AN EVALUATION OF HAZARDS FROM IMMERSION OF PLUTONIUM IN THE MARINE ENVIRONMENT

J. D. TERESI*
U.S. Naval Radiological Defense Laboratory, San Francisco, California 94135

and

C. L. NEWCOMBE
San Francisco State College, San Francisco, California

Abstract—The potential hazard due to a single immersion and subsequent release of plutonium was evaluated on the basis of its possible incorporation in the marine food chains including the alga, *Porphyra*, and the seaweed products agar and alginic acid. The hazards were estimated by comparing the recommended permissible body burdens for large populations (1/10th occupational permissible body burden) with the expected body burden calculated on the basis of very conservative assumptions. The concentration of plutonium from a single release in the sea that would lead to the permissible body burden through the fish food chain was calculated on the basis of published concentration factors. The value of the concentration of plutonium to give this body burden was found to be 6.4×10^{-2} μCi/ml assuming a single intake of 200 gms of fish per individual. The expected body burdens from *Porphyra*, agar, and alginic acid were then calculated on the basis of the calculated plutonium concentration and the published concentration factor for seaweed. Calculations were also made of various diffusion parameters using six published diffusion models to obtain areas of contamination with time and the duration of a hazardous concentration.

INTRODUCTION

The objective of this study was to evaluate the potential hazards from the contamination of the California inshore ocean environment with plutonium following an instantaneous release upon immersion. The analysis considered the ocean environment at those distances from shore at which the ocean depth was about 10 m. At depths greater than this, one would expect the extent of the hazard to be less than that calculated for the 10 m case.

DESCRIPTION OF SOURCES OF HAZARD

Hazards from the existence of plutonium in the marine environment depend upon the physical and chemical properties of its compounds. Equally important are the relative amounts of contaminant in the several parts of the physicochemical and biological environments that may represent avenues of transport of the radioactivity to man.

Released plutonium in direct contact with water, or with naturally occurring suspended particulates of sea water (colloidal particles, sediments, and microscopic organisms), or with sea-bottom materials may result in a wide variety of interactions, most of which are poorly understood. Attention here is limited to those interactions that directly or indirectly affect the availability of plutonium to man.

In California, principal commercial sources of chemically processed marine algae are the phycocolloids agar and algin. Agar is a gelatinous substance produced on the Pacific Coast of the United States from the red alga, *Gelidium cartilagineum*. It is used by the baking industry, in icings, as a stabilizer in pies, as a moisture

* The opinions or assertions contained in this article are the private ones of the writer and are not to be considered as official or reflecting the views of the Navy Department or the Naval Service at large.

regulator in fruit cakes, in cream cheese, in jelly desserts and in puddings. Little is known about the extent to which chemical compounds of plutonium may be adsorbed or absorbed by *Gelidium*.

Algin is a term referring to the polyuronic acid, alginic acid $(C_6H_8O_6)_n$ and its derivatives. Algin occurs in large quantities in many seaweeds. Sodium alginate solutions may be transposed by solutions of salts of a large number of metals, including uranium; e.g. $U(SO_4)_2 \cdot 4H_2O$, to form insoluble alginates.

A dozen or more of the larger algae along the coast of California and lower California, such as the giant kelp, *Macrocystis pyrifera*, are used for the manufacture of algin. Annually, thousands of tons are collected south of Point Conception. The principal uses of algin are in pharmaceuticals, dairy products, soda-fountain drinks, cosmetics, drugs, salad dressings, candy, jelly, and chocolate milk. Algin is also used in stock feed and chicken feed. Although little is known of the extent of uptake of radionuclides by kelp, the potential plutonium body burden in man was estimated by using gross assumptions and available data (see hazard estimates below).

Edible algae, although poor sources of energy because of their indigestibility, possess an important vitamin and mineral content. Irish moss is harvested on the Atlantic Coast in considerable quantities but the amount of commercial utilization for food on the Pacific Coast of the United States is small. *Porphyra* is the only alga utilized directly for food in any quantity in California. In 1929 about 300,000 lb were harvested in Central and Southern California by American Chinese for distribution to Chinese stores or for exporting to the Orient. The potential hazard to the California Chinese is estimated in the section on estimate of the hazards.

