Radioisotopes in the Ocean

What's there? How much? How long?

By David Pacchioli

The release of radioisotopes from the Fukushima Dai-ichi nuclear power plant in March 2011 amounts to the largest-ever accidental release of radiation to the ocean. It came mostly in the form of iodine-131, cesium-134 and cesium-137, the primary radioisotopes released from the reactors, reported Ken Buesseler, a marine chemist at Woods Hole Oceanographic Institution.

All of these substances can cause long-term health problems, said Buesseler, but iodine-131 has a half-life of just eight days and so would be effectively gone from the environment in a matter of weeks. It was cesium-134 and cesium-137, with their half-lives of two and 30 years, respectively, which would remain in the ocean for years and decades to come.

In fact, most of the cesium present in today's oceans, Buesseler noted, is a remnant of atmospheric nuclear weapons testing conducted by the United States, France, and Great Britain during the 1950s and '60s. Lesser amounts are attributable to the Chernobyl nuclear accident in 1986 and to local sources, such as the dumping of low-level waste from England's Sellafield nuclear facility into the Irish Sea.

Prior to Fukushima, however, the levels of cesium-137 off the coast of Japan, as cataloged by Michio Aoyama at the Meteorological Research Institute in Japan and others, were among the world's lowest, at around 2 becquerels per cubic meter (1 becquerel, or Bq, equals one radioactive decay event per second). Against this background, the concentrations measured in early April of 2011 were all the more alarming. At

Photos by Ken Kostel/WHOI

the source waters closest to the Fukushima Dai-ichi plant operated by the Tokyo Electric Power Co., concentrations of up to 60 million becquerels per cubic meter were reported, high enough to cause reproductive and health effects in marine animals.

Most of the cesium from Fukushima came from the millions of gallons of water poured onto the reactors during efforts to cool them, which subsequently flowed into the ocean as runoff or via groundwater. One major leak from flooded buildings at the plant also added cesium to the ocean. Once the leak was plugged in early April, cesium levels close to shore fell off dramatically, Buesseler said. They did not, however, fall out of sight.

"Dilution due to ocean mixing should be enough to cause a decrease in concentration down to background levels within a short period of time," Buesseler told his audience at the Fukushima and the Ocean conference in November 2012. "Yet all the data we have show that measurements around the site remain elevated to this day at up to 1,000 becquerels per cubic meter."

He hastened to put that number into context. "A thousand becquerels is not a big number for cesium. Just for comparison, that's lower than the U.S. Environmental Protection Agency's limit for drinking water. At that level, Buesseler stressed, the cesium in Japanese coastal waters is safe for marine life and for human exposure.

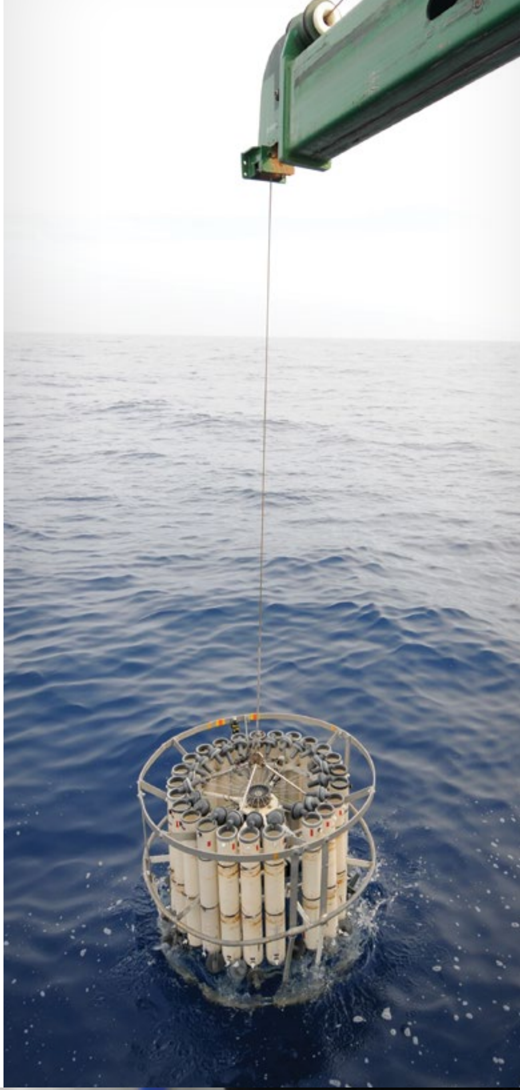
"It's not direct exposure we have to worry about, but possible incorporation into the food chain," he said. That, and the ongoing high levels of radioactive cesium. "The fact that they have leveled off and remained higher than they were before the accident tells us there is a small but continuous source from the reactor site."

