RADIOIODINE IN KELP FROM WESTERN AUSTRALIA

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As part of a program to survey low levels of radioactivity in the marine environment of the southern hemisphere, we have studied the distribution and uptake of ^{131}I found in the subtidal kelp *Ecklonia radiata*, on the west coast of Australia. Concentrations of 5 to 75 fCi/g of ^{131}I exist in this species over a considerable distance along the coast. We have characterized the principal source of the ^{131}I and found a general temporal correlation between the amount of radioiodine discharged from sewer outfalls and its concentration in kelp. Transplant experiments have enabled us to estimate uptake and depuration rates, and our results are consistent with laboratory measurements made by others.

Introduction

This study was a component of the Lawrence Livermore National Laboratory's Marine Sampling Project which was initiated by the "September-79 Event," a putative nuclear explosion off the southeast coast of Africa. The only evidence of radioactivity detected in connection with this event was a very slight indication of ^{131}I in the thyroid glands of sheep slaughtered in southern Australia two months later¹. If a similar event were to occur again in the southern hemisphere, the west

coast of Australia would be an ideal location for monitoring radioactivity advecting eastward in the Indian Ocean.

While visiting the Commonwealth Scientific and Industrial Organization (CSIRO) near Perth in April, 1983, we collected several specimens of *Ecklonia radiata*, a subtidal kelp. We were quite surprised to find ^{131}I in these plants at an average concentration of 24 fCi/g wet weight. The origin of this radionuclide with a half-life of 8.04 d in kelp from an open coast site was a puzzle, as western Australia has no nuclear reactors, much less any nuclear fuel reprocessing plants. We suspected the source of the isotope to be medical use in the local hospital, followed by discharge through the sewer system to the ocean. Inquiries with the Radiation Protection Office at The Queen Elizabeth II Medical Centre in Nedlands, Western Australia, confirmed this was the case. In January 1985, we began an investigation of this interesting occurrence. We have analyzed plants from many locations along the west coast of Australia to determine the geographical extent of the detectable ^{131}I and transplanted plants from clean areas to the principal sewer outfall and back to observe uptake and depuration rates.

Background

Sea water contains about 0.05 ppm of iodine^{2,3,4}, with a variation of 0.01 to 0.07 ppm because of the effects of wave action, temperature variation, and escape of iodine due to the action of iodine oxidase in the presence of oxygen⁵. The chemical forms of iodine are principally iodide and iodate, although methyl iodide has been found in all ocean waters so far examined⁶. Iodide is thermodynamically unstable in oxygenated seawater but⁶ does exist in surface and deep layers. Laboratory experiments have shown that iodide is oxidized to iodate in oxygenated seawater when no organisms are present⁷ and iodate is the predominant species in the surface. In a community of fish and seaweeds, however, both oxidation and reduction

reactions take place⁸ and 20 to 30% of the total iodine may be as iodide, due mainly to reduction of iodate by biological activity.

Seaweeds have long been known as effective accumulators of iodine and are an important commercial resource. In several species iodine is known to be an essential nutrient required for growth^{2,4,9,10}. A reasonable average iodine concentration over several commercial species is 175 ppm, although the range is a factor of 5. Variables affecting the concentration are principally growth rate and harvesting pressure¹¹, but seasonal variations are also considerable, especially in the *Laminaria*¹². The brown algae (Phaeophyta) accumulate iodine to a much higher level than the green (Chlorophyta) or red (Rhodophyta) algae¹³, and members of the order Laminariales (which includes *Ecklonia radiata*) are especially effective concentrators; *Laminaria japonica* from the Sea of Japan has an average dry weight concentration of iodine of 0.56%, three times the average for Rhodophyta and nearly 100 times the average for Chlorophyta³. Depending upon conditions, individual species may have very large concentration factors: 10^4 for *Sargassum* sp., 3×10^4 for *Laminaria digitata*, 10^5 for *Gracilaria corticata* (Rhodophyta)¹⁴. The iodine content varies in different seasons and with locality and also depends on the relative proportion of fronds and stipes and whether the plant is fruiting or sterile⁵.

