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ESTIMATION OF RADIONUCLIDES CONCENTRATIONS IN THE OCEAN AT THE HYPOTHETICAL SUBMERGENCE OF FRESH MOX FUEL PACKAGE

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ABSTRACT

Under INF code and IAEA standard, radioactive materials in Type B package are transported safely on the sea. To gain the supplementary public acceptance for these transports, environmental impact assessments have been made by assuming that a Type B package might be sunk into the sea. A method of the assessment consists of the estimation of release rates of radionuclide from a package, simulation of radionuclide concentration both in coastal and global areas, and dose assessment for the public.

We summarized the radiological impact at the submergence of Type B packages, such as spent fuel (SF), PuO_2 powder, high level wastes (HLW) and fresh mixed oxide (MOX) fuel in coastal areas. The evaluated dose equivalents by radiation exposure to the public for all materials were far below the limit of the ICRP recommendation (1mSv $year^{-1}$). These assessments had a lot of uncertainties especially in the simulation of radionuclide concentration therefore the results might be overestimated.

We developed more realistic methods to simulate both in coastal and global areas to explain the evaluated results of the impacts to the public in an efficient manner. General circulation models for the Japan Sea and for the world ocean were employed at coastal and global areas respectively. It is impossible to validate the method directly because no accident with release of radionuclide in the ocean has occurred. Therefore we simulated background radionuclide concentrations by fallouts (^{137}Cs and $^{239,240}Pu$) to validate the methods. Fallouts were input into ocean by nuclear weapon test since 1945 and mainly in 1960s. And their concentrations in the ocean have been measured to keep monitoring the artificial contaminations now. Observed database is useful to compare with simulated results in both coastal and global areas. Simulated results were in good agreement with observation.

And then, we simulated radionuclides concentrations at the hypothetical release from the submerged Type B package in the Japan Sea and global ocean. Simulated concentrations in the coastal and global areas were quite smaller than the background concentration by the fallouts.

INTRODUCTION

Irradiated nuclear fuels such as spent fuel (SF), PuO_2 powder, high level wastes (HLW) and fresh mixed oxide (MOX) fuel have been transported by sea between Europe and Japan. There is a special safety standard called INF (Irradiated Nuclear Fuel) Code at International Maritime Organization (IMO) for the structure and systems of transport ship of irradiated nuclear fuels. On the other hand, for transport of radioactive materials, there is a safety standard stipulated in "Regulations for the Safe Transport of Radioactive Material" issued by International Atomic Energy Agency (IAEA). During transportations, these irradiated nuclear fuels are contained in the Type B package under INF code and IAEA standard.

To gain the public acceptance for these transports additionally, dose assessments for public at the hypothetical submergence of irradiated nuclear fuel package into the sea were carried out in the past 20 years by Central Research Institute of Electric Power Industry (CRIEPI) (spent fuel (SF)[(1)], PuO2 powder[(2)], high level wastes (HLW)[(3)], high burn-up spent fuel[(4)], and fresh mixed oxide (MOX) fuel[(5)]). And then, CRIEPI summarized the radiological impact at the submergence of Type B packages, spent fuel (SF), PuO_2 powder, high level wastes (HLW) and fresh mixed oxide (MOX) fuel in the coastal area in IAEA TECDOC[(6)] and journal[(7)]. The evaluated results of the dose equivalent based on the ICRP Publication 30 for all materials were far below the dose equivalent limit of the ICRP recommendation (1mSv $year^{-1}$). These assessments had a lot of uncertainties especially in the simulation of radionuclide concentration. Large–scale materials circulation models in the ocean have not been validated directly with the observations. In these assessments, highest concentrations in temporal and spatial were used for the dose assessment for the public. Therefore, the results might be overestimated.

Methods of the impact assessments both for near shore and deep sea areas consists of,

(1) Estimation of release rate of radionuclide from a package,
(2) Simulation of radionuclide concentration
(3) Estimation of dose assessment for the public.

Methods to simulate ocean circulation and diffusion have been improved with rapid progress on the super computing. Ocean general circulation models become useful to simulate the radionuclides concentration in the ocean.

Here we developed methods to simulate radionuclides concentrations in the ocean by Ocean General Circulation Models. More realistic assessments are necessary to gain the public acceptance for sea transport of radioactive materials.

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METHOD

We defined two areas for the impact assessments in this study. One is the location near coast called as "near shore" for smaller and shorter scale, another is the location far from coast called as "deep sea" for larger and longer scale. Two different scales methods should be employed for each area because reasonable spatial and temporal scales are important for the assessments.

