

Sub seabed disposal of high level radioactive wastes

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Diffusion and advection controlled migration of radionuclides through sub seabed sediments is analyzed to determine the flux at the sediment-water boundary which is subsequently translated into radionuclide concentration in ocean water. The emplaced canister is modelled as an area source and analytical solutions are obtained for two modes of releases, viz. instantaneous release and release due to leaching from a fixed inventory, the former being a worst case condition. Results indicate little influence of canister/waste matrix integrity on concentration. Any radionuclide with a half-life $< 10^5$ y and $K_d > 10^3$ ml.g⁻¹ will not appear in ocean surface water when buried at 10 m. Unlike other radionuclides, ¹²⁹I and ²³⁸U show appreciable and more or less uniform concentration for burial depths ranging from 1-50 m under all conditions studied. Sensitivity of pore water velocity, porosity and temperature on concentration is examined and found that they affect the radionuclide concentration by different rates.

In the management and disposal of high level radioactive waste (HLW), several options¹⁻⁴ related to emplacement in geological formations situated in land areas such as salt domes, argillaceous formations (clay) and hard rocks including granite, basalt, limestone and other impermeable rocks have been considered. Stable geological formations underneath the ocean⁵⁻⁸ may also prove suitable for disposal of HLW. Sub seabed disposal of HLW is being considered⁹⁻¹⁵ presently as a potential alternative to geological repository.

In the present study, unidimensional geospheric model has been used to evaluate radionuclide concentration in pore water for 2 types of release modes from HLW buried in sub seabed: (i) instant disintegration of waste matrix followed by immediate entry of radionuclides into pore water and (ii) slow leaching from waste matrix to pore water. The different processes considered in this model include waste matrix/canister integrity, advective and diffusive migration of radionuclides, their adsorption onto sediment and thermal effects on adsorption and diffusion. The concentration of radionuclide in pore water is translated into flux at sediment-water boundary and used as a continuous input source for oceanic concentration model. The oceanic concentration model consisting of processes like diffusional migration, geochemical elimination, physical decay and reflection at boundaries, is used to compute the temporal distribution of radionuclide concentration in ocean

surface water arising out of sub seabed disposal of HLW. Peak concentration of radionuclides in ocean surface water is discussed in relation to different parameters considered for burial options.

Mathematical Formulation

Pore water concentration—The radioactive waste buried in sediment matrix is subjected to advective and diffusive migration through interstitial pore water along with radioactive decay. In addition, adsorption of radionuclides by sediment would retard the mobility of the nuclides in pore water. From a consideration of rate of change of radioactivity in a unit volume of saturated sediment phase consisting of pore water and corresponding solid phase, the following equation¹⁶ is obtained.

$$\epsilon \frac{\partial C}{\partial t} + (1 - \epsilon) \frac{\partial q}{\partial t} = \epsilon D \frac{\partial^2 C}{\partial z^2} - \epsilon U \frac{\partial C}{\partial z} - \epsilon \lambda C - (1 - \epsilon) \lambda q \quad \dots (1)$$

where C is radionuclide concentration in pore water (Bq.ml⁻¹); q is radionuclide volumetric concentration in solid phase (Bq.ml⁻¹); ϵ is total porosity; D is vertical diffusivity in pore water (cm².s⁻¹); U is pore water vertical velocity (cm.s⁻¹); z is burial depth (cm) origin being at the

discharge point; t is time elapsed after the release (s); and λ is physical decay constant (s^{-1}). In Eq. 1, lateral dispersion is neglected in order to maximize the vertical flux entering the sediment-water boundary.

Assuming rapid equilibrium for the adsorption of radionuclides by sediment, concentration of radionuclides in sediment can be represented as $q = [K_d \rho / (1 - \epsilon)] C$ where K_d is concentration factor for a radionuclide in sediment ($ml.g^{-1}$); ρ is bulk density of the sediment ($g.ml^{-1}$). Eq. 1 can be rewritten using the value of q as

$$\frac{\partial C}{\partial t} = D_1 \frac{\partial^2 C}{\partial z^2} - U_1 \frac{\partial C}{\partial z} - \lambda C \quad \dots (2)$$

where $D_1 = D/\alpha$; $U_1 = U/a$ and retardation factor, $a = 1 + (K_d \rho / \epsilon)$.

The solution for Eq. 2 for a unit instant release of radioactivity (1 Bq) is obtained as

$$C = \frac{\exp[-\lambda t] \exp[-(z - U_1 t)^2 / 4 D_1 t]}{2 \epsilon_e A a (\pi D_1 t)^{1/2}} \quad \dots (3)$$

where A is cross sectional area of dump site (cm^2) and ϵ_e is effective porosity.

Sub seabed burial (case 1)—In this case, a situation is envisaged under which the radioactive material is available for migration through pore water shortly after burial in sub seabed due to low integrity of the compacted waste and canister. In such a case the concentration of radionuclides in pore water is governed by Eq. 3.

