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Modeling Study on Nuclide Transport in Ocean– an Ocean Compartment Model

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해양에서의 핵종이동 모델링–해양구획 모델

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Abstract

An ocean compartment model simulating transport of nuclides by advection due to ocean circulation and intertaction with suspended sediments is developed, by which concentration breakthrough curves of nuclides can be calculated as a function of time. Dividing ocean into arbitrary number of characteristic compartments and performing a balance of mass of nuclides in each ocean compartment, the governing equation for the concentration in the ocean is obtained and a solution by the numerical integration is obtained. The integration method is specially useful for general stiff systems. For transfer coefficients describing advective transport between adjacent compartments by ocean circulation, the ocean turnover time is calculated by a two–dimensional numerical ocean model. To exemplify the compartment model, a reference case calculation for breakthrough curves of three nuclides in low–level radioactive wastes, Tc–99, Cs–137, and Pu–238 released from hypothetical repository under the seabed is carried out with five ocean compartments. Sensitivity analysis studies for some parameters to the concentration breakthrough curves are also made, which indicates that parameters such as ocean turnover time and ocean water volume of compartments have an important effect on the breakthrough curves.

요 약

해수유동에 의한 이류와 부유물과의 상호작용에 의한 해양에서의 핵종이동을 모사하여 시간에 따른 핵종농도의 파과곡선을 구할 수 있는 해양구획모델이 개발되었다. 해양을 임의 수의 구획으로 나누어 각 구획간의 질량수지를 통해 지배 방정식을 세우고 이에 대한 해를 stiff한 문제에 유용한 수치적분방법을 이용하여 구하였다. 2차원 해수유동모델을 이용하여 해수교체시간을 계산한 후 각 구획간의 해수유동에 의한 이류수송을 나타내는 이동계수를 구하였다. 개발된 해양구획모델에 대한 계산예로써 해저에 위치한 가상 처분장으로 부터 방출된 저준위방사성 폐기물의 주요 핵종중 Tc-99 Cs-137 및 Pu-238에 대한 파과곡선을 5개구획에 대해 구하였다. 또한 핵종농도 파과

곡선에 대한 파라미터의 민감도 분석을 수행한 결과 구획내의 해수체적 및 해수교체시간과 같은 주요 변수들이 파과곡선에 중요한 영향을 줌을 알 수 있었다.

1. Introduction

Two possible alternatives for disposal of low-level radioactive wastes arising from nuclear power plants are, in general, geological disposal and ocean disposal. The latter, as a possible viable case in view of public acceptance, would be a disposal alternative either under the seabed of the continental shelf or in the vicinity of the ocean. In connection with the assessment of environmental impacts of ocean disposal of wastes on human environment, the behavior of nuclides in the ocean is regarded as one of the key factors which decide the capacity of the ocean as a possible recipient of wastes. In this respect, a theoretical modeling for nuclide migration in the ocean is needed for the first step of safety assessment. To this end a modeling study for nuclides transport entering the ocean directly from the repository located under the seabed or in the vicinity of the ocean is performed. The ocean is divided into arbitrary number of characteristic compartments where instataneous uniform mixing of ocean water occurs. The model is based on assumptions that all nuclides are released into one of the compartments of the ocean and constant transfer rates of nuclides between two adjacent compartments can be parametrized as similarly developed by many authors [1-3].

In this paper the model simulating transport of nuclides by advection due to ocean circulation and intertaction with suspended sediments is developed, by which concentration of each nuclide in each compartment is calculated as a function of time. Performing a balance of mass of nuclides in each compartment, the differential equation system for the concentration of the ocean is obtained and a solution by the numerical integration is obtained. This integration method is specially useful for stiff systems. For transfer coefficients describing advective transport by ocean circulation, the ocean turnover time, which is actually the inverse of advective transfer coefficients, is calculated by a two-dimensional numerical ocean model.

To exemplify the compartment model developed here, a sample case calculation is carried out with five ocean compartments for ocean in the vicinity of repository site. Sensitivity studies for some parameters to the concentration breakthrough curves are also made.

2. Ocean Transport Model

2.1. The Physical System

The ocean is divided into arbitrary number of compartments as shown in Fig. 1. Ocean waters in each compartment is assumed to be well mixed instantaneously. Following assumptions will be also made :

1. Nuclides are discharged into ocean directly from repository located under seabed.
2. Nuclides enters the ocean waters as soon as they are leached from the repository.
3. Ocean water movement between ocean compartments can be modeled assuming instantaneous uniform mixing within defined compartments and constant rates of transfer between two adjacent compartments.
4. Relevant nuclides transport processes are assumed to be largely advection of nuclides dissolved in the ocean water due to ocean circulatiuon and adsorption onto small sediment particles suspended and removal of them from the ocean water by sedimentation to the underlying sedi-

ments and no direct adsorption onto bottom sediments is assumed (see Fig. 1(b)). Also diffusion between adjacent compartments is assumed to be negligible compared to advective transport.

