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Impacts of the Fukushima Nuclear Power Plants on Marine Radioactivity

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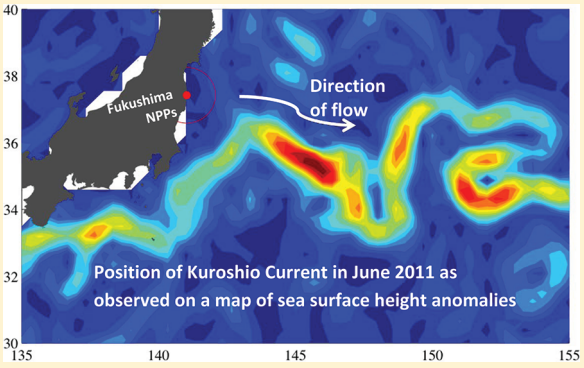
Supporting Information

ABSTRACT: The impacts on the ocean of releases of radionuclides from the Fukushima Dai-ichi nuclear power plants remain unclear. However, information has been made public regarding the concentrations of radioactive isotopes of iodine and cesium in ocean water near the discharge point. These data allow us to draw some basic conclusions about the relative levels of radionuclides released which can be compared to prior ocean studies and be used to address dose consequences as discussed by Garnier-Laplace et al. in this journal.[1] The data show peak ocean discharges in early April, one month after the earthquake and a factor of 1000 decrease in the month following. Interestingly, the concentrations through the end of July remain higher than expected implying continued releases from the reactors or other contaminated sources, such as groundwater or coastal sediments. By July, levels of ^{137}Cs are still more than 10 000 times higher than levels measured in 2010 in the coastal waters off Japan. Although some radionuclides are significantly elevated, dose calculations suggest minimal impact on marine biota or humans due to direct exposure in surrounding ocean waters, though considerations for biological uptake and consumption of seafood are discussed and further study is warranted.



INTRODUCTION

As a result of the earthquake on March 11, 2011, and subsequent tsunami, water as high as 15 m inundated the Dai-ichi nuclear power plants (NPPs) causing loss of power and hence disruption of controls and failed cooling systems shortly after the earthquake. Venting of gases, hydrogen explosions, and the fire in the spent fuel pond of Unit 4 resulted in the primary atmospheric releases of Fukushima radionuclide contaminants, peaking around March 15, with a relatively high atmospheric release rate through March 24.[2] In addition to these atmospheric fallout pathways, the cooling of the reactors with fresh water and seawater, and release of highly contaminated water from the damaged reactor buildings led to radioactive discharges directly to the sea. Some of this was intentional (to leave space for more highly contaminated waters); some was unconstrained and likely resulted from contaminated groundwater discharges as well as direct runoff to the sea. Unlike Chernobyl, there was no large explosive release of core reactor material, so most of the isotopes reported to have spread thus far via atmospheric fallout are primarily the radioactive gases plus fission products such as cesium, which are volatilized at the high temperatures in the reactor core, or during explosions and fires. However, some nonvolatile activation products and fuel rod materials may have been released when the corrosive brines and acidic waters used to cool the reactors interacted with the ruptured fuel rods, carrying radioactive materials into the ground and ocean. The full magnitude of the release has not been well documented, nor is there data on many of the possible isotopes released, but we do have significant information on the concentration of several isotopes of Cs and I in the ocean near the release point which have been publically available since shortly after the accident started.

DATA SOURCES

We present a comparison of selected data made publicly available from a Japanese company and agencies and compare these to prior published radionuclide concentrations in the oceans. The primary sources included TEPCO (Tokyo Electric Power Company), which reported data in regular press releases[3] and are compiled here (Supporting Information Table S1). These TEPCO data were obtained by initially sampling 500 mL surface ocean water from shore and direct counting on high-purity germanium gamma detectors for 15 min at laboratories at the