Release scenarios

Release scenarios for near shore and deep sea are employed from previous studies. Here we summarized the methods in brief. Detailed descriptions are in the references[(5)(6)(7)].

At the case of near shore, the depth of submergence of the Type B package for irradiated nuclear fuel is estimated to be 200m. It would be possible to salvage the package from a depth of less than 200 m, the depth of 200m was conservatively assumed for the assessment in case of submergence near shore. The package will not rupture and keep integrity according to the IAEA transport regulation at the depth of 200 m.

The barrier effect scenario that the presence of package reduced the release rate of nuclides to the ocean is employed.
Accordingly, a conservative scenario was assumed as follows;
(1) The package is submerged on the seabed at the depth of 200m.
(2) After submergence, sealability is lost by a functional disorder of O-ring immediately.
(3) Seawater enters into the cavity of the package.
(4) Radioactive materials are exposed to the seawater .
(5) Nuclides leach the seawater in the cavity of the package.
(6) The solution of nuclides is released to the ocean through the seal gap.
At the case of deep sea, the depth of submergence of the Type B package is estimated to be several thousands meters far from coast. The package will not keep integrity due to the high water pressure. In this case, no package is considered. Accordingly, a conservative scenario was assumed as follows;
(1) The package is submerged on the seabed at the depth of several thousands meters.
(2) Radioactive materials are exposed to the seawater.
(3) Nuclides leach the seawater directly

Simulation of radionuclides concentrations

We developed off-line tracer models using the achieved results from ocean general circulation models both for near shore and deep sea areas to simulate the radionuclides concentration in the ocean at the hypothetical release of radionuclides from a submerged package.

At the case of the near shore, we employed the achieved results from an ocean general circulation model for Japan Sea[8]. This is based on Modular Ocean Model (MOM) version 1.1. The basic equation on the three-dimensional diffusion equation was taken into consideration both nuclides decay and scavenging(nuclides removed from seawater by phenomena that nuclides absorb suspended materials in seawater and settle down the seabed).
Off-line calculation was carried out using Japan sea general circulation model's outputs, which contains monthly mean current and density. The flow field was constructed by liner interpolation of twelve monthly results. Inter-annual change is not considered in this calculation. These equations were calculated by the finite elemental methods.

At the case of the deep sea, we employed achieved results of the NCOM (National Center for Atmospheric Research Ocean Model)[9]. The same basic equation on the three-dimensional diffusion equation as a near shore case was taken into consideration both nuclides decay and scavenging. Off-line calculation was carried out using NCOM's outputs, which contains monthly mean current and density. Flow field was constructed by liner interpolation of twelve monthly results. Inter-annual change is not considered in this calculation. These equations were calculated by the finite elemental methods.

Estimation of effective dose of radiation exposure to the public

At the case of near shore, the internal dose from ingestion of fish in the area of calculation and the external dose by marine operations were calculated in the same manner of previous studies [5][6][7] (referring the model to calculate the internal dose equivalent in ICRP Pub. 72). The external dose was caused by working on shipboard, swimming, working at beach and operation of fishery net.
At the case of the deep sea, the internal dose equivalent from ingestion of fish in the area of calculation was only calculated by similar to near shore case. The external dose is neglected because this case is far from shore.

RESULTS AND DISCUSSION

Case studies are carried out for near shore and deep sea according to the previous study for fresh MOX fuel [5]. Target radionuclides of fresh MOX fuel in this study are ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{241}Am.

Near Shore

A location of the hypothetical submergence of one package of the fresh MOX fuel was supposed at the depth of 200m in Wakasa bay area in the Japan Sea.

Release rate of radionuclides

Release rated for each nuclides are employed from previous study[5] with the barrier effect model. The concentration of nuclides in the cavity was saturated in 0.1 years after submergence of a package, after then, the release rate of ^{240}Pu was about 2.0 x 10^{9} Bq year^{-1} after 2 years.