Concentration factors, and uptake and depuration rates for different forms of iodine have been studied with ¹²⁵I and ¹³¹I. Using carrier-free ¹³¹I, Svetasheva¹⁵ found the uptake rate to be highly dependent on the chemical form. He found iodide to be taken up faster than iodate, but once the iodine was taken up, the depuration rate was independent of the species absorbed. The uptake rate of the excreted form was similar to that of iodide suggesting a biological transformation. He also found the uptake rate to be slower for *Ulva rigida* (Chlorophyta) than for *Cystoseira crinita* (Phaeophyta).

The mechanism of the uptake of several micronutrients in *Macrocystis pyrifera* was studied by Manly¹⁶, who found that there was an initial rapid uptake into free space, followed by a slower rate due to cellular uptake. For iodide the gross uptake rate decreased and approached the cellular uptake rate within 30 min because of the small free space capacity for iodide. He also found that anions exchange more rapidly than cations between free space and the external solution. For all frond lengths in *Macrocystis pyrifera*, the iodide uptake was greater than the assimilation, suggesting transport to other tissues for storage or release. This translocation of iodine was observed by Amat and Srivastava¹⁷ in *Laminaria saccharina*. When they placed ¹²⁵I on a distal mature part of a blade, it was translocated unidirectionally toward the meristematic region at the blade-stipe junction with a velocity of 2 to 3.5 cm/hr. Analysis also showed much higher levels of stable iodine in the stipe and holdfast than in the blade. On a cellular scale, Pedersen and Roomans⁶ found marked differences in localization of iodine in three species of *Laminaria*.

A project similar to ours studied the uptake of ¹³¹I by several species of seaweed near a sewer outfall in Bracclet Bay, Swansea, U.K.¹⁸ Concentration factors of up to 10⁴ were measured for several species including *Fucus*, *Ulva*, and *Porphyra*. *Fucus* had the highest concentration factor, *Ulva* the lowest. The highest concentration of ¹³¹I found in *Porphyra*, a local food source, was 446 pCi/kg, but samples were highly variable and some showed no radioactivity at all, even though they were nearer the outfall than some of the higher ones.

Fig 1 is a diagram of an *Ecklonia radiata* plant growing in the rough water typical of most of our experimental sites. Unlike terrestrial plants that grow from their tips, *Ecklonia* grows from a meristem situated at the junction of stipe and blade. New plant material is added at the base of the plant and moves up in "conveyor belt" fashion to the top and sides of the plant, where it erodes away¹⁹. A small hole punched in the meristem about 10 cm from the base is observed to move

upward and has proved useful as a measure of growth in length. Estimates of total biomass production are difficult, however, because of uncertainties in the rate of erosion at the tips, respiration, and exudation and dissolution of organic material. The growth rate is dependent on water temperature, available light, and nutrient supply. Erosion rate depends mostly on wave action and can be very rapid during storms.

Estimate of ^{131}I Output

Fig 2 shows the general geography in the region of our experiments, the locations of the areas we sampled, and the four sewer outfalls that are the sources of the ^{131}I . We knew that the Swanbourne outfall was the principal source, as all patients receiving ^{131}I therapy were treated in the Nedlands Medical Centre and excreted most of their ^{131}I into the Subiaco and Swanbourne sewer systems, both of which discharge through the same outfall. However, some patients were treated on an outpatient basis and excreted approximately half of their doses to the Cape Peron, Point Peron, and Beenyup outfalls. In order to estimate the ^{131}I output at the various outfalls, several assumptions, based on engineering calculations, were necessary. The estimate was simplest for inpatients, who excreted 100% of the administered dose in the time they were hospitalized. The hospital retention tanks are emptied into the main sewer system at known times, and the ^{131}I arrives at the Swanbourne outfall approximately two days later. Outpatients who reside within the metropolitan Perth area were assumed to have excreted 50% of their doses into the Subiaco system over two days' time, with another two days allowed for transport to the outfall. Patients who resided outside the metropolitan area were assumed to have stayed in the Perth region overnight, and to have excreted 25% of their doses into the Subiaco system. The remainder of their doses were excreted into one of the other three systems. The activity at a particular outfall then was the sum of all doses

multiplied by the fractions excreted and the radioactive decay factor from the time of the dose to arrival at the outfall.