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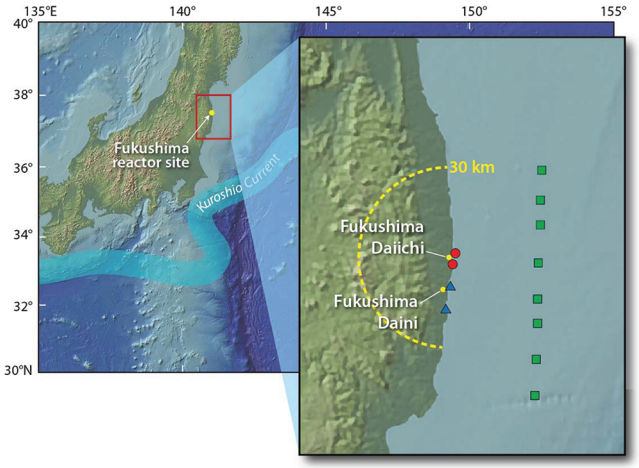

Figure 1. Map of Fukushima sampling locations at the Dai-ichi NPP (1F- yellow dot as indicated). Red dots N and S of the Dai-ichi NPP are discharge channels where samples were collected. Samples were also collected by TEPCO at the Dai-ni NPP (2F- yellow dot as indicated) with sampling indicated from shore near Dai-ni NPP and Iwasawa Beach (blue triangles). Also shown are sampling locations by MEXT 30 km offshore (green squares). For scale, 30 km radius around Fukushima is shown on land. More detailed sampling maps available at TEPCO and MEXT Web sites.[3,4]

Fukushima Dai-ni NPPs. They reported initially results for ^{131}I ($t_{1/2}$ = 8.02 days), ^{134}Cs ($t_{1/2}$ = 2.065 years) and ^{137}Cs ($t_{1/2}$ = 30.07 years). Data from MEXT (Ministry of Education, Culture, Sports, Science and Technology—Japan) were also released on a public Web site[4] and are based on similar direct counting methods. In general MEXT data were obtained by sampling 2000 mL seawater and direct counting on high-purity germanium gamma detectors for 1 h in a 2 L Marinelli beaker at laboratories in the Japan Atomic Energy Agency. The detection limit of ^{137}Cs measurements are about 20 000 Bq m^{-3} for TEPCO data and 10 000 Bq m^{-3} for MEXT data, respectively. These measurements were conducted based on a guideline described by MEXT.[5] Both sources are considered reliable given the common activity ratios and prior studies and expertise evident by several Japanese groups involved in making these measurements. The purpose of these early monitoring activities was out of concern for immediate health effects, and thus were often reported relative to statutory limits adopted by Japanese authorities, and thus not in concentration units (reported as scaling factors above "normal"). Here we convert values from both sources to radionuclide activity units common to prior ocean studies of fallout in the ocean (Bq m^{-3}) for ease of comparison to previously published data.

RESULTS AND DISCUSSION

We focus on the most complete time-series records from the north and south discharge channels at the Dai-ichi NPPs, and two sites to the south that were not considered sources, namely the north Discharge channels at the Dai-ni NPPs about 10 km to the south and Iwasawa beach which is 16 km south of the Dai-ichi NPPs (Figure 1). The levels at the discharge point are exceedingly high, with a peak ^{137}Cs 68 million Bq m^{-3} on April 6 (Figure 2). What are significant are not just the elevated concentrations, but the timing of peak release approximately one month after to the earthquake. This delayed release is

