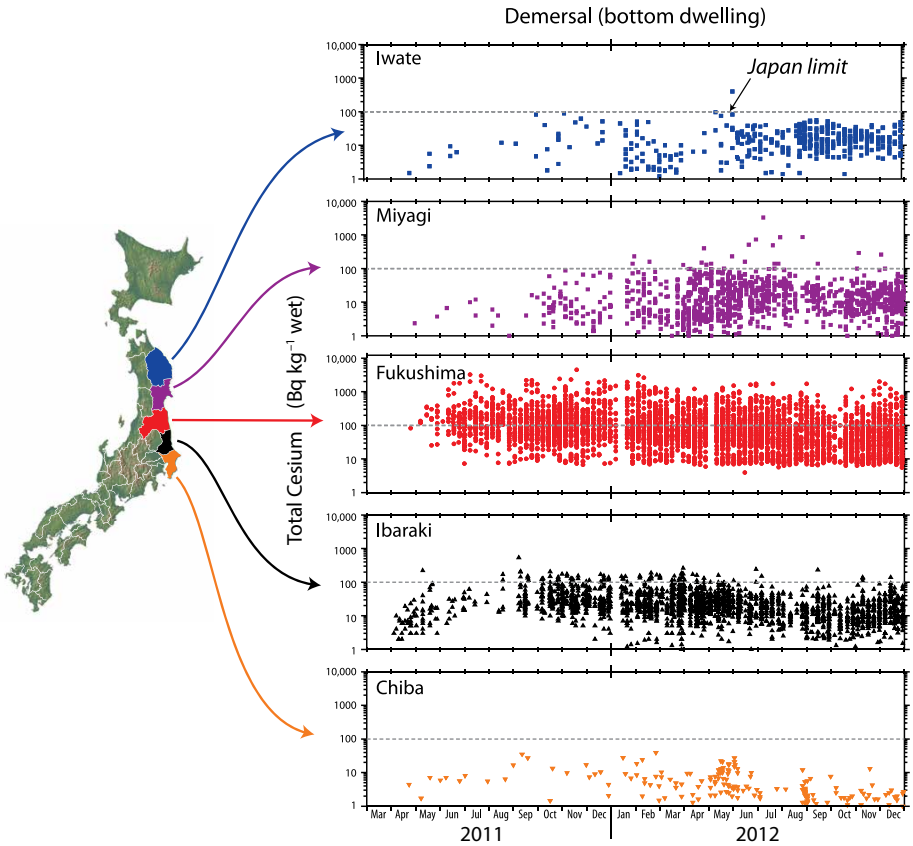

Figure 7. Time series of total ^{134}Cs + ^{137}Cs in demersal fish in Bq per kg wet weight. Original data are from the Japanese Ministry of Agriculture, Forestry and Fisheries. *Adapted from Buesseler (2012) and extended in time to end of 2012*

rates in the populations of Japan or the United States (Fisher et al., 2013).

Clearly, it is important to keep monitoring Cs in fish off Japan, especially in bottom-feeding fish, but also in freshwater fish (which are higher in radioactive Cs due to lower stable Cs in fresh waters). However, measuring fish is not enough to help with management decisions and planning hampered by lack of knowledge about the nature and amount of continuing sources from the NPP, rivers, and bottom sediments; how they might change in the future; and the residence time for Cs in the seafloor sediments off Japan. In addition, recent reports of elevated $^{90}Sr/^{137}Cs$ in groundwater at the NPP suggest that multiple isotopes and their sources and sinks need to be studied to understand the fate of these radionuclides in the ocean and uptake by biota to support targeted decisions about whether or not to re-open fisheries. Strontium is a bone-seeking isotope that has a longer biological half-life in fish; however, unless consuming smaller fish whole, we don't usually consume fish bones. In any case, multiple isotopes and their sources and sinks need to be studied to understand the fate of these radionuclides in the ocean and uptake by biota in order to support targeted decisions about whether or not to reopen fisheries. Such decisions need to come with continued public education about the health benefits of eating seafood that outweigh the low risk from radionuclide doses, as these studies and other data on naturally occurring radionuclides have already shown.