The activity flux¹⁷ at the sediment-water boundary, $\psi_1(z, t)$, is obtained from $A \epsilon_e [U_1 C - D_1 (\partial C / \partial z)]$ as

$$\psi_1(z, t) = \frac{(z + U_1 t) \exp[-\lambda t] \exp[-(z - U_1 t)^2 / 4 D_1 t]}{4 a (\pi D_1 t)^{1/2}} \quad \dots (4)$$

Any radioactive material deposited at sediment-water boundary is subjected to advective transport, diffusion, radioactive decay, geochemical elimination and reflection at boundaries. As is known, the horizontal dispersion in ocean is very large¹⁸ and the material spreads horizontally throughout the entire ocean before any significant vertical migration takes place. The equation¹⁹⁻²¹ governing the concentration of a radionuclide in water for an instant release of 1 Bq at sediment-water boundary is given below.

$$C_w = \frac{\exp[-\lambda_e t] \exp[-x^2 / 4 K t]}{S (\pi K t)^{1/2}} + \frac{\exp[-\lambda_e t]}{2 S h}$$

$$\times \left\{ \operatorname{erfc} \frac{2h+x}{(4Kt)^{1/2}} + \operatorname{erfc} \frac{2h-x}{(4Kt)^{1/2}} \right\} \quad \dots (5)$$

where $\operatorname{erfc}(z) = 1 - (2/\sqrt{\pi}) \int_0^z \exp(-y^2) dy$

and S is area of ocean (cm^2); h is depth of ocean (cm); x is depth coordinate with origin at sediment-water boundary (cm); K is vertical diffusion coefficient in ocean water ($cm^2.s^{-1}$); λ_e , the effective elimination rate coefficient in ocean $= (\lambda + \lambda_g)$; λ_g being the geochemical elimination rate coefficient in ocean for the nuclide (s^{-1}).

The activity concentration of radionuclides in ocean water, C_w , arising out of unit activity release at sub seabed is given by the integral

$$C_w = \int_0^t \psi_1(z, t-t') C_w(x, t') dt' \quad \dots (6)$$

where t is total time and t' is time spent by the nuclide in ocean water.

Eq. 6 is difficult to solve in a closed form and hence its value is computed using summation of an infinite number of instantaneous releases, sequentially arranged with a vanishingly small time interval, Δt , between releases. The summation form of Eq. 6 is given as

$$C_w = \Delta t \sum_{n=1}^{t/\Delta t} \psi_1(z, t-n\Delta t) C_w(x, n\Delta t) \quad \dots (7)$$

Sub seabed burial (case 2)—Another situation is envisaged under which HLW is vitrified with glass and encased in canisters. The vitrified waste matrix would prevent radioactivity coming into pore water for time periods equivalent to its integrity, thereby allowing the nuclide to decay within the canister to that extent. Leach studies conducted on vitrified waste form show rapid release of radioactivity in the beginning (fast component) followed by slow release for long periods of time²⁰ (slow component). Since the integrity of the vitrified waste form is sufficiently higher than the normal integrity of canisters, canister integrity is ignored for computation. The leach flux, $L(t)$, coming out of vitrified waste form can be represented as

$$L(t) = B_1 \exp(-\lambda_{11} t) + B_2 \exp(-\lambda_{12} t) \quad \dots (8)$$

where λ_{11} is the effective removal rate coefficient for fast component $= (\lambda + K I_1)$; $K I_1$ being the leach rate coefficient for fast component (s^{-1}); λ_{12} , is effective removal rate coefficient for slow compo-

nent = $(\lambda + Kl_2)$; Kl_2 being the leach rate coefficient for slow component (s^{-1}). B_1 and B_2 are the fast and slow component fluxes ($Bq.s^{-1}$) which are given by

$$B_1 = Qf_1/[(f_1/Kl_1) + (f_2/Kl_2)]; B_2 = Qf_2/[(f_1/Kl_1) + (f_2/Kl_2)]$$

where Q is the total inventory of waste (Bq); f_1 and f_2 are fractions of fast and slow components respectively.

The pore water concentration arising out of leaching of vitrified waste form in sub seabed is given by the integral

$$C_v = \int_0^t L(t-t') C(z, t') dt' \quad \dots (9)$$

The solution for Eq. 9 is obtained as

$$C_v = \frac{B_1 \exp[-\lambda_{11}t] \exp[Uz/2D]}{4A\epsilon_e a(D_1\gamma_1)^{1/2}} F[z, t, \gamma_1] + \frac{B_2 \exp[-\lambda_{12}t] \exp[Uz/2D]}{4A\epsilon_e a(D_1\gamma_2)^{1/2}} F[z, t, \gamma_2] \quad \dots (10)$$