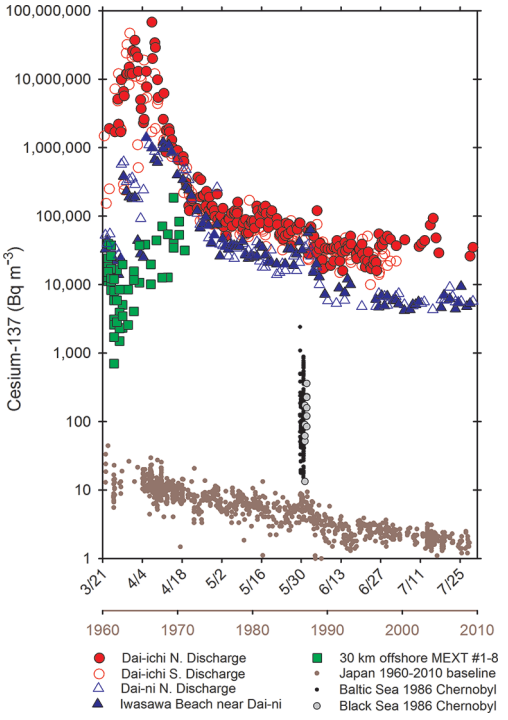

Figure 2. Surface ocean concentrations from March 21 to July 31, 2011 of ^{137}Cs in Becquerels per cubic meter (Bq m^{-3}) for two sites near the Fukushima Dai-ichi nuclear power plant (red circles, north (filled) and south (open) discharge channels[3]), Dai-ni NPPs (10 km to the south of Dai-ichi, blue filled triangles[3]), Iwasawa Beach near Dai-ni (16 km south of Dai-ichi, blue open triangles[3]), and 30km off-shore (green squares, stations 1−8 in original MEXT data[4]). These are compared on the lower *X*-axis (1960−2010) to the historical record of ^{137}Cs off the east coast of Japan (brown circles) and to Chernobyl influenced waters in 1986 in the Baltic and Black Seas.[13,14]

presumably due to the complicated pattern of discharge of seawater and fresh water used to cool the reactors and spent fuel rods, interactions with groundwater, and intentional and unintentional releases of mixed radioactive material from the reactor facility.

During the first month of release data, $^{134}Cs/^{137}Cs$ activity ratios were one (0.99 ± 0.03 for Dai-ichi north and south discharge channels) and extremely uniform (Supporting Information Figure S1). This makes the tracking of Fukushima derived radionuclides in the ocean quite straightforward, since given its relatively short 2 year half-life, the only source of ^{134}Cs in the North Pacific at this time would be the Dai-ichi NPPs. Hence in addition to the elevated Cs activities, the presence of ^{134}Cs is a unique isotopic signature for tracking these waters and calculating mixing ratios. This ratio of Cs isotopes is determined by reactor design and fuel cycle and age. Interestingly a $^{134}Cs/^{137}Cs$ ratio of 1.0 here is considerably higher than 25 years ago when a ratio of 0.54 ± 0.04 was reported in Chernobyl fallout.[6] In the oceans, the behavior of cesium is thought to be conservative, i.e. it is soluble (<1% attached to marine particles) and is carried primarily with ocean waters and as such has been used as a tracer of water mass mixing and transport.[7−9] That being said, even a small fraction at such a high activity release point is a large number, thus the concentrations of Cs in sediments and biota near the NPPs may be quite large, and will continue to remain so for at least 30−100 years due to the longer half-life of

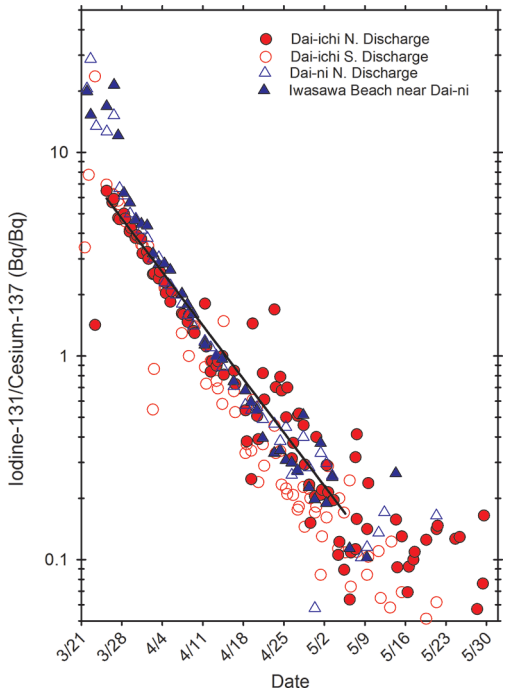

Figure 3. Activity ratio of $^{131}I/^{137}Cs$ at the same four sites at Dai-ichi, Dai-ni and Iwasawa Beach as in Figure 2 plotted on a log activity ratio (*y*-axis) vs time through May 30 (*x*-axis). Solid black line is the decay trend expected for the activity of an isotope with 8 day half-life such as ^{131}I.