Local spawning areas of pelagic fishes near the coast of California and centers of commercial fishing and sportfishing in shallow waters may, due to accidental situations, become centers for the origination of radioactive contamination that reaches man. The release of plutonium in a partially enclosed or restricted local area, as projected in this report, could result in a small-scale dispersal of the plutonium and in a correspondingly localized contamination of the food web of a local area. The scale of the resultant hazard from plutonium would depend in large part on the solubility of the radionuclide in sea water, the rate of diffusion, and the extent of uptake and transport of the contaminant through the food web to man. The scarcity of experimental data necessitates the use of conservative assumptions to make adequate estimates of the likely presence or absence of hazards due to plutonium in water.

ESTIMATE OF THE HAZARDS

The potential hazard due to immersion in seawater and subsequent release of the radionuclide is evaluated on the basis of its possible incorporation in the marine food chains including the alga *Porphyra* and the seaweed products agar and alginic acid.

The hazards from these various sources are estimated by comparing the recommended permissible body-burden for large populations (1/10th the occupational permissible body burden) with the expected body burden calculated on the basis of conservative assumptions.

Estimation of the Hazard Due to the Ingestion of Plutonium Through the Fish Food Chain

Desired information for a detailed description of the hazard has proved, in most instances, to be lacking. However, for present purposes, hazards to man from projected releases of plutonium can be satisfactorily evaluated by utilizing established knowledge of the marine environment and by resorting to necessary assumptions customarily utilized in analytical techniques for the evaluation of hazards. Many of the customarily used mathematical models and useful dosage concepts regarding levels of absorption and tolerance assume an organ-organ system equilibrium and/or an organism-environment equilibrium. Because of the relatively rapid dispersion of radioactivity (see the Appendix) throughout the environment and the consequent decrease with time of the concentration levels, the organism-environment equilibrium referred to above is in fact not likely to be reached in the situations under consideration. Thus the cylindrical-dispersion model is very conservative. Also, due to the behavior patterns of pelagic fish—their schooling habits and mi-

gratory movements—the probability of their remaining in a seriously contaminated area long enough to permit an equilibrium state is expected to be low. Hence, the likelihood of netting a sizeable number of contaminated fish is low. Furthermore, the total commercial catches per boat are proportionately large and, to this extent, the likelihood that a processing plant will turn out consecutive batches of this hypothetically contaminated product is correspondingly remote. Therefore it is reasonable to anticipate a single exposure by the consumer rather than a continuous exposure.

The maximum permissible body burden (MPBB) for the general population is usually assumed to be 1/10 that of the occupational value. In the case of ^{239}Pu the value chosen for the general population is, therefore, 0.004 μCi.

The body burden can be calculated in the following manner (neglecting decay and biological elimination):

$$q = f_w \cdot f_c \cdot I \cdot S \qquad (1)$$

where q = body burden in microcuries,
f_w = retention factor (fraction of total ingested that is retained in the body),
f_c = concentration factor (total concentration through all fish food chains),
I = food intake (g),
S = plutonium concentration in seawater (μCi/cm^3).

The value of f_w used here [1] is 2.4×10^{-5}. The value for f_c is 1 according to Dunster [2] and 13 according to Aten. [3]

The average daily intake of fish food has been estimated to be about 200 g, [4] assuming all protein food is obtained from marine sources.

If q is set at the maximum permissible value of 0.004 μCi and f_c is assumed to be 13, then the value of S will be 6.4×10^{-2} μCi/cm^3. The following expression for S results if we assume an instantaneous plutonium release (C), in curies, with immediate mixing throughout a cylinder of water of depth (D), in meters, and a surface area given by (A). (Other diffusion models are treated in the Appendix.)

$$S = \frac{C}{AD} \ (\mu\text{Ci/cm}^3) \qquad (2)$$

where A = area (m^2).

