

# ABCs of Radioactivity

## A Long and Winding Road to Achieve Stability

To the average layperson, “radioactivity” is a harsh and scary word. But the fact is that radioisotopes, both natural and artificial, are all around us. And for marine scientists in particular, they are important tools. Oceanographer Claudia Benitez-Nelson reviewed some of the fundamentals at the start of the Fukushima and the Ocean conference in Tokyo in November 2012.

To begin at the beginning, radioactivity is a spontaneous emission of radiation resulting from changes in the nucleus of a chemical element, said Benitez-Nelson, who earned her Ph.D. from the MIT/WHOI Joint Program in 1999 and now directs the marine science program at the University of South Carolina. “We radiochemists have a whole suite of elements that we consider radioactive. They’re simply unstable. In the process of getting rid of that instability, they release energy to the surrounding environment in the form of radiation.”

That radiation comes in two broad types. The non-ionizing type, which includes visible light and microwaves, lacks enough oomph to create charged ions, and thereby to alter the structure of an atom. It poses little threat to our health. Ionizing radiation, however, can actually change the atomic structure of living tissue—killing cells or making them cancerous. That’s why we try to avoid direct exposure to medical X-rays and the sun’s ultraviolet rays.

All radioisotopes—also called radionuclides—lose excess energy by emitting ionizing particles such as neutrons, protons, electrons, or photons. In the process, these so-called parent radioisotopes transform, or decay, into daughter isotopes containing different numbers of protons and neutrons. Daughters with the same number of protons are isotopes of the parent element; daughters with a different number of protons are actually different elements, with different chemical properties.

Each change along the way, Benitez-Nelson noted, follows a unique timetable, or half-life. The half-life of an isotope is the time it takes for one-half of the atoms in a given sample to decay. This daughter isotope can decay into another radioisotope, or the daughter isotope, that will continue the radioactive decay chain or decay into a stable element that ends the chain.

For example, one of the most common naturally occurring radioisotopes, uranium-238, has 92 protons and 146 neutrons. It decays into thorium-234 (90 protons, 144 neutrons), which decays into protactinium-234 (91 protons, 143 neutrons), and so on. The half-lives for each of these radioisotopes, respectively, are 4.468 billion years, 24 days, and 1.2 minutes—and each of these elements chemically reacts in a different manner (see next page).

“Because that half-life differs from one radioisotope to the next, we can use it as a simple clock to study a host of ocean processes that take place across different timescales, from days to years to millennia,” Benitez-Nelson said. So-called radioactive “tracers” are present in the ocean for eons—some formed

by the interaction of incoming cosmic rays from space with atmospheric gases, and still others introduced by human activities. These tracers help scientists unravel how fast ocean waters mix, how quickly groundwater from land enters the ocean, and how rapidly carbon and other elements are cycled through air, sea, seafloor, and continents.

Several common naturally occurring radioisotopes—uranium, thorium, and potassium—are always present in seawater.

In fact, noted Benitez-Nelson, the amounts of these isotopes present in the ocean are thousands of times higher than those of even the largest human sources of radioactivity.

Ken Buesseler, a marine chemist at Woods Hole Oceanographic Institution, put it this way: “We live in a sea of radioactivity, but this does not present a problem to us or to sea life because the radioactive elements found in our oceans are at such minute concentrations.” As Buesseler added, “The danger is in the dose.” (See Page 20.)

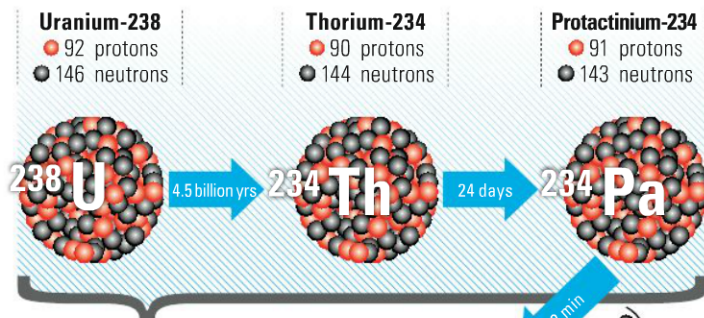
—David Pacchioli

“We live in a sea of radioactivity. The danger is in the dose.”