^{137}Cs which is still detected in marine and lake sediments from 1960s fallout sources.

There was considerable attention given to ^{131}I releases due to its relatively high activities and tendency to accumulate in the human thyroid if ingested via land-based food supply or if bioconcentrated by seaweeds and consumed as part of the Japanese diet. We can see that the ocean release ratio of $^{131}I/^{134}Cs$ must have been relatively constant, with the highest measured activity ratios near 20−30 on March 22 followed by a predictable decrease due to the radioactive decay of ^{131}I with its 8 day half-life (which would plot as straight line in Figure 3). Assuming the source ratios of Cs and I were fixed when the fission process stopped on March 13, the initial source ratio would have been a factor of 2 higher. Reported atmospheric $^{131}I/^{137}Cs$ ratios showed somewhat higher variability early in the releases, ranging from around 10−80 during the period of maximum atmospheric release between March 15 and 24[2]. Some of the ocean data fall off of this ocean decay trend, indicating perhaps different sources (e.g., higher values around April 19−23 at Dai-ichi NPPs, Figure 3), but the general consistency of this decay pattern in waters both at Dai-ichi NPPs and further south indicates that overall the I and Cs sources can be considered uniform even if several reactors and events contributed to the releases.

Ocean currents off Japan would lead to both southward transport of water along the coast via the Oyashio current, and northward driven diversions due to surface wind shifts.[10,11] Clearly the waters 10 km south along the shore at the Dai-ni NPPs were initially lower in overall activity, but rapidly reached activities more similar to the source at Dai-ichi. Coastal water concentrations decreased by close to a factor of 1000 in the month following peak releases (Figure 2). This is a consequence of ocean mixing and a primary radionuclide source that has dramatically abated. Unlike contamination of soils on land, vertical and horizontal mixing rates in the ocean are fast, diluting the primary contaminant signal quite rapidly, particularly in the energetic coastal waters off Japan where the Oyashio waters move south and interact with the rapidly flowing and offshore meandering of the Kuroshio Current. These currents, tidal forces, and eddies mix the waters quite rapidly offshore. Such physical transport and mixing processes can be modeled, but thus far there are no published models that include Fukushima ocean data for comparison (see for example Fukushima fallout predictions in ref 12).

Sampling immediately off shore was sponsored by MEXT in Japan, and we have summarized here the first month's data at eight stations along a transect 30 km offshore from the Fukushima NPPs (along 141° 24′ E. Longitude between 37° 00′ and 37° 40′ N Latitude; Figure 1). From the first measurements on March 22 until about March 28, activities decreased. This decrease can be explained if the initial data reflect elevated ^{137}Cs from direct Fukushima fallout deposition earlier in March,[2] followed by dilution. The waters 30 km offshore increased after March 28 in parallel with the coastal waters at Dai-ni. The offshore waters have roughly 1000 times lower activities during the period of peak discharge in early April and remain at least 10 times lower later in April. The grid sampled by MEXT expanded considerably in April to include additional stations between the coast and 30 km, but after about April 20, the reported MEXT data fall largely below their minimum detectable limit, which was 10 000 Bq m^{-3} for ^{137}Cs.

If the source at Fukushima had stopped abruptly and ocean mixing processes continued at the same rates, one would have expected that the ^{137}Cs activities would have decreased an additional factor of 1000 from May to June but that was not observed. The break in slope in early May implies that a steady, albeit lower, source of ^{137}Cs continues to discharge to the oceans at least through the end of July at this site. With reports of highly contaminated cooling waters at the NPPs and complete melt through of at least one of the reactors, this is not surprising. As we have no reason to expect a change in mixing rates of the ocean which would also impact this dilution rate, this change in slope of ^{137}Cs in early May is clear evidence that the Dai-ichi NPPs remain a significant source of contamination to the coastal waters off Japan. There is currently no data that allow us to distinguish between several possible sources of continued releases, but these most likely include some combination of direct releases from the reactors or storage tanks, or indirect releases from groundwater beneath the reactors or coastal sediments, both of which are likely contaminated from the period of maximum releases.