Concentration in the Japan Sea

Vertical averaged concentrations at the surface layer (0 ~ 100m) are important to assess the radiological impact at a hypothetical submergence accident because the public ingests the seafood mainly caught from ocean surface[5]. Temporal change of surface averaged concentration (0 ~ 100m) is shown in Figure 1. ^{241}Pu concentration is highest for first 50 years. Only ^{241}Am concentration increases for first 100 years by daughter product. Total concentration is highest at 2 years after release and then decreases. Horizontal distribution of total concentration of 6 nuclides is shown in Figure 2. High concentration area occurs north-eastward from release point in front of Wakasa Bay. Radionuclides advect north-eastward from the sea bottom at the depth of 200m although the direction o surface "Tsushima current" is north-westward. Three-dimensional model can represent characteristics of real ocean. The surface concentration is about 7.0 x 10^{-5} Bq m^{-3} when one package of fresh MOX fuel submerged at the depth of 200m in front of Wakasa Bay. This value is about 6 times smaller than the previous study[5] due to difference of the models to calculate concentrations in the ocean. In the previous study, vertical distribution of horizontal current was not considered. Shear effect of the vertical distribution of horizontal current reduced the simulated concentrations of radionuclides in this three dimensional model.

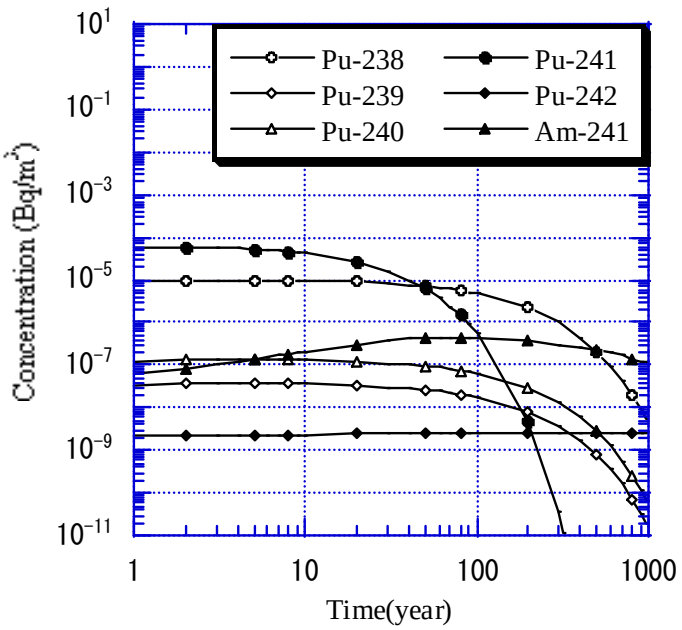

Figure 1. Maximum surface concentration (vertical averaged concentration from 0 to 100m)

Comparison with fallout concentrations

The presence of artificial radionuclides in the ocean is due to the global fallout generated by atmospheric nuclear weapons tests and a nuclear reactor accident. The Meteorological Research Institute (MRI) in Japan has collected historical data (Historical Artificial Radionuclides in the Pacific Ocean and its Marginal Seas (HAM) database [10]) on three anthropogenic radionuclides (^{90}Sr, ^{137}Cs and ^{239,240}Pu(The total of ^{239}Pu and ^{240}Pu was described as ^{239,240}Pu in the database)) in the world ocean, mainly in the Pacific Ocean and its marginal seas. Deposition rate of fallout have been measured since 1958 at the site of the Meteorological Research Institute[11]. The distributions of fallout in the Japan Sea were simulated in considering with input condition from observed deposition rate by this method[12]. Simulated results were in good agreements with observed database. Simulated horizontal distribution of ^{239,240}Pu in the surface layer is from 2 x 10^{-3} to 5 x 10^{-3} Bq m^{-3} in the Japan Sea in 1993. Simulate concentrations at the hypothetical submergence of one fresh MOX fuel package is far smaller than recent background concentration by fallout. The additional concentration by the release from package is too small to detect in comparison with background concentration.

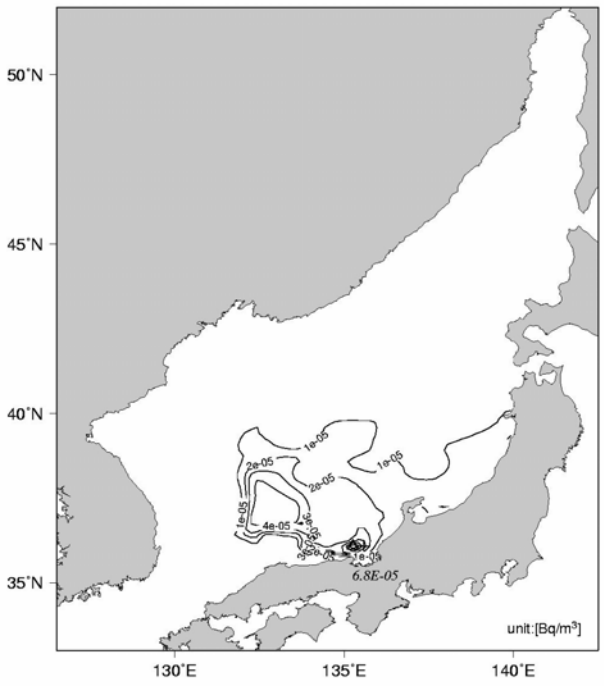

Figure 2. Horizontal distribution of total concentration of 6 radionuclides at 2 years after release from a package at the bottom of 200m depth in front of Wakasa Bay.