While there are certainly many simplifying assumptions included in this model, the dose amounts and times are accurately known, as are the home locations of outpatients. Thus the ^{131}I input data are accurate, and the assumptions are based on engineering calculations and measurements of volumes and flow rates in the various systems. Furthermore, the 8-d half-life of the ^{131}I alleviates any effects of long-term accumulation or unknown holdups. The major uncertainties lie in the assumptions regarding the Woodman Point and Beenyup outputs.

Sampling and Analysis

All of our underwater work was conducted using SCUBA. We constructed underwater frames to hold plants at the Swanbourne sewer outfall and at the Pamela Shoals and Atlantis depuration sites. Plants were transported by open boat but covered with burlap kept wet with sea water to protect them from direct sunlight and dessication. Survivability of transplants was about 90% using this technique. All of the plants transplanted to Swanbourne had identification tags attached and were punched as described above to measure growth rates. As soon as plants were harvested for analysis, they were sealed individually in polyethylene bags and shipped frozen in dry ice to Livermore.

After thawing and draining, plants were packed wet into individual containers for gamma spectroscopic analysis. Most plants were too large to analyze entirely and required trimming to fit the containers. A few preliminary experiments could detect no differences in ^{131}I concentration from various parts of a single plant but, for consistency, plants were always placed in the containers stipe-first and trimmed off the top to fit. Counting weights were typically 200 g. Counting periods were usually 1 wk on any of several Ge-Li detectors, including our high-sensitivity

low-background Compton-suppressed detector which has been discussed²⁰. Spectra were analyzed with the GAMANAL computer program²¹.

Experiments at the Swanbourne Outfall

We transplanted 40 plants from a site we assumed would be free of ^{131}I (Control Site) on 1/19/85 to a frame near the Swanbourne outfall. The Control Site is an exposed reef well offshore and we were surprised to see detectable levels of ^{131}I (confirmed by a second sampling showing even higher concentrations 10 mo later) in these plants. These low starting levels did not present a problem, as after growing 3 mo on the frame at Swanbourne the ^{131}I concentration had increased five-fold to an average of 51.2 ± 19 fCi/g wet weight. We sampled these plants periodically, replacing plants as necessary to maintain an adequate population on the frame, and the results are plotted in Fig 3 along with the 16-d moving average of the estimated output of ^{131}I . The 16-d moving average was selected to smooth the periodic inputs of ^{131}I , which range from 0 to 200 MBq/d (in one instance, 1200 MBq in a day). On the basis of radioactive decay alone, radioiodine released more than 16 d prior to the sampling date would contribute less than 25% of the total detected. Further removal from the frame site would also occur because of dilution and dispersion. As can be seen from Fig 3, there is a definite association of high and low ^{131}I concentration in kelp with high and low inputs at Swanbourne. The association is quite strong, considering the assumptions made in calculating the output at Swanbourne, the largely unknown effects of local currents, and the variable growth and erosion rates of the kelp over the course of the year. The high point on day 144 (1985) may be due to known inputs from Woodman Point carried northward by the current. The points at days 293 (1985), and 18 and 159 (1986), appear to be low, but all are on the leading edges of input peaks and well within the uncertainty of the input estimates.

We made one attempt to determine ^{131}I concentrations in other species of algae. On the day 201 (1985) collection at Swanbourne, when the average ^{131}I concentration in *Ecklonia* was 34.3 fCi/g, we collected samples of *Ulva* and *Cualerpa*, both Chlorophyta, and a sample of an undetermined species of Rhodophyta. These were all indigenous species growing on the outlet pipe. ^{131}I was not detectable in either species of Chlorophyta, and was barely above detection limits at 4.4 fCi/g in the Rhodophyta. This is in general agreement with the findings of others that the brown algae are more effective concentrators of iodine than are the green or red.

