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Canopy-Forming Kelps as California's Coastal Dosimeter: ^{131}I from Damaged Japanese Reactor Measured in *Macrocystis pyrifera*

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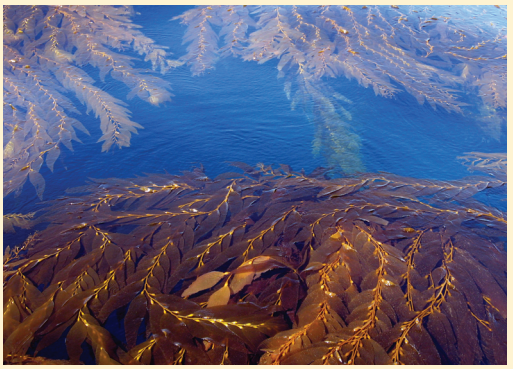
ABSTRACT: The Fukushima Daiichi Nuclear Plant, damaged by an earthquake and tsunami on March 11, 2011 released large amounts of ^{131}I into the atmosphere, which was assimilated into canopy blades of *Macrocystis pyrifera* sampled from coastal California. The specific activity calculated to the estimated date of deposition/assimilation ranged from 0.6 to 2.5 Bq gdwt^{-1}, levels greater than those measured from kelps from Japan and Canada prior to the release. These ^{131}I levels represent a significant input into the kelp forest ecosystem. Canopy-forming kelps are a natural coastal dosimeter that can measure the exposure of the coastal environment to ^{131}I and perhaps other radioisotopes released from nuclear accidents. An organizational mechanism should be in place to ensure that they are sampled immediately and continuously after such releases.



INTRODUCTION

The Fukushima Daiichi Nuclear Plant in Japan, damaged by earthquake and tsunami on March 11, 2011, began to release significant radioactive material into the atmosphere March 12, with the main explosion occurring March 14.[1] Some of the radioactive materials released included ^{131}I ($t_{1/2}$ = 8.0 d), ^{35}S ($t_{1/2}$ = 87.4 d; produced indirectly from cooling the reactor with seawater) with longer lived radionuclides such as ^{133}Xe ($t_{1/2} = 16 \times 10^6$ y), ^{137}Cs ($t_{1/2}$ = 30 y), and ^{134}Cs ($t_{1/2}$ = 2.1 y).[2] Major isotopes released at levels reported by the Japanese government in June were ^{133}Xe (1.1×10^4 PBq, 1 PBq = 10^{15}), ^{131}I (150 PBq), and ^{137}Cs (13 PBq) with the estimated release of radioactive material to the atmosphere being 15% that of the Chernobyl accident.[3,4] Continued analysis of this event suggests that these estimates should be increased, especially for ^{133}Xe and ^{137}Cs.[2] Based on measurements of the radioactive cloud produced from the Chernobyl reactor accident, the most important radioisotopes released in terms of amounts and global radiological impact were ^{131}I and ^{137}Cs.[5] Projected paths of the radioactive atmospheric plume emanating from the Fukushima reactors, best described as airborne particles or aerosols for ^{131}I, ^{137}Cs, and ^{35}S, and subsequent atmospheric monitoring showed it coming in contact with the North American continent at California, with greatest exposure in central and southern California.[1,6] Government monitoring sites in Anaheim (southern California) recorded peak airborne concentrations of ^{131}I at 1.9 pCi m^{-3} from a baseline of zero.[7]

Central and southern California coastal waters are dominated by large surface canopies of Giant Kelp *Macrocystis pyrifera*. Kelps (brown seaweeds of the Order Laminariales) are the strongest accumulators of iodine known; tissue iodine concentrations can be several orders of magnitude (10^4) higher than iodine in the surrounding seawater.[8] Iodine is stored in the tissue as iodide which is effluxed to the thallus surface where it functions as an antioxidant, scavenging reactive oxygen species.[8] Most of the surface area of *Macrocystis* resides in mature canopy blades where iodine uptake is very rapid.[9] Atmospheric ^{131}I is deposited on the earth's surface in precipitation and *Macrocystis* is ideal for detecting ^{131}I present in the atmosphere because the canopies are directly exposed to precipitation at the sea/air interface. Once we became aware of the magnitude of the disaster and the composition of the radioactive plume, a month after the tsunami, we began to obtain samples of *Macrocystis* from local beds that were being visited for other studies, and we sent out requests to several marine biologists along the California coastline, where *Macrocystis* can be found, to send us fresh blades. We also made a request to scientists in Sitka, Alaska. We describe in this report a simple technique by which we detected ^{131}I in *Macrocystis* canopy blades originating from the damaged Japanese plant. We also advocate an organized periodic sampling protocol of kelp bed canopies immediately after such a radioactive release.