5. Transport of nuclides occurs only between two adjacent compartments.

6. These processes can be parametrized into transfer coefficients between the compartments and nuclide concentration profile in each compartment can be calculated by performing a mass balance on each compartment.

$$V_m \frac{dC_m^i}{dt} = \sum_{n \neq m}^{M} \left(\lambda_{nm}^i V_n C_n^i - \lambda_{mn}^i V_m C_m^i \right) - \left(\lambda^i + k_m^i \right) V_m C_m^i + S_m^i \quad (1)$$

The first two terms on the right hand side of Eq. (1) represent nuclide incoming to the compartment m via turnover of ocean water and any incoming from other adjacent compartment. The remaining three terms allow for loss by radioactive

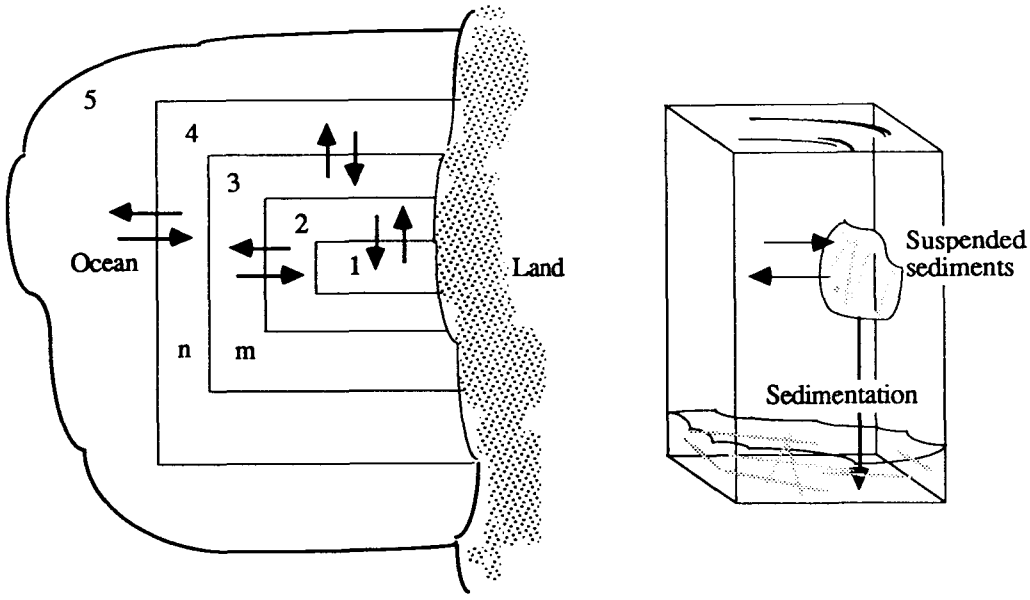

Fig.1. Nuclide transport in the ocean water (a:between adjacent compartments ; b:interaction with suspended sediments).

2.2. Governing Equations

The differential equation system for each compartment m can be obtained by balancing the total mass of nuclides between adjacent compartment n and nuclides interacting with suspended sediments compartment. The equations are [2]

decay, sedimentation and for direct source input from the repository. Only considering main nuclides in low-level wastes, daughter nuclides by decay chain can be ignored.

In the initial state when nuclides are released from the repository nuclide concentration is set to zero for all compartments. Threfore the initial concentration is

$$C_m^i(0) = 0 \quad \text{at } t=0 \tag{2}$$

Eq. (1) may be rewritten in simpler vector form as Eq. (3)

$$\frac{d\tilde{C}^i}{dt} = \mathbf{A}^i\tilde{C}^i(t) + \tilde{S}^i(t) \tag{3}$$

where transfer coefficient matrix A^i is defined as Eq. (4), which can be determined from Eq. (1) and S^i is volume-normalized source term $S_m^i(t)/V_m$

$$\mathbf{A}^i = A_{mn}^i = \begin{cases} \lambda_{nm}^i \frac{V_n}{V_m}, & \text{for } m \neq n \\ -\left(\lambda^i + k_m^i\right) - \sum_{n \neq m} \lambda_{mn}^i, & \text{for } m = n \end{cases} \tag{4}$$

In this case the transfer coefficients between two adjacent ocean water compartments would be given by Eqs. (5) and (6), representing nuclide transport from *m* to *n* and vice versa, respectively.

According to Bolin et al. [4] and Davies [5], in the steady state, the turnover time τ which is equivalent to the nuclide residence time in a compartment can be regarded as the mean time nuclides in the ocean water compartnment have been in there at the moment they are leaving it.

If density of ocean water remains constant, mass can be replaced by volume, and mass flux by volume flux in Eq. (8), which is the inverse of Eq. (5).

$$\lambda_{mn}^i = \frac{1}{\tau_m} \tag{5}$$

$$\lambda_{nm}^i = \frac{V_m}{V_n}\lambda_{mn}^i \tag{6}$$

$$\tau_m = \frac{M_m}{F} \tag{7}$$

$$\tau_m = \frac{V_m}{Q} \tag{8}$$

and also the transfer coefficients from water to the sediments would be given by

$$k_m^i = \frac{\varepsilon(1+R)v_s A_m}{V_m} \tag{9}$$

In Appendix A, Eqs. (5) to (9) is briefly discussed and matrix coefficient $\mathbf{A}^i$ is represented in Appendix B.

2.3. Source Terms

In order to quantify the nuclides into the compartment, nuclides are assumed to enter one of the compartment as soon as they are leached after t_0, at which leaching begins, with constant leach rate *L*. For conservatism, constant infinite release of decaying nuclides is also assumed.