It is prudent to point out though what is meant by "significant" to both ocean waters and marine biota. With respect to prior concentrations in the waters off Japan, all of these values are elevated many orders of magnitude. ^{137}Cs has been tracked quite extensively off Japan since the peak weapons testing fallout years in the early 1960s.[13] Levels in the region east of Japan have decreased from a few 10s of Bq m^{-3} in 1960 to 1.5 Bq m^{-3} on average in 2010 (Figure 2; second *x*-axis). The decrease in ^{137}Cs over this 50 year record reflects both radioactive decay of ^{137}Cs with a 30 year half-life and continued mixing in the global ocean of ^{137}Cs to depth. These data are characteristic of other global water masses.[14] Typical ocean surface ^{137}Cs activities range from <1 Bq m^{-3} in surface waters in the Southern Hemisphere, which are lower due to lower weapons testing inputs south of the

equator, to >10−100 Bq m^{-3} in the Irish Sea, North Sea, Black Sea, and Baltic Seas, which are elevated due to local sources from the intentional discharges at the nuclear fuel reprocessing facilities at Sellafield in the UK and Cape de la Hague in France, as well as residual ^{137}Cs from Chernobyl in the Baltic and Black Seas. Clearly then on this scale of significance, levels of ^{137}Cs 30 km off Japan were some 3−4 orders of magnitude higher than existed prior to the NPP accidents at Fukushima.

An additional comparison can be made, not just to current global ocean ^{137}Cs levels, but to what was measured immediately following the Chernobyl accident in 1986. An increase in 1986 in waters off Japan is barely seen in the time-series record off Japan, but in the Baltic and Black Seas they peaked in 1986 in the 10−1000 Bq m^{-3} range (Figure 2). This is thus well below activities of ^{137}Cs immediately at the discharge point or even the initial 30 km monitoring line of MEXT. As such, despite some uncertainty over the total releases from Fukushima vs Chernobyl to both land and sea, the accidental releases from Fukushima are a larger source to the ocean. That should not be surprising as fallout deposition in general decreases with distance from the source (both in air and ocean). Since the Dai-ichi NPPs are directly adjacent to the ocean and Chernobyl was 500−600 km from the closest ocean bodies of the Baltic and Black Seas, Fukushima has become the largest accidental source of radionuclides to the ocean in terms of measured radionuclide concentrations.

Finally though, while the Dai-ichi NPP releases must be considered "significant" relative to prior sources off Japan, we should not assume that dose effects on humans or marine biota are necessarily harmful or even will be measurable. Garnier-Laplace et al.[1] report a dose reconstruction signal for the most impacted areas to wildlife on land and in the ocean. Like this study, they are relying on reported activities to calculate forest biota concentrations, and TEPCO ocean data for the expected concentration in the ocean and by calculation in marine sediments and the doses to benthic biota. By this calculation the dose effect due to forest soils on land ecosystems were small, 2−6 mSv d^{-1} (converted here from Gy using a relative biological effectiveness factor of 1, appropriate for doses due to I and Cs isotopes, so 1 Sv = 1 Gy). In contrast, these authors report much higher doses for marine benthic biota and ocean birds, in the range of 210−4600 mSv d^{-1}. These authors conclude that impacts to the marine biota would be severe, including marked reproductive effects and possibly mortality due to direct dose effects. While these effects were carefully defined as maximum dose rates (for activities at equilibrium), Figure 2 makes it clear that for the oceans even at the discharge point, there were >1000 times lower radionuclide activities as quickly as one month after peak releases and even lower activities off shore, which would bring these doses to levels quickly below where there is any effect for ecosystems (defined as 0.24 mSv d^{-1} by these authors; see also ref 15).