Substituting for S the value of the concentration that would result in the maximum permissible body burden and 10 for D and transposing, equation 3 results:

$$A = \frac{C}{10 \times 6.4 \times 10^{-2}} = 1.56\,C \ (\text{m}^2). \qquad (3)$$

For any given total release of plutonium the area of the sea containing the concentration 6.4×10^{-2} μCi/cm^3 can be calculated. The radius of the equivalent circular area can then be determined. Examples of contaminated areas for various total amounts of plutonium released to a depth of 10 m are shown in Table 1.

Table 1. *Radii* (r) *of Cross-sectional Areas* (A) *of Contaminated Cylindrical Volumes as a Function of Total Plutonium* (C) *Released to a Depth of* 10 *meters*

C (curies)	A (meters^2)	r (meters)
10^2	1.56×10^2	7
10^3	1.56×10^3	22
10^4	1.56×10^4	70
10^5	1.56×10^5	220
10^6	1.56×10^6	700

The sample results shown in Table 1 can be used to indicate the volume and the areas of offshore water involved under assumptions of instantaneous releases of plutonium in amounts of 10^2 to 10^6 Ci. The calculations based on a concentration factor of 13, indicate that 200 g of fish obtained from these areas during a short period of time while the plutonium concentration is at the level of 6.4×10^{-2} μCi/cm^3 would have to be eaten by each individual of a large population to result in the general population MPBB of 0.004 μCi. This type of estimate of the hazard is a conservative one since it implies that fish concentrate plutonium to the equilibrium value during their stay in the body of water involved. It also assumes that a large population will obtain enough to supply it with food for one day's consumption of 200 g per individual.

Estimation of the Hazard Resulting from the Ingestion of Agar

The value of the plutonium body burden from ingestion of agar processed from seaweed is a function of the concentration factor for seaweed, the yield of agar, and the amount of plutonium carried with the agar.

Agar is prepared from a filtrate obtained from a hot-water treatment under pressure and therefore would probably not contain plutonium. Since there are no data available on this point, it has been assumed that the plutonium of the seaweed will follow the agar in a proportional manner.

According to Tseng,[5] the 650,000 lb of agar used annually in the United States were distributed approximately as shown in Table 2. As can be seen, only about $\frac{1}{2}$ of the total is ingested. Of this total amount used, only 52,000 lb, or 8%, is produced in the United States (Southern California) and, therefore, the amount of the Southern California produced agar ingested is 26,000 lb per year.

Table 2. Annual Distribution of Agar in the United States

Utilization	Amount (lb)
Laxatives	100,000
Microbiological culture	100,000
Bakery	100,000
Confectionery	100,000
Dental impression mold	75,000
Meat packing	50,000
Emulsifier	50,000
Cosmetics	25,000
Miscellaneous	50,000

The total area from which this much agar is produced constitutes about 100 mi^2[6] (2.59×10^8 m^2). As shown in Table 1, the area of concern relative to a plutonium contamination of 6.4×10^{-2} $\mu Ci/cm^2$ is 1.56 m^2 per curie of plutonium released for a cylindrical instantaneous dispersion model. Therefore, the amount of plutonium which could just contaminate this 100/mi^2 area to the above concentration is 1.66×10^8 Ci, and thus the amount of contaminated agar that would be ingested is about 7.1×10^{-2} g/Ci of released plutonium.

If we assume a population of about 10^8 then the *per capita* ingestion of the contaminated agar would be 7.1×10^{-10} g/Ci released. The concentration factor for seaweed is assumed to be 500, which is only a guess given by Dunster,[2] probably based on soluble plutonium compounds.

To calculate the body burden from agar ingestion the following expression is used:

$$q = I_{ag} S f_c f_w$$

where:

q = body burden (μCi),
I_{ag} = intake of agar (g),
S = plutonium concentration in seawater ($\mu Ci/cm^3$),
f_c = concentration factor for seaweed,
f_w = body retention factor.

Substituting 500 for f_c, 2.4×10^{-5} for f_w, 6.4×10^{-2} for S and 7.1×10^{-10}/Ci for I_{ag}, $q = 5.5 \times 10^{-13}$ μCi/Ci of released plutonium.