Therefore for direct nuclides releases, the following equation can be employed:

$$S_m^i(t) = I_0 e^{-\lambda^i t} L^i e^{-L^i(t-t_0)}\delta_{ms} \tag{10}$$

where δ_{ms} is kronecker delta function and subscript **s** represents the compartment receiving unclides released from the repository.

2.4. Solution of the Equations

The differential equation system of Eq. (1) that calculate the nuclide concentrations in each compartment of ocean waters is solved using a semi-implicit third-order Runge-Kutta scheme [6]. This scheme is properly selected because Eq. (1) is likely to be a stiff problem. Brief algorithm for solving a system of first order differential equations of the form $y' = f(x)$ with proper initial conditions is given in Appendix C.

3. Reference Case Calculation and Discussion

In this study the reference case is for a hypothetical disposal site located in the Yellow Sea and the East China Sea which is divided into 5 compartments in contact with underlying sediments. The principal purpose is to exercise the model developed here and not to assess the safety of disposal at certain particular site.

3.1 Ocean Turnover Time

For reference ocean where a hypothetical repository is located, volume and flux in each compartment are necessary to calculate the turnover time of Eq. (8). So, the two-dimensional hydrodynamic equations, considering the curvature of the earth and the variation with latitude of the Coriolis acceleration are taken into account by adopting spherical coordinates. The equations of continuity and depth-mean motion as used in this model are : [7]

$$\frac{1}{r\cos\Phi}\left\{\frac{\partial}{\partial\chi}(Hu)+\frac{\partial}{\partial\Phi}(Hv\cos\Phi)\right\}+\frac{\partial\xi}{\partial t}=0 \quad (11)$$

$$\frac{\partial u}{\partial t}+\frac{u}{r\cos\Phi}\frac{\partial u}{\partial\chi}+\frac{v}{r}\frac{\partial u}{\partial\Phi}-\frac{uv\tan\Phi}{r}-2v\omega\sin\Phi$$
$$+\frac{k_b u\sqrt{u^2+v^2}}{H}+\frac{g}{r\cos\Phi}\frac{\partial\xi}{\partial\chi}=0 \quad (12)$$

$$\frac{\partial v}{\partial t}+\frac{u}{r\cos\Phi}\frac{\partial v}{\partial\chi}+\frac{v}{r}\frac{\partial v}{\partial\Phi}+\frac{u^2\tan\Phi}{r}+2u\omega\sin\Phi$$
$$+\frac{k_b v\sqrt{u^2+v^2}}{H}+\frac{g}{r}\frac{\partial\xi}{\partial\Phi}=0 \quad (13)$$

Eqs. (11) to (13) are vertically integrated hydrodynamic equations. For initial condition, at t=o, u(χ, Φ, t), v(χ, Φ, t) and ξ (χ, Φ, t) are specified for all positions at which the equations are to be solved. At a land boundary, the component of the flow normal to boundary is zero and at an open boundary, elevation is specified as a function of time and position along the boundary. The depths used for the grid elements of the model were obtained from Admiralty charts Nos. 1262, 2347.

The finite difference grid of the two-dimensional numerical ocean model used in these calculation is shown in Fig. 2. Details of the numerical scheme were given by many authors [7, 8] and will not be restated here. The tidal residual currents are used for determining the flux in each compartment and these $\overline{u}_i$ are obtained by integrating the depth mean current over one or more tidal periods, i.e.,

$$\overline{u_i}=\frac{1}{T}\int_0^T u_i(t)\,dt \quad (14)$$

Fig. 3 shows the feature of the computed tidal residual currents from the model and its results are in good agreement with the other ones.[7] In order to compute turnover time of each compartment, ocean is divided into five region as is seen in Fig. 2. The turnover time for each compartment can be readily computed once the volume of the compartment and the flux through it are known. In the numerical model the surface area and averaged depth of each grid element are known exactly. Hence the total volume and surface area for each compartment can be determined.

Since the numerical model calculates residual currents across each side of a grid element, the flux into-and out of each compartment can be readily computed provided the boundaries of the compartment coincide with the grid lines used in the two-dimensional numerical ocean model. Having determined the flux into and the flux out of the compartment, at each grid point on the boundary of the compartment, the total flux into and total flux out of the compartment can be