—Ken Buesseler



# Radioactive Decay Chains



## Atoms

Atoms are made of protons, neutrons and electrons. The number of protons determines what element it is. Atoms have the same number of protons. Atoms with different numbers of neutrons are called isotopes.

Radioactive elements, called radioisotopes or radionuclides, are unstable. They spontaneously "decay," releasing particles and/or energy. In the process, "parent" isotopes become "daughter" isotopes. These may become parent isotopes to their own daughter isotopes.

## Half-lives

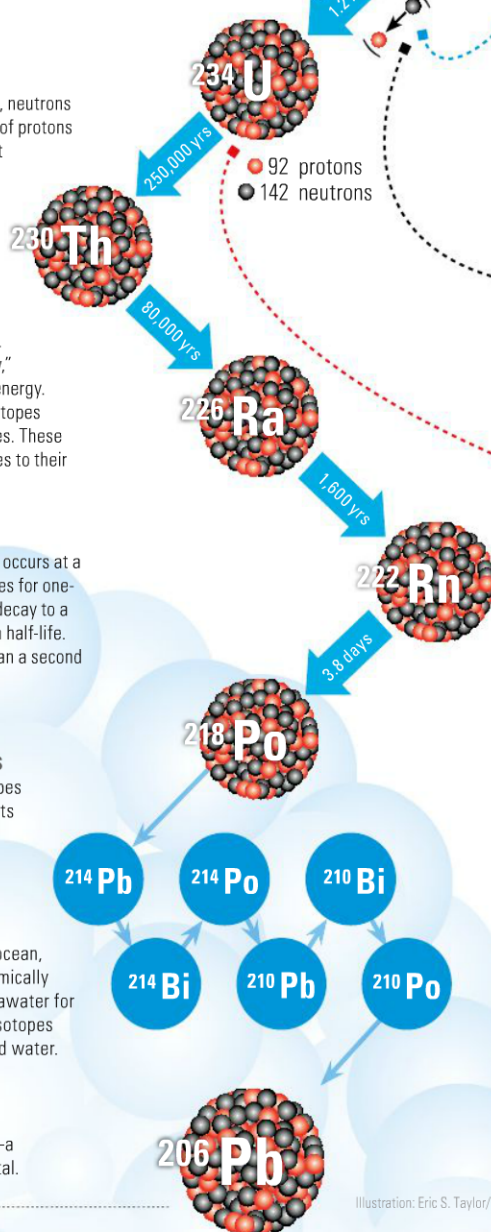
Each decay along the chain occurs at a unique rate. The time it takes for one-half of a parent isotope to decay to a daughter isotope is called a half-life. Half-lives vary from less than a second to billions of years.

## Elemental and chemical changes

Parent and daughter isotopes are often different elements with different physical and chemical properties. Thorium-230 is a metal, but radon-226 is a gas, for example. Th-234 adheres readily to particles in the ocean, while Pa-234 is not as chemically "sticky" and remains in seawater for centuries. All these radioisotopes occur naturally in rocks and water.

## End of the chain

occurs when polonium-210 decays into lead (Pb-206)—a stable, non-radioactive metal.



## Different types of radioactive decay

Radioisotopes can emit three kinds of ionizing radiation from their nuclei:

**Beta particles** are released when a neutron in nuclei turns into a proton, or vice versa. They are charged electrons or positrons that have high speed and energy and can travel far into the body.

**Gamma rays** are emitted during beta decay. They are pure energy on the electromagnetic spectrum that includes light and X-rays. They have no mass or charge but have high energy and can travel much farther than  $\alpha$  or  $\beta$  particles.