The routes and rates of radiation

Away from the coast in the open ocean, the radioactivity that showed up first came from fallout from the atmosphere carried out to sea by winds. The winds limited radioactive exposure on land, as more than 80 percent of the fallout fell on the sea.

Only a few weeks after the accident, a research cruise undertaken by Makio Honda and colleagues at the Japan Agency for Marine-Earth Science and Technology, or JAMS-TEC, detected low levels of both cesium-137 and cesium-134 some 1,900 kilometers (1,180 miles) from Fukushima, from radioactive gases carried out to sea by winds. The importance of the shorter-lived cesium-134 isotope, Buesseler said, was that it offered definitive proof that the contamination had originated from Fukushima. Cesium-134 is not naturally present in the ocean, and it has a half-life of only two years. Any amount of it introduced by weapons testing or other pre-Fukushima sources would have long since disappeared.

In the weeks after the Fukushima nuclear disaster, Ken Buesseler (far left), a marine chemist at Woods Hole Oceanographic Institution, organized an expedition with scientists from different fields and institutions to investigate radioisotopes from the damaged nuclear plant that ended up in the ocean and marine life. They used nets to sample organisms and instruments such as the one at right to collect more than 1,500 water samples in 30 locations off Japan. Water and biological samples were sent to 16 labs in seven countries to detect levels of a variety of radioisotopes.



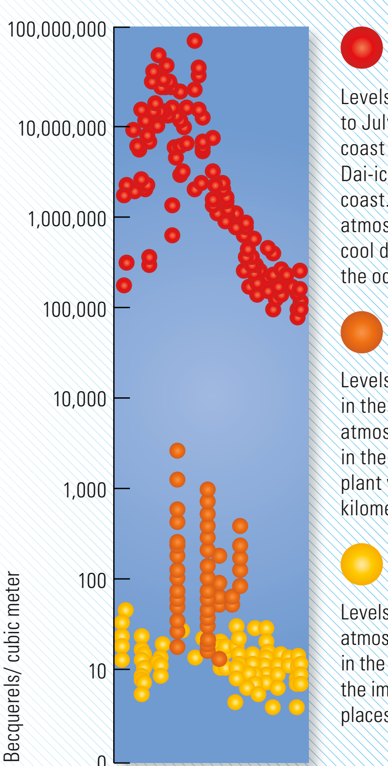
In June 2011, Buesseler led a quickly organized expedition aboard the research vessel *Ka'imikai-o-Kanaloa* that took a comprehensive look at the fate of Fukushima radiation both in the open ocean and in marine life. Beginning 600 kilometers offshore and coming within 30 kilometers of the crippled nuclear plant, the research team sailed a sawtooth pattern, gathering water samples from as deep as 1,000 meters, and collecting samples of phytoplankton, zooplankton, and small fish. (See Page 12.) They also released two dozen drifters to track currents. These instruments move with ocean currents over months and report their positions via satellite.

Like their Japanese colleagues, Buesseler's team measured elevated levels of both cesium-137 and the telltale cesium-134 in the water they collected. These levels varied widely across the sampling area, however, which indicated the complexity of the currents at play—dominated by the mostly eastward flow of the mighty Kuroshio Current, the Pacific Ocean's equivalent of the Gulf Stream in the Atlantic.

"The first thing we noticed was that when we got to the Kuroshio, we lost the cesium-134 signal," Buesseler said. "That confirmed that the Fukushima radioactive fallout from the atmosphere did not reach these more southern latitudes." The scientists believe the Kuroshio acted as a barrier, blocking radioisotope-contaminated waters from flowing through it and to the south.

An Unprecedented Release Of Radioisotopes to the Ocean

The amount of cesium-137 radioisotopes from the Fukushima disaster in surface ocean waters was 10,000 to 100,000 times greater than amounts that entered the ocean from the Chernobyl accident or atmospheric nuclear weapons tests.



Fukushima
2011

Levels of radioisotopes measured from March to July 2011 in surface ocean waters off the coast of Japan following the disaster at the Dai-ichi nuclear power plant located on the coast. Radioisotopes entered the ocean from atmospheric fallout and from water used to cool damaged reactors, which flushed into the ocean.