where $\gamma_1 = [(U^2/4D_1) - Kl_1]$ and $\gamma_2 = [(U^2/4D_1) - Kl_2]$. Depending on whether γ_1 and γ_2 are positive or negative, F functions will become real or complex. The definitions of F functions for both real and complex²¹ are given as

$$F[z, t, \gamma_a] = \exp[-z(\gamma_a/D_1)^{1/2}] \operatorname{erfc}[z/(4D_1t)^{1/2} - (\gamma_a t)^{1/2}] - \exp[z(\gamma_a/D_1)^{1/2}] \times \operatorname{erfc}[z/(4D_1t)^{1/2} + (\gamma_a t)^{1/2}]$$

$$F[z, t, \gamma_a]/i = 2 \sin(2xy) \{ [\exp(-x^2)/2\pi x] - \operatorname{erfc}(x) \}$$

$$+ 4 \exp(-x^2)/\pi \sum_{n=1}^{\infty} \frac{\exp(-n^2/4)}{(n^2 + 4x^2)}$$

$$\times [2x \sin(2xy) + n \sinh(ny)]$$

where x and y are $z/(4D_1t)^{1/2}$ and $(|\gamma_a t|)^{1/2}$ respectively and $i = \sqrt{-1}$.

The activity flux at sediment-water boundary, $\psi_2(z, t)$, is obtained as

$$\psi_2(z, t) = \frac{B_1 U_1 \exp[-\lambda_{11}t] \exp[Uz/2D]}{8a(D_1\gamma_1)^{1/2}} F[z, t, \gamma_1] + \frac{B_2 U_1 \exp[-\lambda_{12}t] \exp[Uz/2D]}{8a(D_1\gamma_2)^{1/2}} F[z, t, \gamma_2] + \frac{B_1 \exp[-\lambda_{11}t] \exp[Uz/2D]}{4a} G[z, t, \gamma_1] + \frac{B_2 \exp[-\lambda_{12}t] \exp[Uz/2D]}{4a} G[z, t, \gamma_2] \quad \dots (11)$$

$$G[z, t, \gamma_a] = \exp[-z(\gamma_a/D_1)^{1/2}] \times \operatorname{erfc}[z/(4D_1t)^{1/2} - (\gamma_a t)^{1/2}] + \exp[z(\gamma_a/D_1)^{1/2}] \operatorname{erfc}[z/(4D_1t)^{1/2} + (\gamma_a t)^{1/2}]$$

Depending on whether γ_1 and γ_2 are positive or negative, the G function will also become real or complex. The complexity in the function is removed by following the procedure of Sarma *et al.*²¹ and the complex G function is obtained as

$$G[z, t, \gamma_a] = 2 \cos(2xy) \operatorname{erfc}(x) + \exp[-x^2] \times [1 - \cos(2xy)]/\pi x + 8x \exp[-x^2]/\pi \times \sum_{n=1}^{\infty} \exp[-n^2/4] \times [\cosh(ny) - \cos(2xy)]/(n^2 + 4x^2)$$

The concentration of radionuclide in ocean water due to burial in sub seabed, therefore, can be obtained from the integral given below

$$C_{w2} = \int_0^t \psi_2(z, t-t') C_w(x, t') dt' \quad \dots (12)$$

Eq. 12 is not solvable in a closed form without some approximations and hence the integration is replaced by summation as has been done earlier. For most of the radionuclides, summation method has been employed to arrive at the concentration profile in ocean water.

The above summation technique using Eq. 12 involves large computer times for evaluation of concentration of long-lived radionuclides having half-life $> 10^6$ y. Hence an approximate solution of Eq. 12 could be obtained by assuming F and G functions to be constant and integrating the rest of Eq. 12 to a time period t_1 equivalent to about 25 times of respective geochemical residence time of a radionuclide. This assumption is valid for those radionuclides having short geochemical residence times compared to the period of computation, since their concentration is mainly governed by their effective half-life during that time period which is nearer to period of interest. For such cases, Eq. 12 can be rewritten as

$$C_{w2} = B_3 \int_0^{t_1} \exp[-\lambda_{11}(t-t')] C_w(x, t') dt' + B_4 \int_0^{t_1} \exp[-\lambda_{12}(t-t')] C_w(x, t') dt' \quad \dots (13)$$

where B_3 and B_4 are constants resulting due to approximation and are equal to

$$B_3 = \frac{B_1 U_1 \exp[Uz/2D] F[z, t, \gamma_1]}{8a(D_1 \gamma_1)^{1/2}} + \frac{B_1 \exp[Uz/2D] G[z, t, \gamma_1]}{4a}$$

$$B_4 = \frac{B_2 U_1 \exp[Uz/2D] F[z, t, \gamma_2]}{8a(D_1 \gamma_2)^{1/2}} + \frac{B_2 \exp[Uz/2D] G[z, t, \gamma_2]}{4a}$$