**Alpha particles** have two protons and two neutrons and a +2 charge. They are 2,000 times larger than  $\beta$  particles and travel relatively slowly and lose energy fast in air. They can't penetrate clothing, skin, or even paper, but they can be ingested or inhaled.

## Radiation and health

When ionizing radiation strikes tissue, cells, and DNA, it can break chemical bonds and otherwise damage cellular machinery. Depending on a variety of factors, health impacts can occur immediately, such as burns, or over longer terms, such as cancer. All three types of radiation can cause damage, but the larger particles, if inhaled or ingested, can cause the most damage to living tissue. (See Page 20.)



Illustration: Eric S. Taylor/WHOI



# How Is Fukushima's Fallout Affecting Marine Life



The Fukushima nuclear disaster delivered an unprecedented amount of radioactivity into the sea over a relatively brief time. How did that pulse of cesium and other radioisotopes make its way through the marine food chain? Scott Fowler, who helped pioneer marine radioecology for more than 30 years at the International Atomic Energy Agency's Marine Environment Laboratories, offered a primer on the subject at the Fukushima and the Ocean Conference in Tokyo in November 2012.

The food chain starts with marine phytoplankton—microscopic plants that account for as much photosynthesis as plants on land. These organisms take up radioactive contaminants from the seawater that surrounds them. As the phytoplankton are eaten by larger zooplankton, small fish, and larger animals up the food chain, some of the contaminants end up in fecal pellets or other detrital particles that settle to the seafloor. These particles accumulate in sediments, and some radioisotopes contained within them may be remobilized back into the overlying waters through microbial and chemical processes.

*By David Patcholi*

Photos by Ken Kostel/WHOI



How much radioactivity gets into marine life depends on a host of factors: How long the organisms are exposed to radioactivity is certainly important, but so too are the sizes and species of the organisms, the radioisotopes involved, the temperature and salinity of the water, how much oxygen is in it, and many other factors such as the life stage of the organisms.

In all this, Fowler said, it's important to remember the omnipresence of natural background radiation. Polonium-210 and potassium-40 are naturally occurring radioisotopes in the ocean, for example. Potassium-40 is the most abundant radioisotope in the ocean, but polonium-210 accumulates more readily in marine organisms.

"Polonium is responsible for the majority of the radiation dose that fish and other marine organisms receive," he said.

In an experiment in the early 1980s, Fowler demonstrated vast differences in how much plutonium was absorbed from seawater by marine life across a spectrum of taxonomic groups. Phytoplankton accumulated roughly 10 times as much plutonium as microzooplankton, which took up 100 times more than clams. Octopi and crabs took up about half as much plutonium as clams, but about 100 times more than bottom-dwelling fish.

Another cross-species comparison showed that organisms took up different amounts of radioactivity depending on which particular radioisotopes were out there, he said.

Radioisotopes are also transferred to marine organisms from contaminated sediments—once again in ways that display a complex range of factors, Fowler noted. In one experiment measuring uptake of americium, worms exposed to contaminated sediments took up significantly more of the radioisotope than clams did. But both worms and clams took up much more of the radioisotopes from Pacific sediments, which contain relatively high amounts of silica minerals, than they did from Atlantic sediments, which contain more carbon minerals.

Food is another pathway into marine organisms and "may be in some cases the most important factor in uptake," Fowler said. Consumed radioisotopes are assimilated internally through the gut, potentially a far more efficient route than if they are absorbed externally from the environment. Marine invertebrates, such as bottom-dwelling starfish and sea urchins, are particularly proficient at absorbing a wide range of ingested radioisotopes, he said, but fortunately, they lose that incorporated radioactivity over time, via excretion.

## From plankton to tuna

Fowler's longtime colleague, Nicholas Fisher, zeroed in on the isotopes that have had the most impact from Fukushima. Fisher, a marine biogeochemist at Stony Brook University, has spent 35 years studying the fate of metals and radioisotopes in marine organisms, including radioisotopes associated with nuclear waste. He and members of his lab participated in the research cruise led by Woods Hole Oceanographic Institution marine geochemist Ken Buesseler off the coast of Japan in June 2011.