Uptake and Depuration Rates

We attempted to measure the ^{131}I uptake rate in *Ecklonia* by transplanting plants from the control site to the frame at Swanbourne, followed by frequent sampling to observe the increase in ^{131}I concentration with time. Table 1 shows the average activity at day 0 (fresh from the Control Site), and after 7, 16, and 29 d at Swanbourne. In Fig 3, these are the points at days 327, 336, and 349. We had little success with this experiment as the Control Site plants that previously had contained 10 fCi/g or less on 1/19/85 had increased to 30 fCi/g, while the Swanbourne output had been low for several weeks prior to the transplant. However, since the plants contained 30 fCi/g when transplanted and 28 fCi/g 7 d later, they had to have taken up an amount of ^{131}I approximately equal to that lost by radioactive decay. We feel that the 16-d and 29-d samples represent effects of the variable concentration at the Swanbourne outfall, rather than uptake or loss by the plants. We conclude from this experiment that the uptake rate observed under oceanic conditions is comparable to, or faster than, the radioactive decay rate. This is in agreement with laboratory studies indicating uptake is very rapid and may be complete within a few hours.

We thought that our first attempt to measure a depuration rate by transplanting labeled plants to a clean area and sampling them over time would be straightforward, since we would not have to contend with variable concentrations at the clean site. Seeking to avoid the Control Site and the problems it had already caused us, we selected Pamela Shoals, farther north and more exposed, as the depuration site. We were therefore quite surprised when our transplants from Swanbourne, labeled at a concentration of 20 ± 4 fCi/g, had increased after 7 d at Pamela Shoals to 43.2 ± 5 fCi/g. Seven days later, the concentration had fallen to 26 ± 8 fCi/g, but even after 35 d at Pamela Shoals the concentration was still 19.7 ± 1 fCi/g. Indigenous plants sampled at this time contained 8.5 ± 2 pCi/g. Clearly this was not the clean site we had expected, but we do not know whether the ^{131}I was due mainly to inputs from Beenyup or to transport from Swanbourne, as the usual northward currents were particularly strong during this experiment. At least the results confirmed our previous finding that the uptake rate is comparable to, or faster than, the decay rate.

Our second attempt to measure the depuration rate was more successful. We transplanted plants from the Swanbourne frame to Atlantis, a site north of Two Rocks, after determining that plants from there did not contain detectable ^{131}I . After correcting for radioactive decay, we found that the plants had lost about 30% of their ^{131}I in the first 3 d (the time of the first sample collection) and essentially none over the next 12 d. This indicates that a substantial fraction of the iodine absorbed is incorporated in the cells or stored in the deeper layers of the plant, as described by Manly¹⁶ and Amat and Srivastava¹⁷ for other species. Although this experiment indicates an initial depuration-rate half-life of approximately 6 d, we have no data earlier than three days, and the true initial depuration rate could easily be much faster.

Geographical Distribution of ^{131}I

While the uptake and depuration studies were being conducted, many opportunities arose for collecting indigenous samples from a variety of locations in order to determine the extent of detectable ^{131}I contamination, and Table 2 presents the results for those sites where ^{131}I was detected. Plants from Two Rocks and from two other locations farther north never contained detectable ^{131}I ($<4 \text{ fCi/g}$), nor did plants from Rottnest Island or Pt. Peron. We think the ^{131}I in samples from Pamela Shoals, First Site, and Control Site comes from a combination of longshore transport from Swanbourne and periodic inputs from Beenyup.

Discussion and Conclusions

As is the case with most environmental studies, this one was subject to a variety of unalterable constraints and to variables whose effects were largely unknown. The most serious constraints were the 8 d half-life of ^{131}I , the time required from sample collection to analysis, the low concentrations being measured, and the number of samples that could be analyzed for each experiment. Typically, the time between sample collection and final data analysis was two weeks, one week in shipment and preparation, and another week in counting and data reduction. Furthermore, counter availability limited us to 6 to 8 samples for each experiment. These constraints, combined with the variable output at Swanbourne, sporadic outputs from Woodman Point and Beenyup, and changing coastal currents made it useless to wait for the results of one sampling before beginning another experiment. By the time results were determined, the conditions at the experimental sites had most certainly changed. Although we received excellent cooperation from the Medical Centre's Radiation Protection Board regarding the amount of ^{131}I released from Swanbourne (and the other outfalls), they were never able to supply us with predictions of time and level of future releases as the number of patients to be

treated and the doses to be administered were not predictable by the hospital. These factors, combined with a desire to minimize underwater time and the expense and discomfort of open boat travel, led us to select the Control Site and Pamela Shoals as experimental areas without surveying them first. They were within reasonable distance from Swanbourne and similar to open coastal sites we had determined to be free of ^{131}I . We had learned our lesson by the time of the second depuration experiment, and took the time and trouble to analyze plants from the Atlantis Site before beginning that study.