MATERIALS AND METHODS

We began sampling canopy blades from southern California and counting ^{131}I gamma emissions in the dried tissue a month (April 15, 2011) after the first atmospheric release (March 14, 2011).[1] We were also successful in obtaining samples of *Macrocystis* blades from several other California locations and one site in Alaska, although in some instances, considerable time had passed (Figure 1 and Table 1). The species *M. integrifolia* was sampled in Sitka because *M. pyrifera* is not found above approximately 38° N. Both are canopy formers and may

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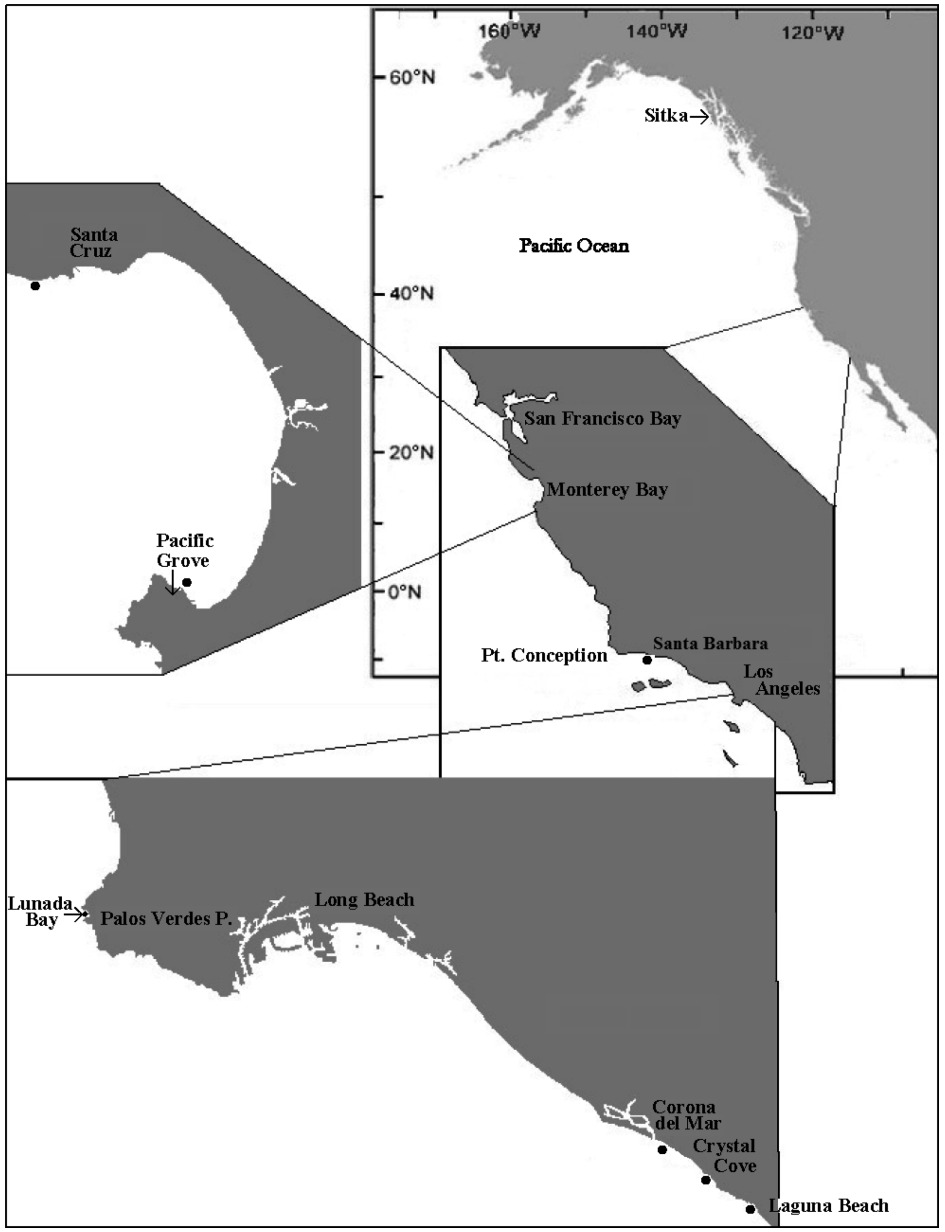