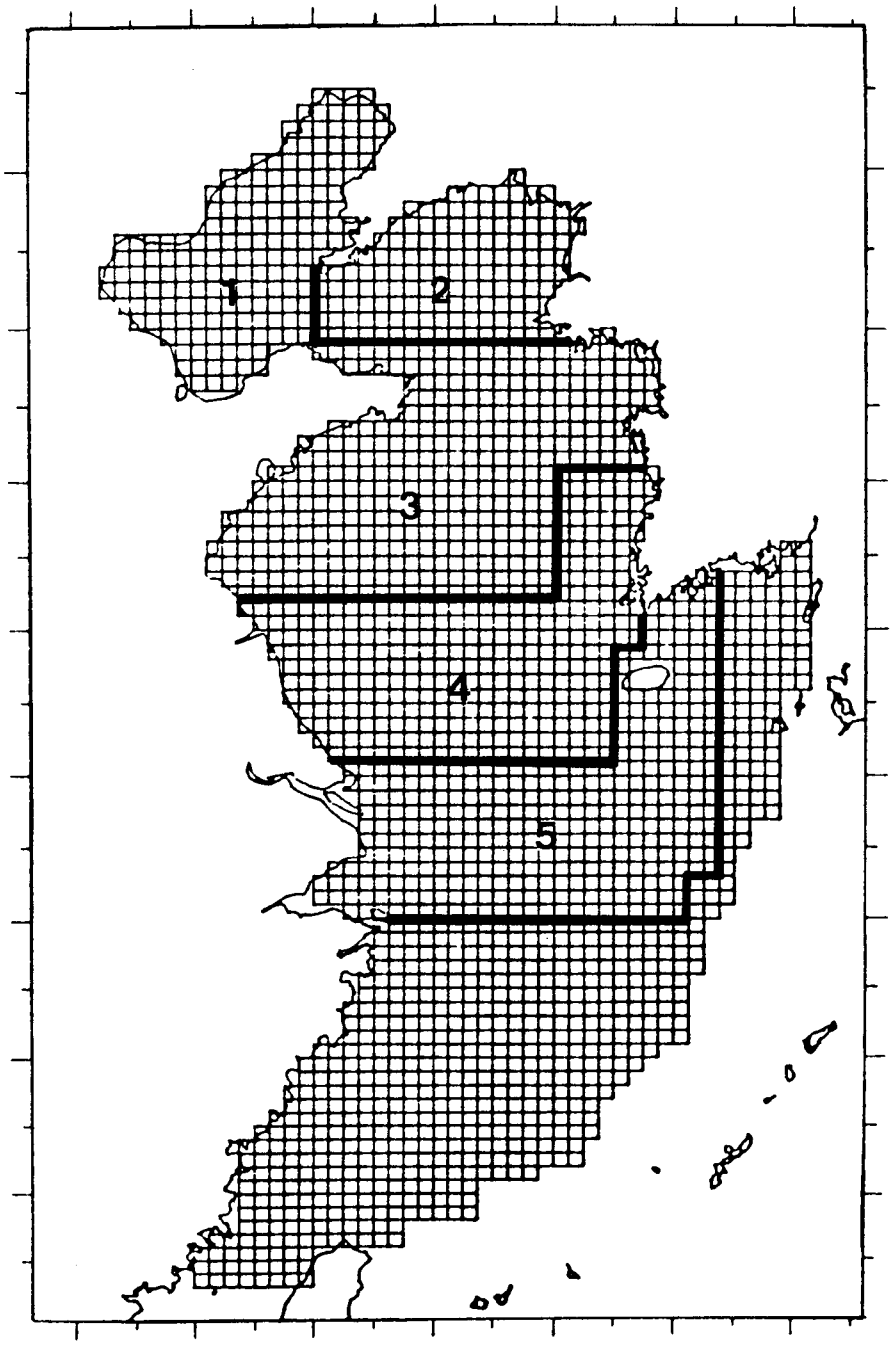

Fig. 2. Finite difference grid of the model, and divided compartments.

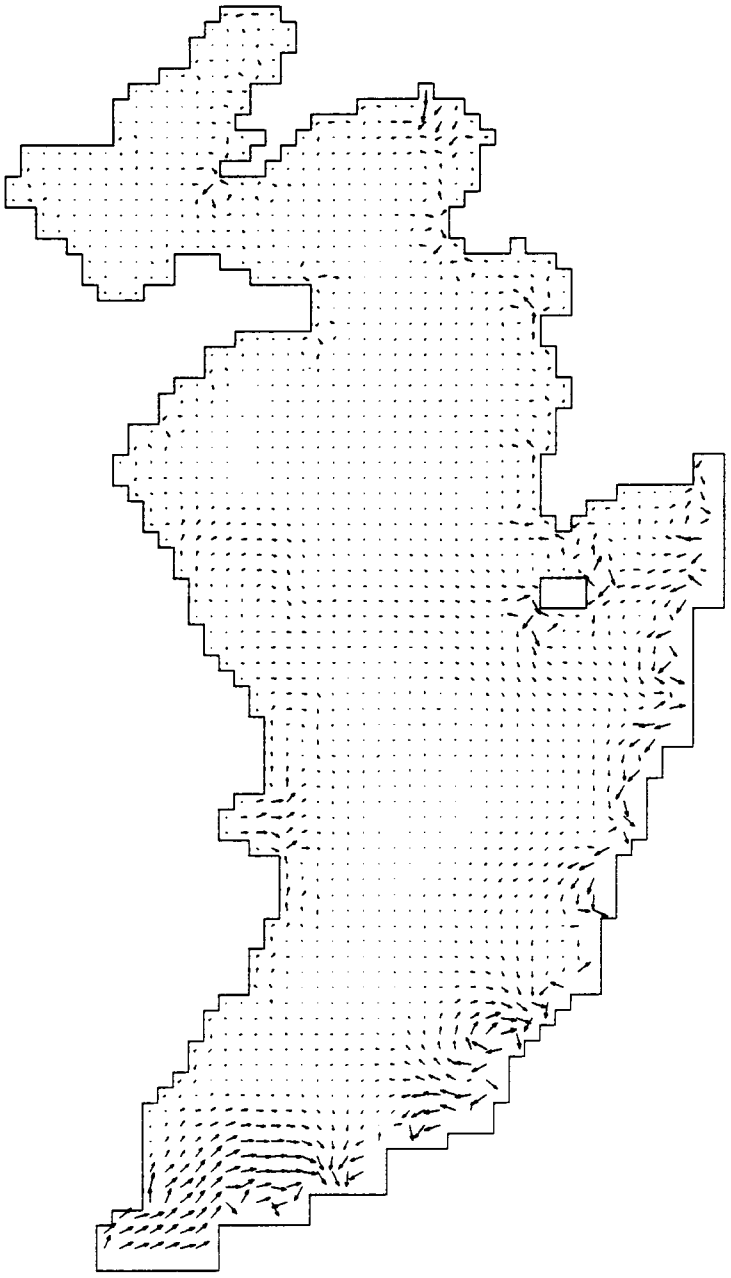

Fig. 3. Computed tidal residual currents.