Defining $\omega_1 = (\lambda_g - Kl_1)$ and $\omega_2 = (\lambda_g - Kl_2)$, solution for Eq. 13 is obtained as

$$C_{w2} = B_3 \exp[-\lambda_{11} t] \left[\frac{F[x, t_1, \omega_1]}{2S(K\omega_1)^{1/2}} + \frac{P[(2h \pm x), t_1, \omega_1]}{4Sh\omega_1} \right]$$

$$+ B_4 \exp[-\lambda_{12} t] \left[\frac{F[x, t_1, \omega_2]}{2S(K\omega_2)^{1/2}} + \frac{P[(2h \pm x), t_1, \omega_2]}{4Sh\omega_2} \right]$$

... (14)

where P function²¹ is defined as

$$P[(2h \pm x), t_1, \omega_a]$$

$$= \exp[-(2h+x)(\omega_a/K)^{1/2}] \operatorname{erfc} \left[\frac{2h+x}{(4Kt_1)^{1/2}} - (\omega_a t_1)^{1/2} \right] + \exp[-(2h-x)(\omega_a/K)^{1/2}]$$

$$\times \operatorname{erfc} \left[\frac{2h-x}{(4Kt_1)^{1/2}} - (\omega_a t_1)^{1/2} \right]$$

$$+ \exp[(2h+x)(\omega_a/K)^{1/2}] \operatorname{erfc} \left[\frac{2h+x}{(4Kt_1)^{1/2}} + (\omega_a t_1)^{1/2} \right] + \exp[(2h-x)(\omega_a/K)^{1/2}]$$

$$\times \operatorname{erfc} \left[\frac{2h-x}{(4Kt_1)^{1/2}} + (\omega_a t_1)^{1/2} \right]$$

$$- 2 \exp(-\omega_a t_1) \left[\operatorname{erfc} \frac{2h+x}{(4Kt_1)^{1/2}} + \operatorname{erfc} \frac{2h-x}{(4Kt_1)^{1/2}} \right]$$

When the value of ω_a becomes negative, the P function becomes complex and its value²¹ can be rewritten as

$$P[(2h \pm x), t_1, \omega_a]$$

$$= 2 \cos(2x_1 y) \operatorname{erfc}(x_1) + \exp(-x_1^2) [1 - \cos(2x_1 y)] / \pi x_1$$

$$+ 2 \cos(2x_2 y) \operatorname{erfc}(x_2) + \exp(-x_2^2) [1 - \cos(2x_2 y)] / \pi x_2$$

$$- 8x_1 \exp(-x_1^2) / \left\{ \pi \sum_{n=1}^{\infty} \frac{\exp(-n^2/4)}{(n^2 + 4x_1^2)} \right\}$$

$$\times [\cos(2x_1 y) - \cosh(ny)]$$

$$- 8x_2 \exp(-x_2^2) / \left\{ \pi \sum_{n=1}^{\infty} \frac{\exp(-n^2/4)}{(n^2 + 4x_2^2)} \right\}$$

$$\times [\cos(2x_2 y) - \cosh(ny)]$$

$$- 2 \exp(-\omega_a t_1) [\operatorname{erfc}(x_1) + \operatorname{erfc}(x_2)]$$

where x_1 , x_2 and y are $(2h+x)/(4Kt_1)^{1/2}$, $(2h-x)/(4Kt_1)^{1/2}$ and $(\omega_a t_1)^{1/2}$ respectively.

It should be clearly understood that Eq. 14 is applicable only for radionuclides having long half-lives and short geochemical residence times. Further, time period of computation t should be sufficiently greater than t_1 so that $t-t_1$ can be approximated to t .

Data Source

Eqs 7 and 12 have been used to compute radionuclide concentration in ocean surface water arising out of sub seabed disposal of HLW for case 1 and case 2 respectively. Eq. 14 has been employed for long-lived nuclides having short geochemical residence time to compute their concentration at very long time periods. About 22 radionuclides commonly encountered in HLW have been chosen for this study. The physical characteristics of radionuclides such as half-life, K_d (at 30°C) and geochemical residence time are obtained from literature^{22,23} (Table 1). Studies²⁴ show that K_d generally decreases with increase in temperature. However, there are a few reported cases^{7,15} wherein K_d increases with increase in temperature mainly due to different reactions taking place at higher temperatures. An empirical relation depicting reduction of K_d with temperature is developed based on experimental data²⁴ as $K_d(T) = K_d(30) \exp[-0.023 (T-30)]$. Use of lower K_d for higher temperature leads to higher concentration which is a conservative approach.