Figure 1. Location of sampling sites designated with ● or by arrow tip for Sitka and Lunada Bay. For specific location coordinates see Table 1.

Table 1. Sampling Information and Specific Activity of ^{131}I in *Macrocystis* Blades[a]

location/position	collector	date collected	date counted	^{131}I counted (dpm)	date assimilated	elapsed[b]	tissue ^{131}I (Bq/gdwt)
Main Beach Laguna Beach, CA 33° 32′ 00″ N 117° 46′ 56″ W	D. Burnett	15-Apr	19-Apr	14.5	21-Mar	3.6	1.4
Crystal Cove St Park Orange County, CA 33°33′ 57″ N 117° 50′ 20″ W	D. Burnett	15-Apr	20-Apr	12.5	21-Mar	3.7	1.4
R. E. Badham St. Marine Conservation Area Corona del Mar, CA 33° 35′ 21″ N 117° 52′ 20″ W	D. Burnett	15-Apr	22-Apr	19.6	21-Mar	4.0	2.5
Lunada Bay Los Angeles County, CA 33°46′49″ N 118°26′0″ W	S. Manley	22-Apr	25-Apr	6.2	21-Mar	4.4	1.1
Goleta Pt. Santa Barbara, CA 34°24′12″ N 119°50′39″ W	D. Chapman	5-May	9-May	1.0	21-Mar	6.1	0.58
Point Cabrillo Pacific Grove, CA 36°37′5″ N 121°53′50″ W	B. Mahoney	26-Apr	29-Apr	3.2	22-Mar	4.7	0.70
Natural Bridges Santa Cruz, CA 36°56′46″ N 122°03′48″ W	K. Lyons	4-May	5-May	4.5	22-Mar	5.5	2.0
Japonski Is Sitka, AK 57°3′20″ N 135°22′25′ W	M. Chapman	9-May	16-May	nd[c]	—	—	—

[a]Specific activity calculated to date of assimilation. [b]Elapsed = half-lives elapsed between time of counting and estimated time of assimilation. [c]nd = not detected.

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be the same species.[10] Samples obtained from southern California were immediately transported in seawater to our laboratory and processed the same day. Samples outside southern California were shipped to us wrapped in seawater-saturated paper towels and sealed in plastic bags on ice by overnight air delivery for immediate processing. Because the samples remained moist, cold, and sealed from air, we assume there was no loss in ^{131}I from the samples. We were limited in our ability to sample a given site with greater temporal frequency because of limited boat access and other logistical issues.

Kelp tissues were prepared in a manner similar to that of the brown seaweed *Fucus* which was sampled after the Chernobyl release of radiosotopes.[11] Mature canopy blades of *Macrocystis* (n = 3) were dried at 60 °C for 24 h. Dried tissue was milled (Thomas-Wiley Mini-Mill) into a powder (20 mesh) and 2 g of dry weight was placed into a polystyrene counting tube (12 mm diameter × 15 mm; Perkin-Elmer). Samples were counted for γ radiation (Perkin-Elmer Wizard 3) 12−24 h to ensure a counting error <7%. The instrument was normalized to major emission peak of ^{133}Ba (250−500 Kev; peak at 356 Kev) because no ^{131}I was immediately available. This window setting includes the main emission peak for ^{131}I of 364 Kev. Having subsequently obtained ^{131}I toward the end of this study, the counting efficiency using these counting parameters was measured at 36% for ^{131}I. The method limit of detection based on 3.14 times the standard deviation of 7 blanks was 3.6 dpm. The brown seaweed *Fucus* spp. processed in a similar manner showed no loss of ^{131}I from volatilization and we assume that this was the case for *Macrocystis*.[11]