Thus, on the basis of the conservative assumptions used in the calculations, a release of about 7×10^9 Ci of plutonium would be required to result in an average body burden due to agar ingestion equal to the MPBB of 0.004 μCi.

Estimation of the Hazard Resulting from the Ingestion of Algin

The value of the plutonium body burden from ingestion of algin from seaweed is a function of the concentration factor for seaweed, the yield of algin, the plutonium content of seawater, the amount of plutonium carried with the algin, and the body retention factor for plutonium. According to Tseng,[8] fresh kelp, *Macrocystis pyrifera*, is digested with soda, which reacts with alginic acid in the kelp to form soluble sodium alginate. After filtration, the filtrate is acidified, precipitating the alginic acid.

Since alginic acid is prepared by an alkaline treatment of kelp, the plutonium would probably be in an insoluble form and, therefore, the amount of plutonium contamination in the alginic acid should be negligible. However, since the amount of contamination actually expected is not known, the calculation of the body burden is based on the assumption that plutonium

is not precipitated out in the production and purification of alginic acid.

According to Tseng, [9] the annual production of algin in the United States is estimated at two to three million pounds. More than 70% of the total algin produced comes from Southern California.

As in the case of agar, the total area from which this much algin is produced constitutes about 100 mi^2 (2.59×10^8 m^2). The amount of contaminated algin that would be ingested assuming a 3-million-pound total production and an area of concern, shown in Table 1, of 1.56 m^2 per curie released, relative to a seawater plutonium concentration of 6.4×10^{-2}, is 1.8×10^{-2} lb/Ci released (8.2 g/Ci released). Based on a population of 10^8, the *per capita* ingestion of contaminated algin would be 8.2×10^{-8} g/Ci released.

The following expression is used to calculate the body burden expected from ingestion of algin:

$$q = I_{al} \cdot S \cdot f_e \cdot f_w$$

where

I_{al} = intake of algin (g).

After substituting the values of S, f_e, f_w as above for the agar calculations and the value of 8.2×10^{-8} g/Ci released for I_{al},

$q = 6.3 \times 10^{-11}$ μCi/Ci of released plutonium.

Thus, on the basis of the conservative assumptions used in the calculations, a release of about 6.3×10^7 Ci of plutonium would be required to result in a body burden due to algin ingestion equal to the MPBB of 0.004 μCi.

Estimation of the Hazard Resulting from the Ingestion of Porphyra

About 300,000 lb of *Porphyra* [8] are harvested annually from an area of about 100 mi^2. Since the contaminated area as shown above would be about 1.56 m^2 per curie of plutonium released, the amount of *Porphyra* harvested from the area of possible contamination would be 1.8×10^{-3} lb/Ci released (0.82 g/Ci released).

If one assumes that this amount is distributed to the 94,000 California Chinese only, then the *per capita* ingestion of contaminated alga would be 8.7×10^{-6} g/Ci released.

The body burden can be calculated from the following equation:

$$q = I_p \cdot S \cdot f_e \cdot f_w$$

where I_p = intake of *Porphyra* (g).

By substituting the values S, f_e, f_w given above and the value of 8.7×10^{-6} g/Ci of released plutonium for I_p, the value for the body burden q per curie of plutonium released is 6.7×10^{-9} μCi.

It is seen then that a release of about 6×10^5 Ci of plutonium would be required to result

Table 3. Summary of Calculated Body Burdens obtained for Values of Parameters shown

Concentration factor fish food web		13*
Concentration in seawater to give body burden of 0.004 μCi (fish food)		6.4×10^{-2} μCi/cm^3
Area of 10 m depth containing 6.4×10^{-2} μCi/cm^3		1.56 m^2/Ci released
Calculated body burden from ingestion of:		
	Agar	5.5×10^{-13} μCi/Ci released
	Algin	6.3×10^{-11} μCi/Ci released
	Porphyra†	6.7×10^{-9} μCi/Ci released

* This is a conservative factor since it was obtained with soluble nuclide under equilibrium conditions. The actual exposure time of the fish food web is a matter of minutes or, at most, a few hours (see Appendix).