With respect to dose effects on humans, at a level approaching 100 000 Bq m^{-3} for ^{134}Cs and ^{137}Cs found at the Dai-ichi discharge channels in June the dose due to direct exposure during human immersion in the ocean can be calculated to be 1 μSv d^{-1}, and would be at least a factor of 10 lower if on a ship above and not in direct contact with the water. This is insignificant relative to the average dose from all sources to the Japanese population of about 1.5 mSv yr^{-1}. This low dose should not be surprising, as levels of the most abundant naturally occurring radionuclide in the oceans, potassium-40, are comparable to ^{137}Cs offshore, with typical ocean value of 12 000 Bq m^{-3}. Levels of ^{137}Cs in June and July at Dai-ni and Iwasawa Beach of 4000 to 10 000 Bq m^{-3} were comparable to permissible drinking water limits for ^{137}Cs, which in the US are 7400 Bq m^{-3} (EPA limit of 40 μSv yr^{-1} calculated here for 1 L per day consumption) and 10 000 Bq m^{-3} recommended by the World Health Organization. Thus even at the observed concentrations at the discharge channels in June and July there will be no significant direct dose effect on humans, and only a short distance away, ^{137}Cs concentrations would be below drinking water limits for Cs isotopes.

This dose assessment does not, however, consider bioaccumulation and consumption of seafood and seaweeds and possible impacts on humans. The waters immediately adjacent to Japan remain a continued source of radionuclides that is keeping the discharge waters elevated in ^{137}Cs, and thus likely other Fukushima-derived radionuclides that have not yet been reported. Locally elevated marine sediment concentrations are expected and this would imply possible additional pathways for assimilation in biota near shore by filter feeding shellfish and benthic marine biota. Brown seaweeds are of particular concern as they are a major crop in Japan and highly efficient at concentrating ^{131}I (concentration factors of 10 000), though with an 8-day half-life, the ^{131}I activities have rapidly declined (Figure 3). The provisional regulatory value in fish established by the Fisheries Agency of Japan is 500 Bq kg^{-1} for radioactive Cs. Early reports off Japan's coast suggest that the only seafood concentrations above safety limits were for sand eels, though a few seafood samples above these levels continue to be found (see for example the July 2011 Japanese Fisheries Agency report[16]). With a Cs concentration factor of 100 for fish,[17] one would approach unacceptable levels of Cs in fish if in equilibrium with ocean activities>5000 Bq m^{-3} for combined Cs isotopes, which we see at the discharge point in July. Also specific pathways such as preferential uptake of ^{90}Sr (not yet measured) in fish bones will need to be considered if whole small fish are consumed. Continued monitoring and bans on fishing in Fukushima impacted waters is thus warranted given the steady elevated levels near the NPPs. Given that Japan has the highest seafood consumption rate in the world, understanding concentrations and assimilation in marine biota is an important task.

Japanese authorities raised the severity of the Fukushima Daiichi nuclear power plant incident to level 7, the highest level on the international scale and comparable only to the Chernobyl incident 25 years ago. With respect to the oceans, however, the impact of Fukushima exceeds Chernobyl if measured by the changes in radionuclide activities in the surface ocean. However, a decrease in Fukushima activities by a factor of 1000 since its peak in early April and dilution off shore greatly lessens direct impacts to humans and marine biota. That being said, it is important to note that we still do not have sufficient field data to estimate the ocean radionuclide inventories, the full range of isotopes released, the aerial extent of contamination, the fraction delivered as coastal runoff vs atmospheric fallout, the sedimentary burden near the Dai-ichi NPPs, and the biological uptake in the marine food chain beyond a limited number of plankton samples and monitoring of the food supply. Given that this is the largest accidental source of radionuclides to the ocean, it is encouraging to see that international collaborations for comprehensive field measurements are beginning, though it will take some time before results are available to fully evaluate the impacts of this accident on the ocean.

■ ASSOCIATED CONTENT

Supporting Information. Figure with $^{134}Cs/^{137}Cs$ release data for first month at Dai-ichi (Figure S1) and Table S1 with complete TEPCO data selected for discussion in this manuscript. This material is available free of charge via the Internet at http://pubs.acs.org.

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