We know of several important variables affecting our results, but we do not know what these effects are. We know the current generally flows northward along the coast driven by westerly winds, but with variable speed depending on wind strength. The plants grow and erode at variable rates as discussed above, and also may shed large amounts of mucous outer covering from time to time. Finally, there are the periodic inputs of ^{131}I from the Woodman Point and Beenyup treatment plants that mix into the coastal inventory in unknown ways.

In spite of the difficulties, we have been able to draw some conclusions:

1) As far as we know, this is the first study to detect low levels of ^{131}I in the coastal waters of Western Australia or in Ecklonia radiata.

2) The Swanbourne sewer outfall is the principal source of the ^{131}I we see in the kelp, but periodic inputs from Woodman Point and Beenyup treatment plants also contribute.

3) There is a general correlation between ^{131}I concentrations seen in kelp growing near the Swanbourne outlet and the output as estimated from the Radiation Protection Board data.

4) The ^{131}I uptake rate is at least comparable to the ^{131}I half-life, probably much faster. The indicated initial depuration rate (half-life approximately

6 d) could also be much faster. Both rates are in general agreement with laboratory studies.

5) The ^{131}I appears to be distributed uniformly throughout the plant, at least within our ability to detect differences at these low concentrations. While others have found marked localization of stable or long-lived radioiodine^{6,17}, the 8-d half-life of ^{131}I does not permit this type of accumulation from the highly variable source at Swanbourne.

6) There is an unusually low variance of ^{131}I concentrations within each sample set. This is illustrated in Tables 1 and 2 for plants containing rather low levels of ^{131}I , but was even more evident in the plants exposed at Swanbourne. A set of 6 or 7 samples would often have an ^{131}I concentration standard deviation of 10% or less, and it rarely exceeded 25%, especially for concentrations above 20 fCi/g. Although an occasional plant from a set would be below detection limits, as they were in the Bracelet Bay study¹⁸, the remainder would agree very closely, a result seldom encountered in our other experiences in environmental monitoring. It may be that (1) the ^{131}I is radiochemically exchanged with the stable iodine in sea water and is thus not subject to the vagaries of tracer concentrations, or (2) the short biological exchange times and half-life of ^{131}I may prevent the more efficient plants from accumulating significantly higher concentrations in their deeper layers. The concentrations we measured may be representative of only the previous few hours' to a day's exposure.

7) There is no evidence of any substantial amounts of other artificial radionuclides; however, we did observe ^{137}Cs in a few samples at or slightly above our detection limit of 3 fCi/g.

8) It can be very difficult to predict where low levels of environmental contamination will occur. In our coastal survey, we detected ^{131}I in what appeared to be very unlikely locations, and we found plants from similar locations to have

quite different concentrations. Temporal variations no doubt account for much of this variability.

9) *Ecklonia radiata* can be transplanted successfully if care in handling is observed.

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Table 1
Uptake Rate Measurement

Time at Swanbourne (d)	^{131}I (fCi/g)
0 (Control Site)	30.6 ± 4
7	28.4 ± 5
16	10.4 ± 2
29	20.3 ± 5

Table 2
 ^{131}I Concentration in Indigenous Plants*
 (fCi/g wet weight, average of two or more plants $\pm$ 1 S.D.)

Date	Location		
	Control Site	First Site	Pamela Shoals
4/1/83		24.0 $\pm$ 2	
1/19/85	9.9 $\pm$ 1		
5/11/85		7.3 $\pm$ 3	
11/16/85	30.6 $\pm$ 4		
3/2/86			8.4 $\pm$ 2
5/12/86		15.5 $\pm$ 1	
6/8/86	<4		

*No ^{131}I was ever detected in plants from Two Rocks and points north, Rottneest Is., or Pt. Peron.

Figure Captions

Fig. 1. *Ecklonia radiata* growing in rough water.

Fig. 2. Coastal geography in the vicinity of Perth,
Australia, showing the locations of our sampling
sites and the sewer outfalls that are the sources
of the ^{131}I .

Fig. 3. ^{131}I discharges from Swanbourne (and Woodman
Pt.) and concentrations in *Ecklonia radiata*.