Table 1. Compartment Dimensions Calculated for Reference Ocean.

Compartment	Sediment area,A_{mn} (m^2)	Interface area,A_m (m^2)	Volume,V_m (m^3)	Turnover time, τ (year)
1	8.45×10^{10}	4.60×10^{6}	1.64×10^{12}	1.8
2	6.82×10^{10}	1.71×10^{7}	2.71×10^{12}	1.7
3	1.73×10^{11}	4.66×10^{11}	8.77×10^{12}	11.9
4	1.47×10^{11}	3.82×10^{11}	6.86×10^{12}	1.7
5	1.69×10^{11}	9.59×10^{11}	1.10×10^{13}	2.5

computed by separately summing the individual fluxes.

Calculated results of turnover time in each compartment as well as dimension of compartments are represented in Table 1. As is seen in Fig. 3, residual currents in the third compartment are relatively weak and that is the reason why turnover time in that compartment is relatively long as represented in Table 1 and then hypothetical repository is assumed to be located there.

3.2 Breakthrough Curves in Ocean Compartments

Only horizontal nuclide transport between compartments by advection and diffusion are considered, as are the effects of adsorption onto suspended sediments and sedimentation. But diffusive transport is assumed to be negligible between adjacent compartments compared to advective transport. The site area is shown in Fig. 2. The repository is assumed to be located under the seabed of the third compartment, arbitrarily. There is only 1.0 Ci of each nuclide in the repository and it is being released infinitely after 100 years since disposal.

For the purpose of an example, only three representative nuclides in low-level radioactive wastes are considered. They are Tc-99, Cs-137 and Pu-238. Tc-99 has long half-life and shows little adsorption capability onto sediments while other two nuclides have short half-life and comparatively large K_d values. Assumed oceanographic- and nuclide specific parameter values used in this study are represented in Tables 2 and 3, respectively. Parameter values such as ocean compartment volumes, area and turnover time calculating transfer coefficients between two adjacent compartments are used as calculated in section 3.1, and its values are shown in Table 1.

Sample results for the reference case is shown in Fig. 4. The figure represents concentration breakthrough curves for the third compartment in normalized volume specific activities Ci/m^3 as a function of time of three nuclides.

The breakthrough curves for both Cs-137 and Pu-238 show a typical exponential decrease with time corresponding to decay. But for Tc-99,

Table 2. Oceanographic parameter values [1]

Parameter	Value
Sediment porosity, ϵ	0.70
Sedimentation rate, v_s	2.5×10^{-5} [m/year]
Sediment bulk density, ρ	8.1×10^{2} [kg/m^3]
Diffusivity, D	3.15×10^{-2} [m^2/year]

Table 3. Nuclide-specific data [1]

Nuclide	Half-life (year)	K_d^i (m^3/kg)
Tc-99	2.1×10^{5}	0.0
Cs-137	30.2	1.0
Pu-238	87.8	10^2

which is not adsorbed onto the sediments and has a long half-life shows a different trend. In the first 10^5 years there is an increase in concentration resulting from the continuing release of nuclides into the ocean. For Tc-99 removal of nuclides relies on decay process only.

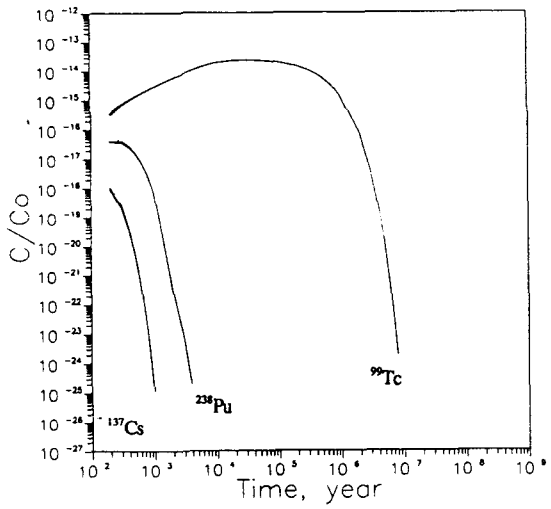

Fig. 4. Concentration breakthrough curves in the 3rd compartment

3.3. Sensitivity Analyses

Sensitivity studies are conducted to examine the effect of compartment model parameters on the predicted nuclide concentrations in the ocean compartments. To this end the concentration breakthrough curves in the 3rd compartment is considered. The parameters in the sensitivity analyses include: distribution coefficients, sedimentation rates, intercompartment transfer rates or ocean turnover time equivalently, compartment volumes, and sediment porosity.