† The evaluation of the body burden from *Porphyra* is conservative since it is based on the assumption that the *Porpyhra* harvested is consumed by 94,000 Californians.

in a body burden of 0.004 μCi in 94,000 Chinese due to the ingestion of *Porphyra.*

SUMMARY

Table 3 presents a summary of information used and values obtained in the analyses presented above.

APPENDIX

Calculation of Dispersal and Concentration of Substances Released into the Ocean

Equations to describe the change with time of the concentration of a diffusing substance have been described by Okubo.[9] These are presented below and may be applied to the release of plutonium.

1. *Joseph and Sendner Solution:*

$$\frac{\partial S}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(Pr^2\frac{\partial S}{\partial r}\right) \qquad \text{(A1)}$$

From this was obtained

$$S(r, t) = \frac{nM}{2\pi D(Pt)^2}\, e^{-\frac{r}{Pt}} \qquad \text{(A2)}$$

where $n = 360°/\theta_n$

θ_n = the angle of the sector to which diffusion is restricted (180° for uniform coastline and 360° for open ocean),

S = concentration in Ci/m^3,

r = distance from origin in meters,

D = thickness of the contaminated layer in meters,

P = "diffusion velocity"* (m/hr) = 54 m/hr,

M = amount of radioactivity released in curies.

The maximum concentration is at the center of the diffusing volume and is given by

$$S_o(t) = \frac{nM}{2\pi D(Pt)^2} \qquad \text{(A3)}$$

2. *Modified Ozmidov Solution:*

$$S(r, t) = \frac{M}{6\pi\gamma^3 t^3 D}\, e^{-\frac{r^{2/3}}{\gamma t}} \qquad \text{(A4)}$$

where γ = "energy-dissipation parameter" (m$^{2/3}$/hr)
$= 1.17$ m$^{2/3}$/hr

$$S_o(t) = \frac{M}{6\pi\gamma^3 t^3 D} \qquad \text{(A5)}$$

3. *Modified Okubo and Pritchard Solution:*

$$S(r, t) = \frac{M}{\pi W^2 t^2 D}\, e^{-\frac{r^2}{W^2 t^2}} \qquad \text{(A6)}$$

where W = "diffusion velocity" (m/hr)
$= 86.4$ m/hr

$$S_o(t) = \frac{M}{\pi W^2 t^2 D} \qquad \text{(A7)}$$

4. *Okubo Solution:*

$$S(r, t) = \frac{4M}{3\pi^{3/2}\bar{\alpha}^3 t^3 D}\, e^{-\frac{r^{4/3}}{\bar{\alpha}^2 t^2}} \qquad \text{(A8)}$$

where $\bar{\alpha}$ = "energy-dissipation parameter" in m$^{2/3}$/hr
$= 1.67$ m$^{2/3}$/hr

$$S_o(t) = \frac{4M}{3\pi^{3/2}\bar{\alpha}^3 t^3 D} \qquad \text{(A9)}$$

5. *Obukhov Solution:*

$$S(r, t) = \frac{M}{\pi\beta^3 t^3 D}\, e^{-\frac{r^2}{\beta^3 t^3}} \qquad \text{(A10)}$$

where β = "energy-dissipation parameter" (m$^{2/3}$/hr)
$= 2$m$^{2/3}$/hr

$$S_o(t) = \frac{M}{\pi\beta^3 t^3 D} \qquad \text{(A11)}$$

6. *Schonfeld Solution:*

$$S(r, t) = \frac{M\omega t}{2\pi D(\omega^2 t^2 + r^2)^{3/2}} \qquad \text{(A12)}$$

* The values for "diffusion velocity" and "energy dissipation parameter" used in the various solutions were taken from a table of data on diffusion of radioactivity reported in ref. 9.

where ω = the "diffusion velocity" (m/hr) = 68.4 m/hr

$$S_0(t) = \frac{M}{2\pi D\omega^2 t^2} \quad \text{(A13)}$$

Expressions for the time, t_m, required to reach the maximum distance, r_m, for a given concentration, S, were obtained by differentiation with respect to t and setting dr/dt equal to zero in each of the diffusion equations. The resulting expressions are given below.