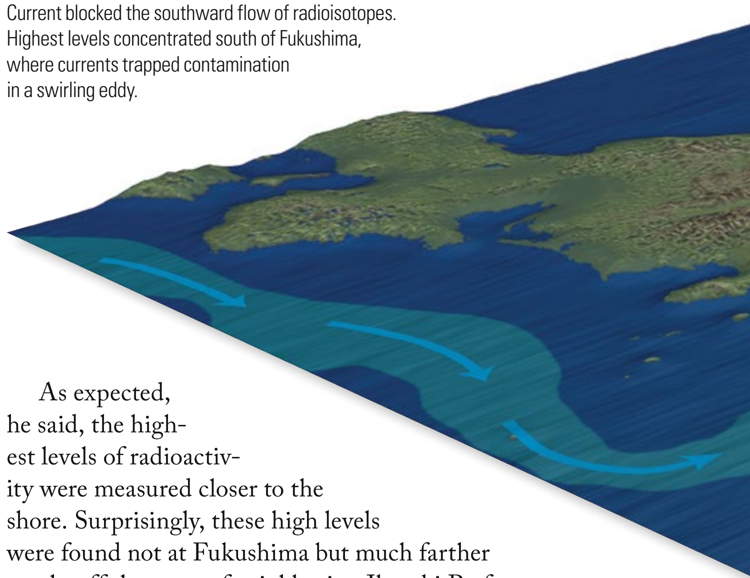
Chernobyl
1986

Levels of radioisotopes measured in 1986 in the Baltic Sea and Black Sea from atmospheric fallout following the disaster in the Chernobyl Nuclear Power Plant. The plant was located in Ukraine, hundreds of kilometers inland.

Weapons Testing
1960s

Levels of radioisotopes from fallout from atmospheric nuclear weapons tests, measured in the 1960s off Japan. Levels were higher in the immediate areas near Bikini Atoll and others places where weapons were detonated.

Starting 375 miles off Japan, researchers on the *Ka'imikai-o-Kanaloa* measured radioisotope levels in various locations. They found that the Kuroshio Current blocked the southward flow of radioisotopes. Highest levels concentrated south of Fukushima, where currents trapped contamination in a swirling eddy.



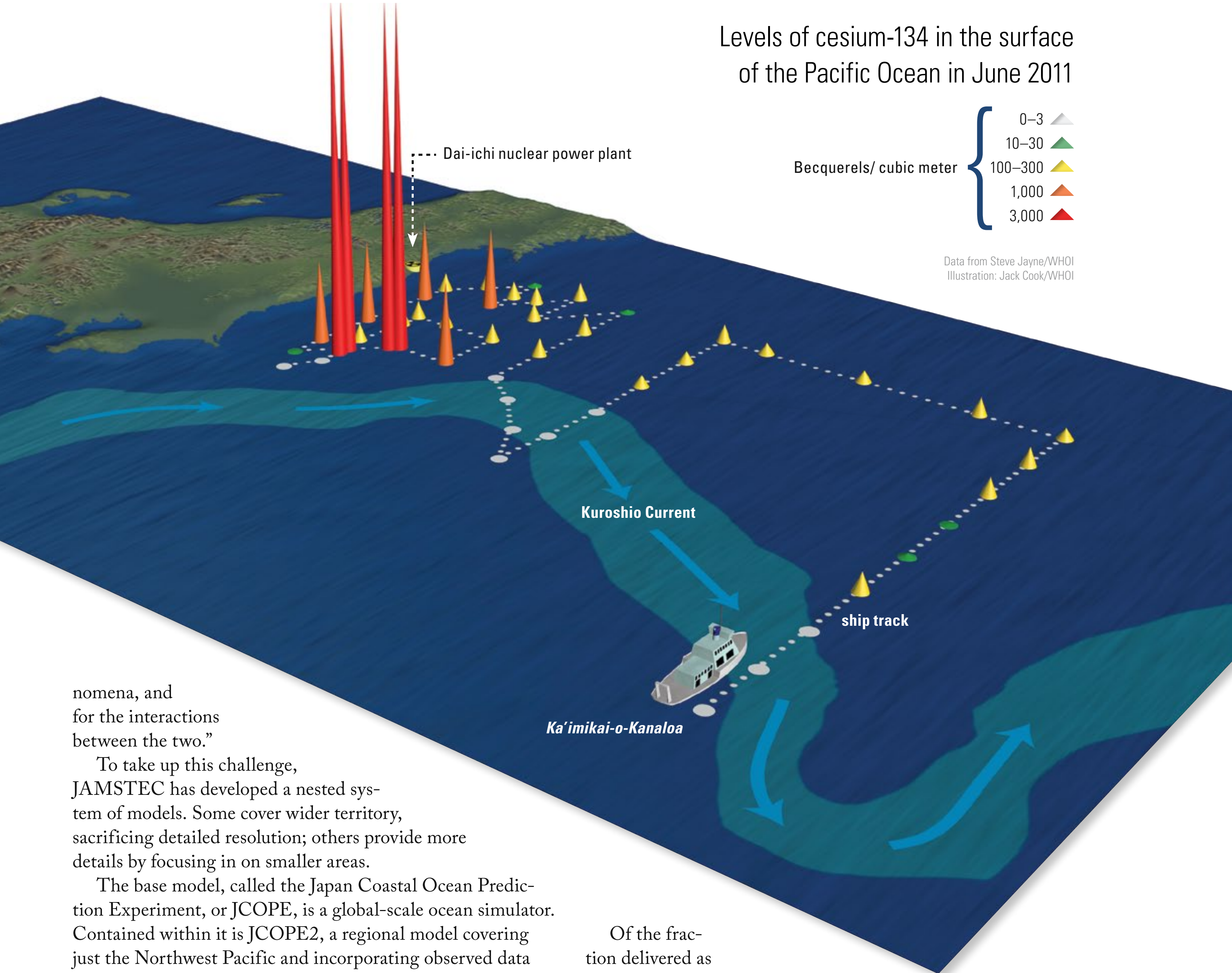
As expected, he said, the highest levels of radioactivity were measured closer to the shore. Surprisingly, these high levels were found not at Fukushima but much farther south, off the coast of neighboring Ibaraki Prefecture. The drifter data showed that this seeming anomaly was the result of a large circular flow, or eddy, that had trapped contamination close to shore south of the plant.