Distribution coefficients and sediment porosity are a measure of adsorption capacity which takes place with suspended sediments in the ocean water. Sedimentation rate may only affect very strongly adsorbed nuclides such as Pu-238. Intercompartment transfer rates are calculated based on the horizontal mixing process such as advection between adjacent compartments. Compartment volumes directly affect the degree of dilution. For nuclides not effectively removed by decay, adsorption or sedimentation, dilution is the main means of lowering nuclide concentrations. For the sensitivity analysis, single parameter values are set to reference values and these values are varied to one order of magnitude lower or higher.

Consequently the sensitivity analysis indicates that one order of magnitude decrease or increase shows no significant effects on the breakthrough curves except for intercompartment transfer coefficient (or turnover time), and compartment volume for all nuclides. In this paper, figures for sensitivity analysis of other parameters except for tansfer coefficient and compartment volume were not presented because of negligible differences. Figs. 5 to 8 show a result of sensitivity studies for two nuclides, Cs-137 and Tc-99 for these two dominant parameter values. In Figs. 5 and 6, for both nuclides turnover times which are actually the inverse of intercompartment transfer rates are unlikely to lead to order of magnitude changes in breakthrough curves. Also raising and lowering one order of magnitude change of volume of each compartment lead to order of magnitude changes in concentrations of both nuclides as is seen in Figs. 7 and 8.

In this study, ocean compartment model is developed to simulate migration of nuclides in ocean with first attempt. The model is only considered ocean circulatio due to tide, but meteorologically induced currents (due to wind stress or atmospheric pressure gradients) et al. should be involved in the next study to compute ocean turnover time. Ocean is also divided into compartment of arbitrary number at this stage, but in future it should be divided with the regions which current profile is similar to one another roughly.

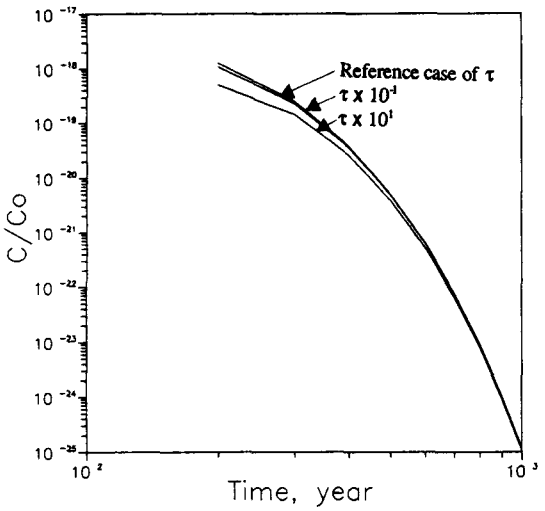

Fig. 5. Sensitivity of concentrations for ^{137}Cs to ocean turnover times for the 3rd compartment.

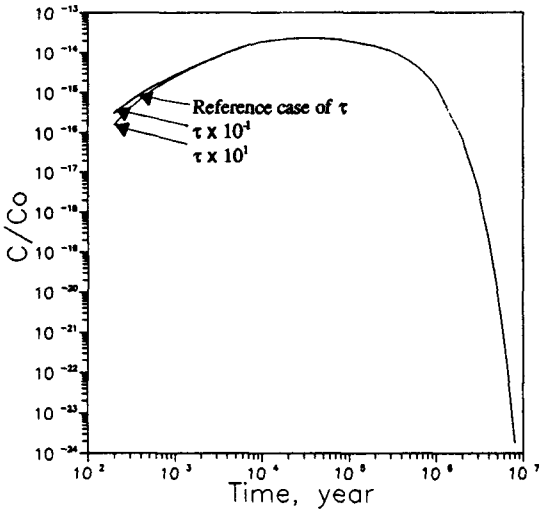

Fig. 6. Sensitibity of concentrations for ^{99}Tc to ocean turnover times for the 3rd compartment.

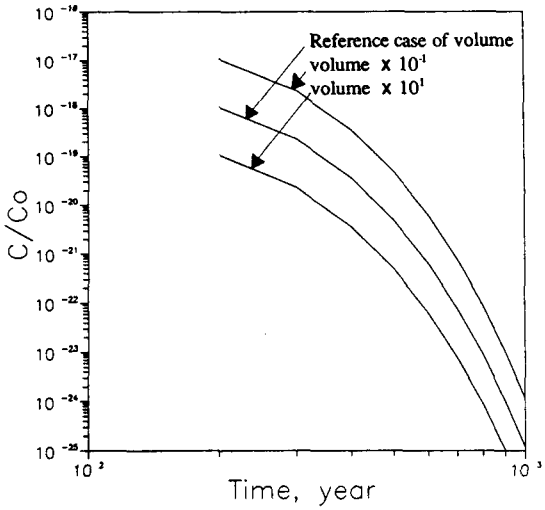

Fig. 7. Sensitivity of concentrations for ^{137}Cs to the 3rd compartment volume.