Vertical migration of pore water in sediments under natural conditions is estimated to range¹⁵ from 3×10^{-8} to 3×10^{-10} cm.s⁻¹ and Duursma *et al.*²⁴ estimated convective pore water velocity of the order of 10^{-9} cm.s⁻¹ even at high temperature (70°C). In the present study pore water velocities are varied from 3×10^{-8} to 3×10^{-6} cm.s⁻¹

Table 1 - Parameters used for computation

Nuclide	Half-life (y)	Geochemical residence time (y)	Concentration factor (ml.g ⁻¹) at	
			30°C	70°C
²⁴¹ Pu	13.2	2.0(2)*	5.0(4)	2.0(4)
²¹⁰ Pb	21.0	1.0(3)	1.0(4)	4.0(3)
⁹⁰ Sr	28.1	4.0(6)	5.0(2)	2.0(2)
¹³⁷ Cs	30.2	6.0(5)	5.0(2)	2.0(2)
²⁴³ Cm	32.0	6.0(2)	5.0(4)	2.0(4)
²³⁸ Pu	86.0	2.0(2)	5.0(4)	2.0(4)
²⁴¹ Am	4.58(2)	6.0(2)	5.0(4)	2.0(4)
²²⁶ Ra	1.60(3)	4.0(4)	5.0(2)	2.0(2)
¹⁴ C	5.73(3)	1.0(5)	1.0(2)	4.0(1)
²⁴⁰ Pu	6.58(3)	2.0(2)	5.0(4)	2.0(4)
²⁴³ Am	7.37(3)	6.0(2)	5.0(4)	2.0(4)
²⁴⁵ Cm	9.30(3)	6.0(2)	5.0(4)	2.0(4)
²³⁹ Pu	2.44(4)	2.0(2)	5.0(4)	2.0(4)
²³¹ Pa	3.25(4)	5.0(1)	5.0(3)	2.0(3)
²³⁰ Th	8.00(4)	2.0(2)	5.0(6)	2.0(6)
⁹⁹ Tc	2.12(5)	1.0(4)	1.0(4)	4.0(3)
²⁴² Pu	3.79(5)	2.0(2)	5.0(4)	2.0(4)
⁹³ Zr	1.50(6)	4.9(2)	1.0(4)	4.0(3)
²³⁷ Np	2.14(6)	6.0(2)	5.0(4)	2.0(4)
¹²⁹ I	1.70(7)	4.0(5)	1.0(2)	4.0(1)
²³⁸ U	4.49(9)	3.0(6)	5.0(2)	2.0(2)
²³² Th	1.41(10)	2.0(2)	5.0(6)	2.0(6)

Surface area of ocean, $S = 7.412 \times 10^{17} \text{ cm}^2$ Vertical diffusivity in ocean, $K = 5 \text{ cm}^2 \text{ s}^{-1}$ Pore water vertical diffusivity, $D = 3 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ Porosity, $\epsilon = 0.1$ Bulk density of sediment, $\rho = 1.8 \text{ g.ml}^{-1}$ *Read as 2×10^2 and similar elsewhere

mainly to account for abnormal situations leading to higher pore water velocities in some regions. Similarly, molecular diffusion coefficient corrected for temperature^{15,24} has been used for describing diffusional transport of pore water.

Leach rates of vitrified wastes range from 0.1 $\mu\text{g.cm}^{-2}.\text{d}^{-1}$ for glass to 0.1 $\text{g.cm}^{-2}.\text{d}^{-1}$ for cement matrix²⁰. Experiments carried out with vitrified waste²⁰ show 2 component release pattern; fast (90%) and slow (10%) components having half-lives of about 1100 y and 22000 y respectively. These values have been employed in case 2 as fractions (f_1, f_2) and leach rate coefficients (KI_1, KI_2) respectively. The parametric data for sub seabed matrix is somewhat scanty and liable to large variations from place to place. As such, parameters chosen for computation are on the conservative side so that even under worst conditions ocean surface water concentrations may not exceed the computed value.

Discussion

Radionuclides studied presently fall distinctly into 2 categories and represent a wide spectrum of nuclides encountered in HLW management. Category 1 nuclides have either short ($< 10^3$ y) or moderate ($< 10^5$ y) half-lives with high K_d values ($> 10^3 \text{ ml.g}^{-1}$). Category 2 nuclides are long-lived having either small or high K_d values. Peak concentrations of category 1 nuclides computed with and without canister/waste matrix integrity are given in Table 2 for 2 pore water velocities and 2 temperatures. Scrutiny of data reveals that peak concentrations of category 1 nuclides are either nil or negligible when buried at 1 m under all conditions studied. This indicates that category 1 nuclides are highly sensitive to burial depth and a disposal at 10 m would confine them completely in the sediment phase. This is because their migration times (z/U_1) are long compared to half-

Table 2 - Peak concentration of radionuclides in ocean surface water (Bq.ml^{-1}) due to sub seabed burial of unit activity (Bq) at 1 m