Over time, the researchers' drifters helped to establish the routes and rate of radiation transported out into the wider ocean and revealed the complexity of currents in the region. A year after the drifters were released, WHOI oceanographer Steve Jayne showed that their circuitous tracks extended halfway across the Pacific, remaining mostly north of the Kuroshio Current. Combined with surface-water samples taken by commercial "ships of opportunity" in a program organized by Aoyama, these data show cesium mixing down into the ocean and flowing east at a rate of about 7 kilometers per day. At that rate, Buesseler said at the November 2012 conference, it would take another year for small but measurable amounts of cesium to show up off the U.S. West Coast.

Modeling the ocean

Lacking dipsticks in the ocean to measure the ongoing dispersal of radioisotopes, or regular research cruises, scientists must rely on mathematical ocean models to predict where radioactivity from Fukushima is flowing. These incorporate the best-known physics of ocean current dynamics, but modeling the unique currents off Japan's Pacific coast can be a frustrating task, JAMSTEC's Yukio Masumoto said at the Tokyo conference.

In particular, the waters off Fukushima are squeezed between the Kuroshio, which flows northeastward near Chiba Prefecture and then turns eastward, and the colder and fresher Oyashio Current, which flows south from the Arctic Ocean. The collision of these two major currents in the western North Pacific creates a region of dynamically mixing waters that makes for prolific fishing grounds, Masumoto said, and also produces 100- to 1,000-kilometer-wide eddies and complicated small-scale effects, all of which must be incorporated into any large-scale simulation. "In addition to these open-ocean phenomena," he said, "we must also account for coastal phe-



nomena, and for the interactions between the two."

To take up this challenge, JAMSTEC has developed a nested system of models. Some cover wider territory, sacrificing detailed resolution; others provide more details by focusing in on smaller areas.

The base model, called the Japan Coastal Ocean Prediction Experiment, or JCOPE, is a global-scale ocean simulator. Contained within it is JCOPE2, a regional model covering just the Northwest Pacific and incorporating observed data on temperature and salinity in order to produce realistic ocean currents. Tucked within the regional model, and offering still finer resolution, is JCOPET, a coastal model that can simulate tidal effects.

Masumoto stressed that the accuracy of each of these dispersion models—and several others currently being run at his and other institutions in Japan and elsewhere—depends largely on actual measurements put into the models before they are set in motion.

"The observed data are quite limited in time and space, and we cannot say which model is the best," Masumoto said—especially given the ongoing uncertainty about the amount of radioisotopes that entered the ocean.

Buesseler reviewed the range of current estimates of the total cesium releases. Their totals vary widely, he noted, but are "beginning to converge" on a total cesium-137 release of between 15 and 30 petabequerels (10^{15} Bq). In comparative terms, he said, this is slightly more than the amount put into the sea by Chernobyl—although the total environmental release from that accident, at 85 PBq, was much higher.

Of the fraction delivered as atmospheric fallout, most estimates lie between 10 and 15 PBq. The numbers for direct discharge, 3 to more than 15 PBq, are much less certain, Buesseler noted. Nor do these total numbers account for the release of other isotopes from lingering plant discharges, including strontium-90, which, although present in quantities much lower than cesium, is of concern: It accumulates in the bones of fish that could be eaten.

To understand the ultimate fate of Fukushima cesium in the oceans, scientists will have to uncover not just the paths and speed of ocean currents that transport cesium across the Pacific, but also how cesium mixes into deeper layers of the ocean and how much accumulates in particles of organic detritus, or "marine snow," that sinks down and settles on the seafloor. Smaller but ongoing radioactive inputs from rivers and groundwater in the vicinity are being reported by Japanese scientists, and will also have to be considered. Although some of these puzzle pieces are beginning to fit together, Buesseler said, significant gaps in knowledge remain.