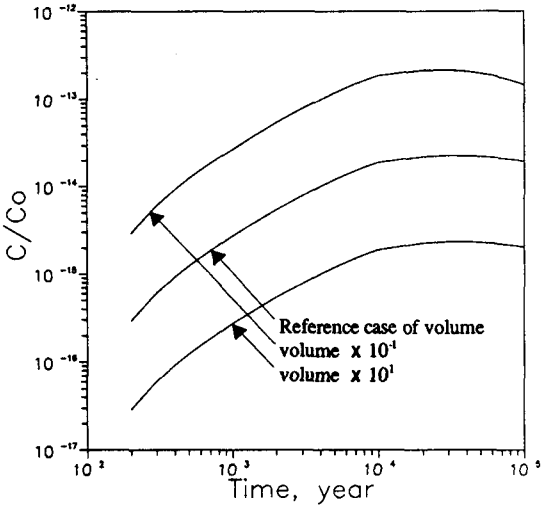

Fig. 8. Sensitivity of concentrations for ^{99}Tc to the 3rd compartment volume.

4. Conclusion

The work presented in this paper represents a simplified modeling study for nuclide behavior in the ocean. To this end the ocean is divided into several characteristic compartments. Performing a balance of mass of nuclides in each compartment, the differential equations for the concentration in the ocean is obtained and a solution by a numerical integration which would be useful for stiff systems is obtained.

To exemplify the model, a reference case calculation is carried out for the ocean divided into five compartments. Sensitivity analysis studies for some parameters to the concentration break-

through curves are also made, and the results show that parameter values such as ocean turnover time and ocean water volumes of compartments have an important effect on the breakthrough curves.

For further study more realistic parametrization for the transfer coefficients and more realistic model for the nuclide injection into the ocean will be needed. However model proposed in this study will be useful in case that safety assessment of the repository in the vicinity of the ocean should be carried out.

Acknowledgment

The authors are indebted to Prof. B.H.Choi for using his shelf circulation model and helpful comments.

Notation

A_m sediment area of compartment *m*, m^2

A_{mn} interface area between adjacent compartments *m* and *n* m_2.

A^i transfer coefficient of nuclide *i* defined in (4).

A^i_{mn} transfer coefficient of nuclide *i* between compartments *m* and *n* defined i (4).

C^i concentratio vector of nuclide *i* defined in (3).

C^i_m concentration of nuclide *i* in compartment *m*, Ci/m^3.

C^i_n concentration of nuclide *i* in compartment *n*, Ci/m^3.

D diffusivity in ocean water, m^2/year.

F mass flow rate between two adjacent compartments, kg/year.

g gravitational acceleration, $m/year^2$.

H total ocean water depth, equal to $h+\xi$, m.

h undisturbed ocean water level, m.

I_o initial nuclide inventory, Ci/m^3.

K^i_d distribution coefficient of nuclide K^i_d, m^3/kg.

k_b coefficient of ocean bottom friction.

k^i_m removal rate of nuclide *i* from compartment *m*, $year^{-1}$.

L^i leach rate of nuclide *i*, $year^{-1}$

M arbitrary number of compartments.

M_m mass of ocean water compartment *m*, kg.

R retardation factor defined in (A12).

r radius of the Earth, m.

S^i source term vector of nuclide *i* in defined in (3).

S^i_m source term vector of nuclide *i* in compartments *m* defined in (3).

T lunar tidal period, sec.

t time, year.

t_o time at which leaching begins, year.

u, v components of depth–mean current, m/sec.

u', v' components of current in the directions of ξ, ϕ, respectively at a depth z, m/sec.

u_i instantaneous tidal velocity, m/sec.

u_i residual currents, m/sec.

V_m volume of compartment *m*, m^3.

V_n volume of compartment *n*, m^3.

v_s sedimentation rate, m/year.

Q water flow rate between two adjacent compartments, m^3/year.

x distance between the center of adjacent compartments, m.

δ_{ms} kronecker delta function.

ϵ porosity of sediment.

λ^i decay constamt of nuclide *i*, $year^{-1}$.

λ^{adv}_{mn} transfer coefficient of nuclide by advection defined in (A1), $year^{-1}$.

λ^{diff}_{mn} transfer coefficient of nuclide by diffusion defined in (A2), $year^{-1}$.

λ^i_{mn} transfer coefficient of nuclide *i* from compartment *m* to *n*, $year^{-1}$.

ξ elevation of ocean surface above the undisturbed level, m.

ρ bulk density of sediment, kg/m^3.

τ_m ocean turnover time in compartment m, year.

Φ latitude, degree.

χ east–longitude, degree.

ω angular speed of the Earth's rotation, degree/year.

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Appendix A. Derivation of Transfer Coefficients

A.1. Transfer Coefficients Between Adjacent two ocean water compartments m and n

The mass transport rates by advection and diffusion between adjacent compartments is expressed respectively as

$$\lambda_{mn}^{adv} C_m V_m = \nu A_{mn} C_m, \tag{A1}$$

and

$$\lambda_{mn}^{diff} C_m V_m = -A_{mn} D \frac{C_n - C_m}{x} \tag{A2}$$

Setting nuclide initial concentration in compartment n in Eq. (A2), total transfer coefficient for two adjacent compartments is

$$\lambda_{mn} = \frac{\nu A_{mn}}{V_m} + \frac{D A_{mn}}{V_m} \tag{A3}$$

If diffusion is negligible, Eq. (A3) is approximated to

$$\lambda_{mn} = \frac{\nu A_{mn}}{V_m} \tag{A4}$$

Also if in steady state water flow rate $Q=\nu A$ is assumed to be constant, the volume exchange rate $\lambda_{mn}V_m$ will be the same for adjacent two compartments :

$$\lambda_{mn} V_m = \lambda_{nm} V_n \tag{A5}$$

or

$$\lambda_{mn} = \frac{V_n}{V_m} \lambda_{nm} \tag{A6}$$

which means the inverse of the ocean turnover time from the relation $\tau = V_m/\nu A$, i.e.,

$$\lambda_{mn} = \frac{1}{\tau_m} \tag{A7}$$

A.2 Transfer Coefficients for the Interaction of Nuclides With Suspended Sediments