Analyzing plankton and fish sampled on the cruise, they consistently found cesium-134 and cesium-137. Not surprisingly, they found no iodine-131, the isotope which along with cesium had been released in highest quantity from the

damaged Dai-ichi nuclear power plant. Iodine-131, with its half-life of a mere eight days, was undetectable after a couple of months, Fisher explained.

Cesium, of course, is a different story. The ocean and its denizens continue to bear traces of cesium-137 that date from the atmospheric weapons testing during the Cold War era of the 1960s. Cesium-134, while much shorter-lived, will persist for a number of years.

The chemical properties of radioactive cesium are similar to those of non-radioactive cesium and naturally occurring potassium and sodium, which are abundant in seawater. So all these end up in the same tissues, particularly muscle, of fish and other marine organisms. But potassium and sodium are much less abundant in fresh water, so cesium uptake is much higher in freshwater organisms than in sea life.

Fish also excrete cesium fairly efficiently, losing a few percent per day. So if fish are no longer exposed to new contamination sources, the levels in their tissue should decrease fairly quickly.

Of particular concern for top-level consumers is the potential that these radioisotopes will be concentrated as they make their way up the food chain—what ecologists call biomagnification. Fortunately, cesium shows only modest biomagnification in marine food chains—much less than mercury, a toxic metal, or many other harmful organic compounds such as the insecticide DDT and polychlorinated biphenyls (PCBs), Fisher said.

On the 2011 cruise, he and his team measured cesium in everything they sampled. "These were primarily zooplankton and some fish," he reported. As expected, concentrations were higher in organisms sampled closer to shore. Radioactive silver ( $^{110m}\text{Ag}$ ) was also detected in all zooplankton samples. In all cases, however, the amounts of cesium and silver isotopes were much lower than those of naturally occurring potassium-40 in the same samples.

On an expedition in June 2011, biologists collected samples of phytoplankton, zooplankton, shrimp (right) and fish, including the tiny hatchetfish at left, to learn if radioisotopes from Fukushima were accumulating in marine life.





## Tracking radioisotopes in marine life

- 1 Radioisotopes released into the atmosphere from the Dai-ichi nuclear power plant fell into the ocean.
- 2 Water used to cool reactors flushed radioisotopes into the sea.
- 3 Microscopic marine plants (phytoplankton) take up radioisotopes from seawater around them.
- 4 Contaminants move up the food chain from phytoplankton to tiny marine animals (zooplankton), fish larvae, fish, and larger predators. Different radioisotopes are taken up at different rates by different species.
- 5 Some contaminants end up in fecal pellets and other detrital particles that settle to the seafloor and accumulate in sediments.
- 6 Some radioisotopes in sediments may be remobilized into overlying waters and absorbed by bottom-dwelling organisms.