RESULTS AND DISCUSSION

Based on measurements performed by others of ^{131}I in air samples over southern California, concentrations spiked on March 21, 2011 (Figure 2), five days after a major release from

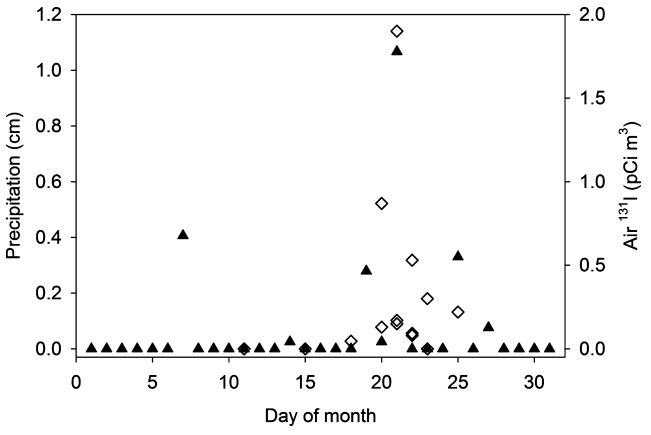

Figure 2. Precipitation (▲) at Newport Harbor, and concentration of ^{131}I (◊) in air measured for the month of March 2011 in Anaheim (both in Orange County, CA.).[7,12]

the Japanese reactor (March 12−16, 2011).[7] This was also a period of significant rain in southern California as shown for Newport Harbor which is the nearest weather station to the Corona del Mar kelp bed (Figure 2).[12] Rainfall and not particulate deposition was the probable vector of ^{131}I transport from the atmosphere to the intertidal brown seaweed *Fucus* during the Chernobyl event, and the same was most likely true for ^{131}I transport from Fukushima even.[11,5] Assimilation most likely occurred shortly after ^{131}I deposition, which we estimated as being March 20−22, 2011 for coastal southern California (Figure 2). This period is consistent with the arrival of ^{35}S in sulfate aerosol from the Japanese reactor to the southern California coastline.[6] In the San Francisco Bay area, preliminary measurements of atmospheric ^{131}I concentrations show a peak occurring March 22−23, 2011 corresponding with precipitation events in the Monterey Bay which includes the sampling sites off Santa Cruz and Pacific Grove.[12,13]

As time progressed, decreasing amounts of radioactivity were detected in all kelp samples repeatedly counted, as shown for kelp collected off Corona del Mar, CA (Figure 3a) and for kelp collected in Lunada Bay, CA (Figure 4). The decay leveled off between 25 and 30 days to approximately 12 dpm or 0.1 Bq gdwt^{-1} for all samples. These low level constant emissions or "residual counts" were significantly above background (no tissue blanks) and presumably came from other radionuclides present in the dried tissue. They were subtracted from the total counts revealing radioactive decay indicative of ^{131}I (Figures 3b, 4). The half-lives calculated from the exponential decay equation derived from exponential regressions were between 7.6 and 8.8 days (e.g., 7.63 days Corona del Mar samples and 8.81 days for the Lunada Bay samples), well within experimental error to the known half-life of 8.04 days for ^{131}I. These results indicated the presence of ^{131}I in the kelp tissues. Samples collected 4 weeks and longer after the initial collection in the southern California showed similar stable low-level counts (12 dpm) indicating no detectable ^{131}I remained and no additional deposition of ^{131}I had occurred.

The radioactive material producing the constant low background gamma emission, or residual counts, in the dried kelp samples is not known. Low levels of gamma emitting radioisotopes have been measured in a variety kelp species: ^{40}K ($t_{1/2}$ = 1.2 × 10^9 y), ^{137}Cs ($t_{1/2}$ = 30 y), and ^{226}Ra ($t_{1/2}$ =1601 y).[14] Only ^{137}Cs and ^{226}Ra may have contributed to the residual counts. Cesium-137, a product of past atmospheric nuclear-bomb tests and nuclear power plant release, has a major gamma emission peak (662 KeV) that is outside the energy window counted in our samples, although some broad low energy emissions occur in the energies measured. Radium-226 ($t_{1/2}$ = 1600 y), a key decay product from the decay of naturally occurring uranium, has several minor emission peaks within the energy window used in our measurements, and is the only radioisotope of the three that was detected in the brown seaweed *Fucus*.[11] Although ^{35}S-sulfate released during the disaster also traveled to the California coast and may have been assimilated by *Macrocystis,* it is a β particle emitter and would not have been detected using our method.