Nuclide	$U = 3(-8) \text{ cm.s}^{-1}$		$U = 3(-6) \text{ cm.s}^{-1}$	
	30°C	70°C	30°C	70°C
With canister/waste matrix integrity				
²⁴¹ Pu	NG*	NG	NG	NG
²¹⁰ Pb	NG	NG	NG	NG
⁹⁰ Sr	NG	2.05(-47)	NG	9.37(-45)
¹³⁷ Cs	NG	8.18(-47)	NG	3.55(-44)
²⁴³ Cm	NG	NG	NG	NG
²³⁸ Pu	NG	NG	NG	NG
²⁴¹ Am	NG	1.48(-70)	NG	3.45(-67)
²⁴⁰ Pu	NG	2.96(-43)	1.14(-60)	1.33(-40)
²⁴³ Am	3.00(-74)	4.75(-42)	6.44(-58)	2.57(-39)
²⁴⁵ Cm	7.59(-70)	2.88(-41)	8.78(-54)	1.79(-38)
²³⁹ Pu	1.36(-56)	1.19(-38)	2.01(-43)	2.62(-36)
²³⁰ Th	NG	6.03(-65)	NG	3.58(-62)
Without canister/waste matrix integrity				
²⁴¹ Pu	NG	NG	NG	NG
²¹⁰ Pb	NG	NG	NG	NG
⁹⁰ Sr	NG	8.35(-46)	NG	3.35(-43)
¹³⁷ Cs	NG	4.74(-45)	NG	1.90(-42)
²⁴³ Cm	NG	NG	NG	NG
²³⁸ Pu	NG	NG	NG	NG
²⁴¹ Am	NG	1.46(-69)	NG	8.42(-67)
²⁴⁰ Pu	NG	7.54(-43)	2.78(-60)	2.76(-40)
²⁴³ Am	7.06(-74)	1.03(-41)	9.73(-58)	3.80(-39)
²⁴⁵ Cm	1.53(-69)	7.28(-41)	1.17(-53)	2.67(-38)
²³⁹ Pu	2.11(-56)	1.84(-38)	2.64(-43)	3.41(-36)
²³⁰ Th	NG	7.14(-65)	NG	3.88(-62)

*Negligible ($< 10^{-75}$) and similar elsewhere.

lives thereby allowing decay before reaching the sediment-water boundary. Canister/waste matrix integrity has little influence on concentration; however higher concentration is observed at elevated temperature and pore water velocity. A rise in temperature would decrease the K_d and increase the pore water diffusivity. An elevated pore water advection would decrease the migration time and hence minimize the decay. All these factors enhance the flux of radioactive material into ocean water.

Concentration of category 2 radionuclides in ocean surface water have been computed assuming a 2 component leaching system (case 2) and the corresponding peak values are tabulated in Table 3 for relatively high pore water velocity (3×10^{-6} cm.s $^{-1}$). Similar data are tabulated in the same table assuming no canister/waste matrix integrity (case 1). For both cases, the temperature at the burial site is assumed to be equal to 30°C. Table 3 shows that 8 radionuclides such as ^{14}C , ^{99}Tc , ^{242}Pu , ^{93}Zr , ^{237}Np , ^{232}Th , ^{129}I and ^{238}U appear in ocean surface when they are buried at 50 m. Radionuclides other than ^{129}I and ^{238}U do not show any significant concentration in ocean surface water even when buried at smaller depths. Unlike other nuclides ^{129}I and ^{238}U show appreciable activity in ocean surface water and do not exhibit any functional dependency of concentration on burial depth. Both these radionuclides have long half-lives, long geochemical residence times and small K_d values. Hence their travel times to reach sediment-water boundary are small compared to half-lives thereby not allowing decay during transit through sediment media. Migration of other nuclides like ^{93}Zr , ^{99}Tc and ^{237}Np through sediment matrix is retarded by their high K_d values. As a result, they move through sediment matrix much slower than ^{129}I and ^{238}U . The retarded pore water velocity (U_1) provides sufficient time for decay during transit. Comparison of data in Table 3 reveals that canister/waste matrix integrity does not act as a significant factor to reduce the concentration in ocean surface water. This again can be explained in terms of larger travel time in comparison to the retention time provided by the canister/waste matrix integrity. The retardation time provided by the sediment is significantly higher than the maximum retention time obtained by increasing the canister/waste matrix integrity.

An overview of the above data reveals that burial of HLW in sub seabed at depths ranging 10 to 50 m would allow all short-lived radionuclides to decay within the matrix and all radionuclides

Table 3 – Peak concentration of radionuclides in ocean surface water (Bq.ml $^{-1}$) due to sub seabed burial of 1 Bq activity at different depths ($U = 3 \times 10^{-6}$ cm.s $^{-1}$, $T = 30^\circ\text{C}$)