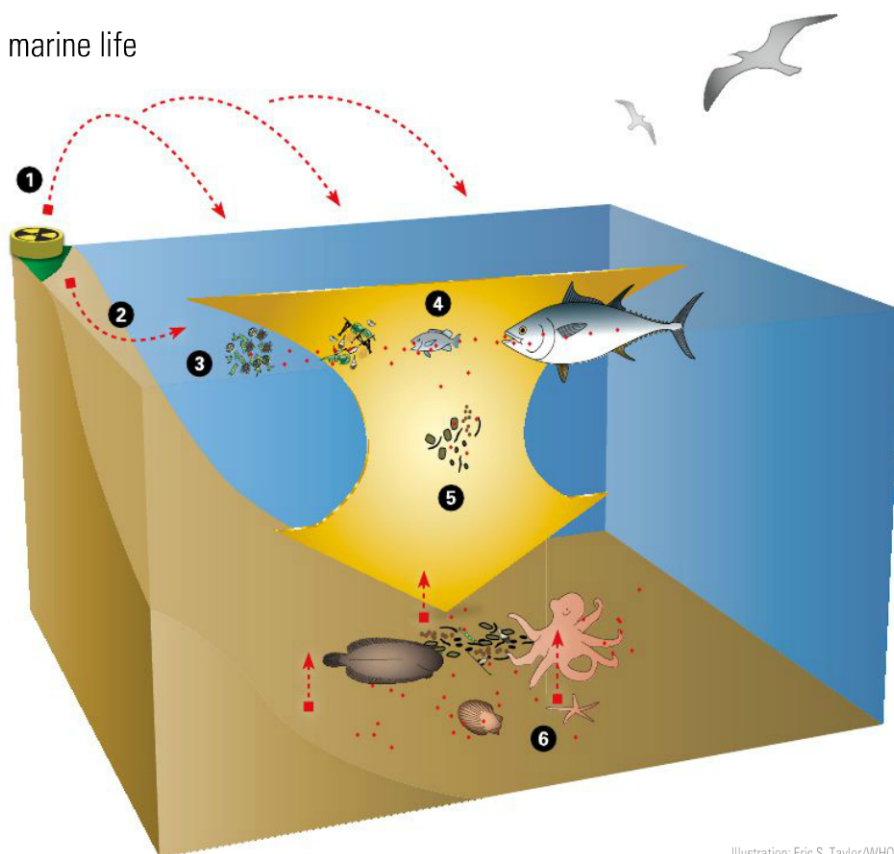


Illustration: Eric S. Taylor/WHOI

“The radioactivity of the fish we caught and analyzed would not pose problems for human consumption,” he said. Which is not, he noted, the same thing as saying that all marine organisms caught in the region are perfectly safe to eat.

### Persistently higher-than-normal levels

What’s puzzling to Fisher, Buesseler, and many other scientists is the persistence of these low but significant levels of radioactivity in the ocean. Jota Kanda, an oceanographer at the Tokyo University of Marine Science and Technology, has extensively studied coastal waters off Fukushima and calculated the amount of cesium still present in coastal waters shallower than 200 meters (660 feet) and in sediments on the seafloor. By his reckoning, what remains is less than three percent of the total discharge, with the rest long since flushed out to the open ocean.

Yet levels of the cesium radioisotopes are still being measured at several tens to hundreds of becquerels per cubic meter in this area, Kanda noted, considerably higher than the levels prior to the Fukushima disaster. More importantly, levels measured in coastal sediments and in some species of fish are higher than those in the surrounding water.

As Kanda sees it, there are three sources responsible for this stubborn presence. One is river runoff—the fallout washed by rainfall into nearby rivers that drain to the sea. He also suggested that a small amount of contaminated water from basement compartments in the reaction unit housing is continuing to leak from the plant itself. But the biggest culprit—the only plausible explanation for the steady levels of radioactive cesium

being measured in fish tissue—is continuous input through a food source. And that, he said, points to sediments.

Kanda has estimated that a total of 95 terabecquerels of cesium ( $10^{12}$  becquerels) is present in coastal sediments. The question, he maintained, is how it got there. It could have drifted down to the seafloor in the fecal pellets of plankton that consumed it at the surface—and in fact, plankton in shallow waters sometimes showed elevated levels of cesium. It could also be arriving with organic bits and pieces carried along by river water. It could have adhered to clay particles that came in contact with contaminated water; such radioactive cesium is tightly bound to clay particles and may not be easily transferred to marine life.

Sediment is complex stuff, he explained. Viewed up close, a single grain of what looks like sand is likely a mélange of mineral, organic matter, and pore water—the liquid trapped in the tiny gaps between particles. How contaminants are taken into these agglomerations is not well understood. Echoing Scott Fowler, Kanda noted that the composition and properties of sediments can vary dramatically.

Solving the mystery of the ongoing radioactivity will require a thorough analysis of the seafloor off Fukushima’s coast, he stressed. “Local communities are concerned. They want to know ‘When can we resume fishing?’ We scientists will have to answer this question.”