A biological half-life of ^{131}I from *Fucus* was 10.3 days derived from the apparent half-period of 4.5 days and known half-life of radioactive decay.[11] Because of limited boat availability, we did not sample blades from a single location at time intervals which would have allowed for the determination of the biological half-life of ^{131}I in *Macrocystis.* Studies suggest, however, a biological half-life of greater than 10 days. The efflux rate of iodine from *Macrocystis* mature blade tissue was measured at 80 pmol d^{-1} cm^{-2} or 91 μmol d^{-1} L^{-1} tissue water.[9] This efflux rate is a very small fraction of the total iodine content of kelp blades (present almost entirely as iodide at ~50 mM), and is less than 1% the uptake rate of iodide at an environmentally relevant concentration of 100 nM.[8,9] In response to external reactive oxygen species, kelps release stored iodide where it can react with an extracellular haloperoxidase generating diiodomethane

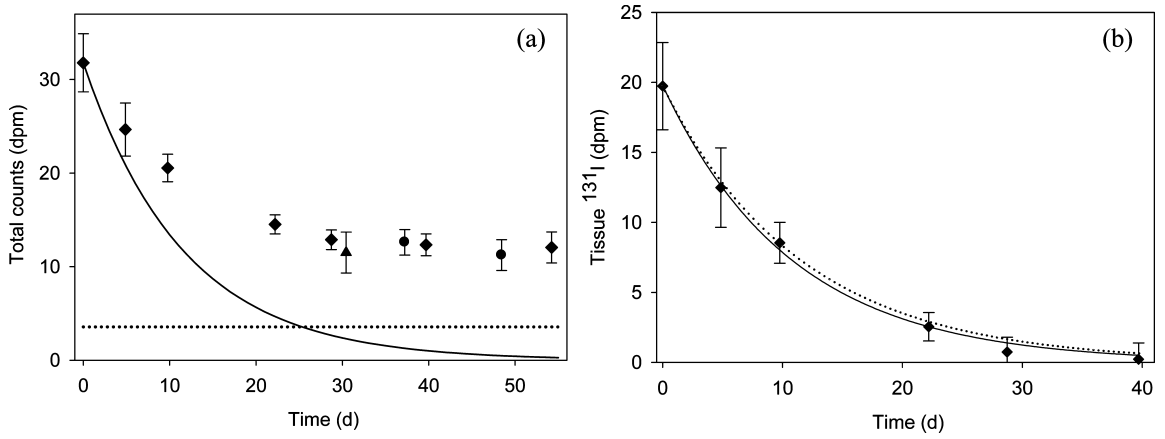

Figure 3. Gamma emission ± sd (n = 3) from kelp collected at Corona del Mar CA. (a) ♦ Collected April 15, 2011 (t = 0), ▲ collected May 19, ● collected May 26. Solid line represents theoretical decay pattern if entirely ^{131}I. Horizontal dotted line shows method detection limit. (b) Gamma emissions, ± sd (n = 3) minus residual activity of 12.1 dpm from kelp collected from Corona del Mar CA on April 15, 2011. Dashed line shows theoretical decay for ^{131}I; solid line shows exponential regression (r^2 = 0.99, p < 0.01).