SUMMARY

The fact that we live in a radioactive world and that the world ocean already contains a wide range of natural and anthropogenic radionuclides in no way diminishes the scale and nature of events at Fukushima. For the ocean, the continued, unintentional releases from the site of the damaged nuclear power plant are unprecedented and will likely resonate in the local ecology and both national and international policy for decades to come. The Fukushima disaster provides a critical tool for the ocean science community to study fundamental physical, chemical, and biological processes in the Pacific Basin for as long as we can detect traces of the suite of long-lived radionuclides that originated there. It should serve as a call for ocean scientists to turn their attention to all aspects of this event, which continues to unfold and to present a plethora of new challenges, opportunities, and questions.

> "THE FACT THAT WE LIVE IN A RADIOACTIVE WORLD AND THAT THE WORLD OCEAN ALREADY CONTAINS A WIDE RANGE OF NATURAL AND ANTHROPOGENIC RADIONUCLIDES IN NO WAY DIMINISHES THE SCALE AND NATURE OF EVENTS AT FUKUSHIMA."

ACKNOWLEDGEMENTS

I am grateful to the many scientists in Japan who have freely shared results and samples, supported our own work and assisted in field collaborations, attended and organized several conferences, written and co-authored manuscripts, and encouraged international participation in these studies in general. Editorial assistance from K. Kostel on this manuscript is also greatly appreciated. Reviews from N. Fisher and S. Fowler are also appreciated and helped improve the final manuscript. Support for our Fukushima studies at the Woods Hole Oceanographic Institution (WHOI) has come primarily from the Gordon and Betty Moore Foundation, with additional support from the Deerbrook Foundation, US National Science Foundation, and, for outreach activities, the Japan Center for Global Partnership and the Morss Colloquia Endowed Fund at WHOI.

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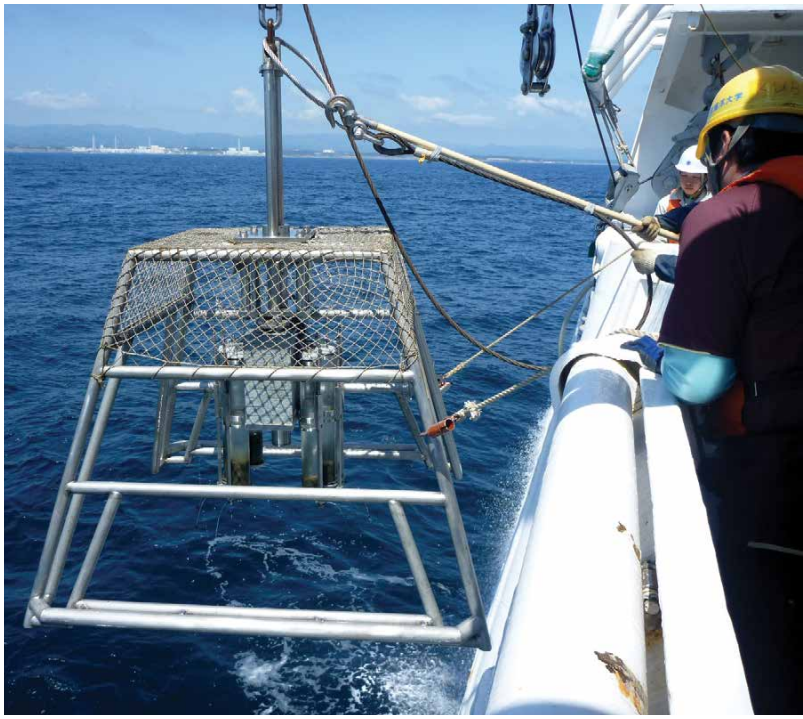

Plankton net (left) and seafloor corer (right) near Fukushima Dai-ichi in May 2013 on R/V *Umitaka Maru*. *Photos by K. Buesseler*

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