The expression for distribution of nuclides between water and sediments is derived from the following relationship.

$$\varepsilon C_m + \rho K_d C_m = C_n \tag{A8}$$

where the distribution coefficient K_d is defined as the ratio of nuclide *i* adsorbed per mass of sediment to nuclide dissolved per volume of water. In this case instantaneous equilibrium and linear exchange isotherm is assumed.

Then the nuclide concentration in the sediments is given by

$$C_n = \varepsilon\left(1 + \rho K_d\right) C_m \tag{A9}$$

Now the rate of removal of nuclides by sedimentation can be expressed by introducing sedimentation rate v_s ;

$$C_m V_m k_m^i = v_s A_m C_n \tag{A10}$$

from which the transfer coefficient for adsorption onto the sediments is given by

$$k_m^i = \frac{\left(\varepsilon + \rho K_d^i\right) v_s A_m}{V_m} \tag{A11}$$

If retardation coefficient is defined as

$$R = \frac{\rho K_d^i}{\varepsilon} \tag{A12}$$

Eq.(A10) is reduced to

$$k_m^i = \frac{\varepsilon\left(1 + R\right) v_s A_m}{V_m} \tag{A13}$$

Appendix B. Transfer coefficient matrix for 5-compartment model

In Eq.(B1) rewritten Eq.(3),

$$\frac{d\tilde{C}^i}{dt} = \mathbf{A}^i \tilde{C}^i(t) + \tilde{S}^i(t) \tag{N1}$$

the transfer coefficient matrix A^i **is expressed as**

$$\mathbf{A}^i = \begin{pmatrix} -\left(\lambda^i + k_1\right) - \lambda_{12}^i & \lambda_{21}^i \frac{V_2}{V_1} & & & 0 \\ \lambda_{12}^i \frac{V_1}{V_2} & -\left(\lambda^i + k_2\right) - \lambda_{21}^i - \lambda_{23}^i & \lambda_{32}^i \frac{V_3}{V_2} & & \\ & \lambda_{23}^i \frac{V_2}{V_3} & -\left(\lambda^i + k_3\right) - \lambda_{32}^i - \lambda_{34}^i & \lambda_{43}^i \frac{V_4}{V_3} & \\ & & \lambda_{34}^i \frac{V_3}{V_4} & -\left(\lambda^i + k_4\right) - \lambda_{43}^i - \lambda_{45}^i & \lambda_{54}^i \frac{V_5}{V_4} \\ 0 & & & \lambda_{45}^i \frac{V_4}{V_5} & -\left(\lambda^i + k_5\right) - \lambda_{54}^i \end{pmatrix}$$

and S as **(B2)**

$$\mathbf{S}^i = \begin{pmatrix} \frac{S_1^i}{V_1} \\ \frac{S_2^i}{V_2} \\ \frac{S_3^i}{V_3} \\ \frac{S_4^i}{V_4} \\ \frac{S_5^i}{V_5} \end{pmatrix} \quad \text{(B3)}$$

Appendix C. Solution algorithm of(1)

For initial value differential equation of the form

$$\frac{\mathrm{d}}{\mathrm{dx}}\mathbf{y} = \mathbf{f}(\mathrm{x}) \quad \text{(C1)}$$

with initial elements,

$$\mathbf{y}(x_n) = \mathbf{y}_n \quad \text{(C2)}$$

Letting the Jacobian **F** be given by

$$F_{ij} = \left(\frac{\partial f_i}{\partial y_j}\right)_{y_i} \quad \text{(C3)}$$

third–order method can be constructed with proper constants, which is called semi–implicit Runge–Kutta method. Then the solution at $t_n + h$, y_{n+1} is found from

$$\begin{cases} \mathbf{k}_1 = h(\mathbf{I} - ha\mathbf{F})^{-1}\mathbf{f}(y_n) \\ \mathbf{k}_2 = h(\mathbf{I} - ha\mathbf{F})^{-1}\mathbf{f}(y_n + b_2\mathbf{k}_1) \\ \mathbf{k}_3 = (\mathbf{I} - ha\mathbf{F})^{-1}(b_{31}\mathbf{k}_1 + b_{32}\mathbf{k}_2) \\ y_{n+1} = y_n + R_1\mathbf{k}_1 + R_2\mathbf{k}_2 + \mathbf{k}_3 \end{cases}$$

where

$$a^3 - 3a^2 + \frac{3}{2}a - \frac{1}{6},\ a = 0.43586652\ldots$$

$$b_2 = 0.75$$

$$b_{31} = \frac{-1}{6a}(8a^2 - 2a + 1)$$

$$b_{32} = \frac{2}{9a}(6a^2 - 6a + 1)$$

$$R_1 = \frac{11}{27} - b_{31}$$

$$R_2 = \frac{16}{27} - b_{32}$$