The key may be how long cesium stays put and the pathways for its uptake into the food chain. Given the 30-year half-life of cesium-137, the sediments could be a possible source of contamination in the food chain for decades to come.

# Tale of the Tuna

Understanding the movement of Fukushima-derived radioactivity through marine ecosystems may come down to getting a better handle on the tiniest of creatures—the microscopic plankton that take up so much volume in the sea. But one species that has become emblematic of the disaster is a shimmering giant: the Pacific bluefin tuna.

Increasingly overfished, Pacific bluefins are among the most prized table fish in the world. A single 500-pound specimen recently fetched \$1.76 million in a Tokyo auction. Beyond their allure as high-end sushi material, however, they are amazing migratory animals. Spawned in the waters off Japan and the Philippines, these fish as juveniles swim the entire 6,000-mile breadth of the Pacific—a four-month journey—to fatten up in food-rich waters off California. Years later, larger and sexually mature adults undertake a return crossing to spawn.

As respective experts on radioisotope uptake in marine life and tuna migration patterns, Nicholas Fisher of Stony Brook University and Daniel Madigan of Stanford's Hopkins Marine Station knew that young bluefins caught off California during the summer of 2011 likely would have spent their early days in contaminated waters off Fukushima. Would these fish act as “biological vectors” transporting radioisotopes between distant shores?

To find out, Fisher and Madigan obtained tissue samples from tuna caught by sport fishermen off San Diego in August 2011, and analyzed them in Fisher's lab. “Every single bluefin we tested—15 out of 15—had both cesium-134 and cesium-137”—telltale evidence of contamination from the damaged Dai-ichi nuclear power plant, Fisher said. “We were quite surprised to see that.”

The radiation levels they measured were “very, very low,” Fisher stressed. In bluefin tuna caught off San Diego, the total radioactive cesium levels were 10 becquerels, only three percent above radiation levels from naturally occurring potassium-40, and far below safe-consumption levels set by the United States and Japanese governments.

Estimating that the migrating tuna would have lost two percent of any absorbed cesium per day as they crossed—but also would have picked up traces of Cold War-era cesium-137 during their journey—Fisher and his colleagues back-calculat-

ed that when the fish left Japanese waters, the concentrations in them were likely to be 15 times higher, about 150 becquerels per kilogram.

Fisher and Madigan ruled out the possibility that the cesium they measured had been carried on ocean currents or through the atmosphere by also sampling yellowfin tuna, which reside off California but do not migrate across the Pacific Ocean. They found no cesium-134 in these fish, and only background levels of cesium-137.

Fisher and Madigan published their results in late May of 2012, and the response was titanic. “Seven hundred U.S. newspapers, and 400 elsewhere, carried this story,” Fisher recalled, “often on the front page.” He submitted to countless interviews and made several television appearances to try to explain his findings.

“People were genuinely terrified of radioactivity,” he said, “and yet few people could even define it.” To address the anxiety, Fisher and French colleagues calculated dosages, comparing the radiation a person would ingest from eating these bluefin tuna (0.000008 millisieverts) to that received from eating a banana, with its natural potassium (0.0001 mSv), getting a dental X-ray (0.005 mSv), or taking a transcontinental flight (0.04 mSv). He would be more concerned about the health impacts of mercury in these fish, he said, than about radiation.

The scientists continue to analyze radioactivity in bluefins, and Madigan, Fisher, and Zofia Baumann, also in Fisher's group, recently reported that bluefins caught off San Diego in 2012 had less than half the radioactive cesium of the 2011 tuna, indicating that radioactive cesium concentrations in tissues were indeed declining.

But to Fisher, the real importance so far of these findings is that the presence of Fukushima radioisotopes could be used as unequivocal tracers of migratory patterns of bluefin tuna and possibly other large migratory animals such as sharks, seabirds, and loggerhead turtles. And understanding the timing and routes of migration patterns can help manage fisheries and devise more effective conservation strategies for threatened species.

—David Pacchioli

Fukushima radioisotopes can track the routes and rates of migratory species.