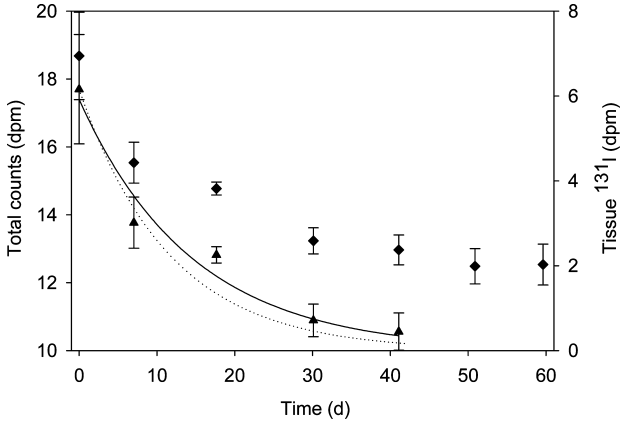

Figure 4. Gamma emission ± sd (n = 3) from kelp collected April 22, 2011 from Lunada Bay CA (♦). Gamma emissions, ± sd minus residual activity of 12.5 dpm (▲). Solid line shows exponential regression (r^2 = 0.97, p < 0.01). Dashed line shows expected decay for tissue ^{131}I.

and mixed iodohalomethanes.[8] Volatilization of ^{131}I may have occurred, however, emission rates of these volatile compounds by kelps are very low in the range of 2−20 ng d^{-1} g^{-1} fresh weight.[15,16]

After subtracting residual counts from the total counts of collected samples, all but those collected from Sitka, AK contained detectable amounts of ^{131}I (Table 1). The ^{131}I distribution pattern in *Macrocystis* is consistent with the projected atmospheric radioactive plume which primarily encountered the central and southern California coast due to the dominant midlatitude westerly winds, with minimal exposure in Sitka, Alaska.[1,6] The late time of sampling in Sitka may have also contributed to ^{131}I not being detected. The specific activities of ^{131}I found in *Macrocystis*, extrapolated to the date of deposition−assimilation (Table 1), are similar in magnitude to those found in the *Fucus* from southwestern British Columbia and northern Washington State (0.23−4.7 Bq gdwt^{-1}) sampled 28 days after the Chernobyl incident of April 26, 1986.[11] *Fucus* sampled from California at that time was below the detection limit. We measured the highest ^{131}I values in the Corona del Mar *Macrocystis* population (Table 1). This particular site directly receives runoff from an urban watershed of 291 km^{-2} via San Diego Creek through Newport Harbor. This runoff may have exposed the neighboring kelp bed to additional ^{131}I deposited from inland precipitation. The other southern California kelp beds would not have received the volume of direct runoff as the Corona del Mar bed. No further input of ^{131}I to the Corona del Mar population was observed at later sampling dates (Figure 2A) indicating that the ^{131}I present came from a single period of exposure to ^{131}I from the Japan release.

These elevated levels of ^{131}I found in *Macrocystis* tissue are well above those reported in other kelps prior to the Fukushima release. The Australian subtidal kelp, *Ecklonia radiate*, was shown to contain low levels of ^{131}I (20−73 mBq g^{-1} fresh weight or 130−490 mBq gdwt^{-1} assuming 15% dry wt), originating from nuclear medicine treatments released from coastal outfalls.[17] Lower values of ^{131}I were measured in kelps along the Japanese coastline prior to the reactor leak: 0.01−0.37 Bq kg^{-1} fresh weight (0.067−2.5 mBq gdwt^{-1} assuming 15% dry wt).[18] In contrast to the levels in our samples, <9 mBq ^{131}I gdwt^{-1} was detected in dried commercially available *Macrocystis integrifolia* blades from coastal British Columbia ten years prior to the Japanese release.[14] It was estimated, however, that seaweed near the Fukushima reactor leak could be as high as 10^8 Bq kg^{-1} fresh weight (or 10^6 Bq gdwt^{-1}).[19]

Macrocystis is the keystone species supporting the kelp forest ecosystem, which is highly productive with complex community structure. Using a canopy density of 3.5 kg fresh weight m^{-2}, the amount of ^{131}I present in California kelp ranged from 0.3 to 1.3 kBq m^{-2} of canopy.[11] Our estimate of the area covered by the Corona del Mar *Macrocystis* canopy was 33 000 m^2 yielding a total of 40 MBq ^{131}I sequestered into this kelp bed canopy. The total amount of ^{131}I taken up by the canopy was most likely larger than that measured in the canopy tissue because iodine taken up by canopy blades is translocated throughout the kelp body as iodide.[9]