1. *Joseph and Sendner Solution* ($n=1$):

$$t_m = K_1 e^{-1} \quad \text{(A14)}$$

where $K_1 = \left(\frac{M}{2\pi DP^2S}\right)^{1/2}$

$$r_m = 2Pt_m = \sqrt{2}\left(\frac{M}{\pi DS}\right)^{1/2} e^{-1} \quad \text{(A15)}$$

2. *Modified Ozmidov Solution:*

$$t_m = K_2 e^{-1} \quad \text{(A16)}$$

where $K_2 = \left(\frac{M}{6\pi\gamma^2 DS}\right)^{1/3}$

$$r_m = (3\gamma t_m)^{3/2} = \frac{3}{\sqrt{2}}\left(\frac{M}{\pi DS}\right)^{1/2} e^{-3/2} \quad \text{(A17)}$$

3. *Modified Okubo and Pritchard Solution:*

$$t_m = K_3 e^{-1/2} \quad \text{(A18)}$$

where $K_3 = \left(\frac{M}{\pi W^2 DS}\right)^{1/2}$

$$r_m = Wt_m = \left(\frac{M}{\pi DS}\right)^{1/2} e^{-1/2} \quad \text{(A19)}$$

4. *Okubo Solution:*

$$t_m = K_4 e^{-1/2} \quad \text{(A20)}$$

where $K_4 = \left(\frac{4M}{3\pi^{3/2}\bar{a}^3 DS}\right)^{1/3}$

$$r_m = \left(3/2\bar{a}^2 t^2_m\right)^{3/4} = \left(\frac{6}{\pi}\right)^{1/4}\left(\frac{M}{\pi DS}\right)^{1/2} e^{-3/4} \quad \text{(A21)}$$

5. *Obukhov Solution:*

$$t_m = K_5 e^{-1/3} \quad \text{(A22)}$$

where $K_5 = \left(\frac{M}{\pi\beta^3 DS}\right)^{1/3}$

$$r_m = \left(\beta^3 t^3_m\right)^{1/2} = \left(\frac{M}{\pi DS}\right)^{1/2} e^{-1/2} \quad \text{(A23)}$$

Table A1. *Values, as a Function of Diffusion Model, of the time, t_m, required for the concentration to reach a value of* 6.4×10^{-2} *μCi/cm^3 at a maximum distance, r_m, and the time, t_o, required for the center of contamination (release point) to reach* 6.4×10^{-2} *μCi/cm^3*

Model	$M = 10^4$ Ci			$M = 10^8$ Ci		
	r_m (meters)	t_m (hr)	t_o (hr)	r_m (meters)	t_m (hr)	t_o (hr)
Joseph & Sendner ($n = 1$)	36	0.34	0.92	3600	33.8	92
Ozmidov	33	2.9	8	3300	63	172
Okubo & Pritchard	43	0.49	0.8	4300	49	81
Okubo	38	5.6	9.2	3800	121	199
Obukhov	43	6.1	8.5	4300	131	183
Schonfeld	31	0.32	0.72	3100	28.6	72.4

6. *Schonfeld Solution:*

$$t_m = K_6 \left(1/3\right)^{3/4} \tag{A24}$$

where $$K_6 = \left(\frac{M}{2\pi DS\omega^2}\right)^{1/2}$$

$$r_m = \sqrt{2}\omega t_m = \left(1/3\right)^{3/4} \left(\frac{M}{\pi DS}\right)^{1/2} \tag{A25}$$

Values of the maximum distance at which the concentration of 6.4×10^{-2} μCi/cm^3 will be found can be calculated from the summary equations presented above. The time required for this maximum distance to be reached can also be calculated. The time, t_0, required for the center of the contaminated area (release point) to reach the concentration of 6.4×10^{-2} μCi/cm^3 can also be calculated from the equations for S_0. These values are shown in Table A1 for two values of the amount of plutonium released (M).

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