Nuclide	Burial depth			
	1 m	10 m	20 m	50 m
^{226}Ra	4.57(–31)	5.47(–47)	2.62(–64)	NG
	2.32(–30)	3.52(–46)	1.54(–63)	NG
^{14}C	2.72(–28)	1.99(–29)	2.33(–30)	2.32(–33)*
	1.04(–27)	1.10(–28)	1.14(–29)	1.03(–32)†
^{231}Pa	4.19(–34)	2.86(–42)	3.84(–51)	NG
	8.15(–34)	4.07(–42)	6.17(–51)	NG
^{99}Tc	9.54(–31)	1.46(–33)	2.16(–36)	1.22(–44)
	1.25(–30)	1.60(–33)	2.39(–36)	1.33(–44)
^{242}Pu	1.66(–34)	1.11(–41)	2.93(–49)	9.59(–72)
	1.68(–34)	1.09(–41)	2.89(–49)	9.50(–72)
^{93}Zr	6.94(–32)	7.47(–33)	1.19(–33)	1.48(–34)
	7.35(–32)	1.05(–32)	3.09(–33)	1.40(–34)
^{237}Np	3.57(–33)	4.15(–35)	2.49(–36)	1.57(–40)
	3.13(–33)	6.10(–35)	2.01(–36)	1.26(–40)
^{129}I	1.60(–27)	1.60(–27)	1.57(–27)	1.58(–27)
	1.81(–27)	1.81(–27)	1.81(–27)	1.66(–27)
^{238}U	3.89(–28)	3.88(–28)	3.88(–28)	3.87(–28)
	3.94(–28)	3.94(–28)	3.94(–28)	3.94(–28)
^{232}Th	9.24(–38)	2.86(–38)	1.93(–38)	9.46(–39)
	9.00(–38)	2.70(–38)	1.88(–38)	9.96(–39)

*First line of each nuclide corresponds to concentration with canister/waste matrix integrity.

†Second line of each nuclide corresponds to concentration without canister/waste matrix integrity.

having moderate half-lives with high K_d would decay to innocuous levels during their transit through sediment media. Few radionuclides that reach ocean surface water have either long or moderate half-lives with low K_d values. Peak concentration of these nuclides is either low or peaks are commencing after prolonged periods of time. Figs 1A-B represent time profile of their concentration in ocean surface water for different pore water velocities from a hot (70°C) burial zone of 50 m. For lowest pore water velocity (3×10^{-8} cm.s $^{-1}$), only 7 radionuclides such as ^{129}I , ^{238}U , ^{14}C , ^{93}Zr , ^{237}Np , ^{99}Tc and ^{232}Th appear in ocean surface water and their peak arrival time varies from 10^5 to 10^{11} y. For highest pore water velocity considered (3×10^{-6} cm.s $^{-1}$), 3 more nuclides such as ^{226}Ra , ^{231}Pa and ^{242}Pu appear in ocean water and their peak arrival time varies between 10^4 and 10^9 y. It can be concluded that peak arrival time decreases with increase in pore water velocity and concentration of radionuclide increases with increase in pore water velocity. Com-

putations made beyond 10^6 y after disposal may not have much relevance as the stability of burial site and other characteristics are uncertain. However, computation is extended beyond 10^6 y only to prove that given the site stability, some radionuclides could appear in ocean surface water.

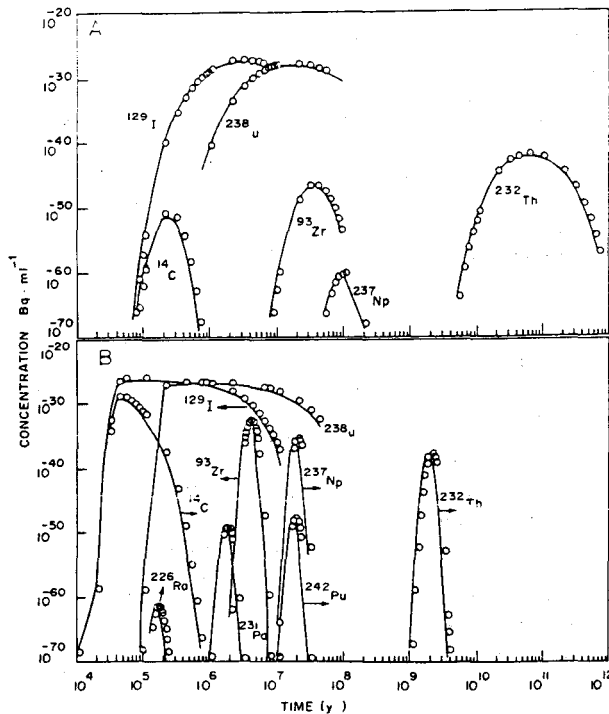


Fig. 1 – Concentration of radionuclides as a function of time due to burial of unit activity (Bq) in sub seabed [Burial depth = 50 m; Temperature = 70°C. A = pore water velocity 3×10^{-8} cm.s $^{-1}$; B = for pore water velocity 3×10^{-6} cm.s $^{-1}$]