The degree to which trophic transfer of ^{131}I to other species occurred is unknown. To our knowledge there have been no studies on the trophic transfer of iodine involving grazers of *Macrocytis*. Laboratory studies of ^{131}I trophic transfer from the red seaweed, *Chondrus crispus*, to the grazer periwinkle, *Littorina littorea*, measured a 0.07 ± 0.02 transfer factor (TF) suggesting that there is no biomagnification in the transfer of ^{131}I from this seaweed to this gastropod grazer.[20] Sea urchins

are important benthic grazers of *Macrocystis* tissue in terms of the amount of biomass consumed, although they feed on primarily detached senescent tissue. By the time *Macrocystis* tissue that had assimilated ^{131}I reached their habitat, it would have disappeared from decay. Grazing crustaceans, gastropods, and fish occur primarily in the midwater and canopy regions and feed on attached tissue oftentimes meristematic. Meristematic regions of *Macrocystis* are supplied with a translocated fluid rich in mannitol, other nutrients, and iodide.[9] Those regions could have been high in assimilated ^{131}I and if ingested may have exposed these organisms to ^{131}I. There could be targeted accumulation of ^{131}I into several fish grazers because their thyroid endocrine system contains iodinated hormones thyroxine and triiodothyronine.[21] Fish possibly affected would be the Opaleye, *Girella nigricans*, and the Halfmoon, *Medialuna californiensis*, which feed on the meristematic apical tissues of *Macrocytis*, and the Senorita, *Oxyjulis californica*, which may incidentally ingest tissue while feeding on encrusting invertebrates.[22]

Although we did not attempt to detect ^{137}Cs in our kelp samples, it may have been present because it was also released in large quantities from the Fukushima plant, and traveled in the atmospheric plume.[1,2] After the Chernobyl reactor release, ^{137}Cs levels were 0.3 Bq g^{-1} fresh weight (2 mBq $gdwt^{-1}$, assuming 15% dry wt) in the Japanese kelp, *Undaria pinnatifida*, which was above baseline levels of <0.1 Bq g^{-1} fresh weight (~0.67 mBq $gdwt^{-1}$).[23] The low baseline levels of ^{137}Cs most likely originated from past atmospheric nuclear-bomb testing. Commercially distributed edible dried kelp of various species, including *M. integrifolia* from British Columbia coast, were analyzed for ^{137}Cs prior to the Fukushima release and found to contain 6−53 mBq $gdwt^{-1}$ (<15 mBq $gdwt^{-1}$ for *M. integrifolia*;).[14] To our knowledge, naturally occurring nonradioactive cesium (^{133}Cs) has not been measured in kelp tissue. Whereas iodine is concentrated in kelp sieve tube sap (0.53−0.59 mg mL^{-1}) as compared to its concentration in seawater (concentration factors 9000×), the concentration of cesium in sieve tube sap (2.2−320 ng L^{-1}) suggests that it may be only slightly concentrated over its concentration in seawater.[24,25] Clearly, studies on Cs assimilation and tissue content in kelps are needed.

Macrocystis and the other ecologically dominant surface canopy-forming kelps such as *Nereocystis leutkeana* are found off the west coast of North America from California to Alaska. *Nereocystis* beds are common in coastal waters north of Pt. Conception, CA overlapping *M. pyrifera* in distribution until replacing *M. pyrifera* approximately north San Francisco. Unlike intertidal seaweeds such as *Fucus,* the *Macrocystis* and *Nereocystis* canopies are positioned to rapidly take up deposited radionuclides independent of tidal levels. These canopy species should be used as regional coastal dosimeters to monitor ^{131}I and possibly other radionuclides (e.g., ^{137}Cs) released from nuclear accidents. The method described in this study is cost-effective, simple, and rapid. The greatest difficulty using this method to monitor coastal exposure is coordinating early extensive sampling after such a disaster. We recommend that a coastal network be established along the west coast of North America for rapid deployment in response to such accidents to sample and count kelp for ^{131}I and other radionuclides. The resulting data would reveal the pattern of plume dispersal and the degree of contamination of the coastal community.

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Notes

The authors declare no competing financial interest.

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