Sensitivity analysis – Effluxing of a radionuclide from sub seabed into ocean water depends on characteristics of site, waste form and nuclide. Site characteristics include burial depth, porosity, pore water advection and diffusion whereas waste form characteristics are its integrity and temperature. Precise values for nuclide characteristics such as half-life and K_d are known with sufficient degree of accuracy. However, some of the parameters like pore water velocity, temperature and porosity are uncertain as sufficient experimental data are not available at present. In order to test the sensitivity of these 3 parameters, concentration of category 2 nuclides have been computed using a range of such parameters. Results of a few selected radionuclides are presented in Table 4 for a burial depth of 50m. Computations show that when pore water velocity is decreased from 3×10^{-6} to 3×10^{-7} cm.s $^{-1}$, concentration of nuclides like ^{93}Zr , ^{99}Tc , ^{237}Np , ^{14}C and ^{232}Th is reduced by several orders of magnitude. Further reduction by an order of magnitude causes the disappearance of all nuclides except ^{232}Th , ^{129}I and ^{238}U from ocean surface water. Except ^{129}I and ^{238}U , all other radionuclides are highly influenced by thermal effect by several orders of magnitude especially at lower pore water velocity. ^{129}I and ^{238}U show little variation in concentration with decrease of pore water velocity or increase of temperature. Concentration of all radionuclides rises with increase in porosity by different rates. ^{129}I and ^{238}U show an increase of about 6 times while all other nuclides record a rise by 4-10 orders of magnitude when the porosity is increased from 0.1 to 0.4.

Variation of pore water velocity, porosity or

Table 4 – Peak concentration of radionuclides in ocean surface water (Bq.ml $^{-1}$) due to burial of 1 Bq activity at 50 m for different pore water velocity (U, cm.s $^{-1}$), porosity (ϵ), and temperature (T, °C) – A sensitivity analysis

Condition			Nuclides					
U	T	ϵ	^{93}Zr	^{237}Np	^{242}Pu	^{14}C	^{232}Th	^{129}I
3(-8)	30	0.1	NG	NG	NG	NG	3.72(-49)	1.13(-28)
		0.4	6.07(-57)	NG	NG	NG	8.33(-43)	1.67(-27)
	70	0.1	1.20(-47)	2.14(-61)	NG	6.75(-52)	3.35(-43)	3.87(-28)
		0.4	2.98(-39)	1.56(-46)	9.39(-66)	7.73(-39)	1.29(-40)	5.09(-27)
	30	0.1	1.09(-51)	NG	NG	5.02(-72)	2.69(-41)	1.07(-27)
		0.4	1.48(-37)	8.81(-50)	NG	3.09(-40)	3.69(-39)	6.11(-27)
3(-7)	70	0.1	8.07(-40)	1.92(-51)	NG	2.30(-42)	4.04(-40)	3.59(-27)
		0.4	2.87(-34)	3.17(-39)	1.27(-55)	4.50(-32)	1.09(-38)	1.57(-26)
	30	0.1	1.48(-34)	1.57(-40)	9.59(-72)	2.32(-33)	9.46(-39)	1.58(-27)
		0.4	6.29(-32)	2.23(-34)	5.84(-43)	7.07(-29)	1.81(-37)	6.39(-27)
	70	0.1	4.64(-33)	3.58(-36)	1.45(-48)	5.84(-30)	1.64(-38)	3.94(-27)
		0.4	2.22(-31)	2.58(-33)	3.43(-37)	1.04(-26)	4.78(-37)	1.60(-26)

temperature ultimately changes the travel times of nuclides and determines the flux at the sediment-water boundary. When the half-lives of radionuclides are small compared either to canister/waste matrix integrity or to migration time within the sediment phase, the quantum of activity entering ocean water is considerably reduced or in some cases completely eliminated. In general, it can be stated that nuclides having long half-lives compared to their travel times are less sensitive to these parametric variations, whereas nuclides having half-lives comparable to their travel times are highly sensitive. This analysis indicates that it is necessary to have an accurate knowledge of pore water velocity, porosity and temperature at the site in order to predict the concentration of long-lived radionuclides due to sub seabed disposal of HLW.

Sedimentation rate—Effect of sediment deposition at seabed on upward migration of radionuclides is not considered in above computations, and it may become significant in case of nuclides having long half-lives and high K_d . Radionuclides from sub seabed migrate upwards at a certain rate depending on pore water velocity and K_d while deposition of suspended sediment increases the effective burial depth. When upward migration is compensated with downward sedimentation rate radionuclides would not be able to breach the sediment barrier. Rough calculations indicate that radionuclides having $K_d > 10^5$ would migrate upwards at a rate < 1 mm/1000 y while sedimentation rate in deep regions of Pacific is about 1 mm/1000 y and in other oceans sedimentation rate is much higher. Hence radionuclides like ^{232}Th and other transuranics, because of their high K_d are not likely to enter ocean water and one may have to only consider nuclides like ^{129}I and ^{238}U for exercising option of burial of HLW.

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