Result of the effective dose of radiation exposure to the public.

If the package submerges on the seabed off the Wakasa bay, the maximum effective dose in temporal and spatial of radiation exposure to the public was estimated. The effective dose of radiation exposure to the public is 4.3 x 10^{-8} mSv $year^{-1}$ at 2 years after release of radionuclides which is far less than the effective dose limit (1 mSv $year^{-1}$) by the ICRP recommendation

Deep Sea

A location of the hypothetical submergence of one package of the fresh MOX fuel was supposed at the depth of 2500m near Japan

Release rate of radionuclides

Release rated for each nuclides are employed from previous study[5] without the barrier effect model. In this scenario, a release rate of the nuclides to the ocean is equal to the leaching rate of nuclides. Release rate of ^{241}Pu was about 1.0 x 10^{13} Bq $year^{-1}$ for first 20 years and decreased by decay. The daughter products increase release rate of ^{241}Am.

Concentration in the world ocean

Temporal change of surface averaged concentration (0-100m) is shown in Figure 3. ^{241}Pu concentration is highest at 70 years after release and then decrease. Total concentration is also

highest at 200 years after release and keep constant for 1000 years. Horizontal distribution of total concentration of 6 nuclides at 200 years after release is shown in Figure 4. High concentration area occurs near Japan. And radionuclides advect and diffuse worldwide even if the concentration is quite low far from release point. Maximum surface concentration is about 3.0 x 10^{-5} Bq m^{-3} when one package of fresh MOX fuel submerged at 2500m depth near Japan. This value is larger than the result by compartment model in previous study[5] which is about 3.0 x 10^{-7} Bq m^{-3}. Mesh size in this model is smaller than the one in previous compartment model. Mesh size of this model is about 4.0 x 104 km2 and mesh sizes of compartment model vary from 5.0 x 10^{5} km^{2} to 2 x 10^{7} km^{2}. When the simulated concentrations by an ocean

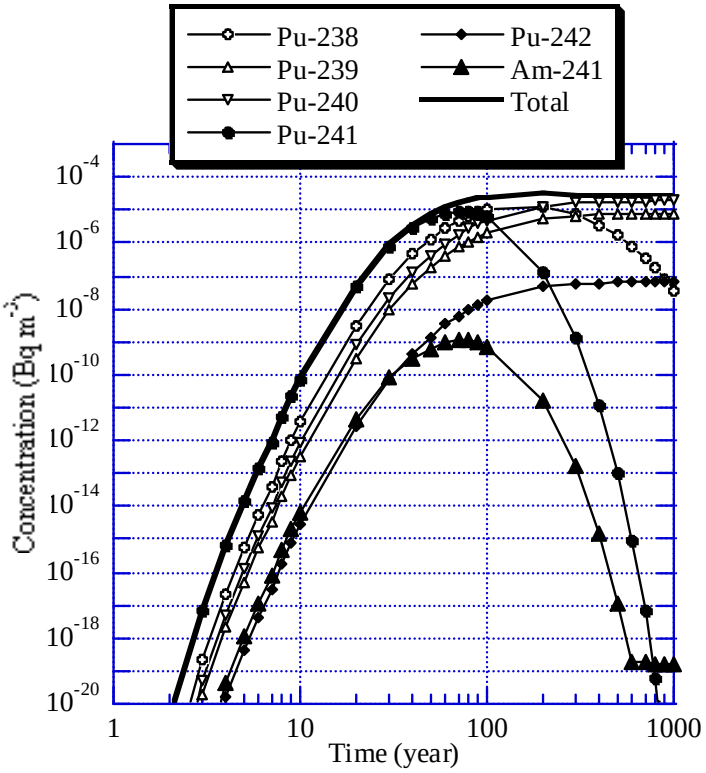

Figure 3. Maximum surface concentration (vertical averaged concentration from 0 to 100m)

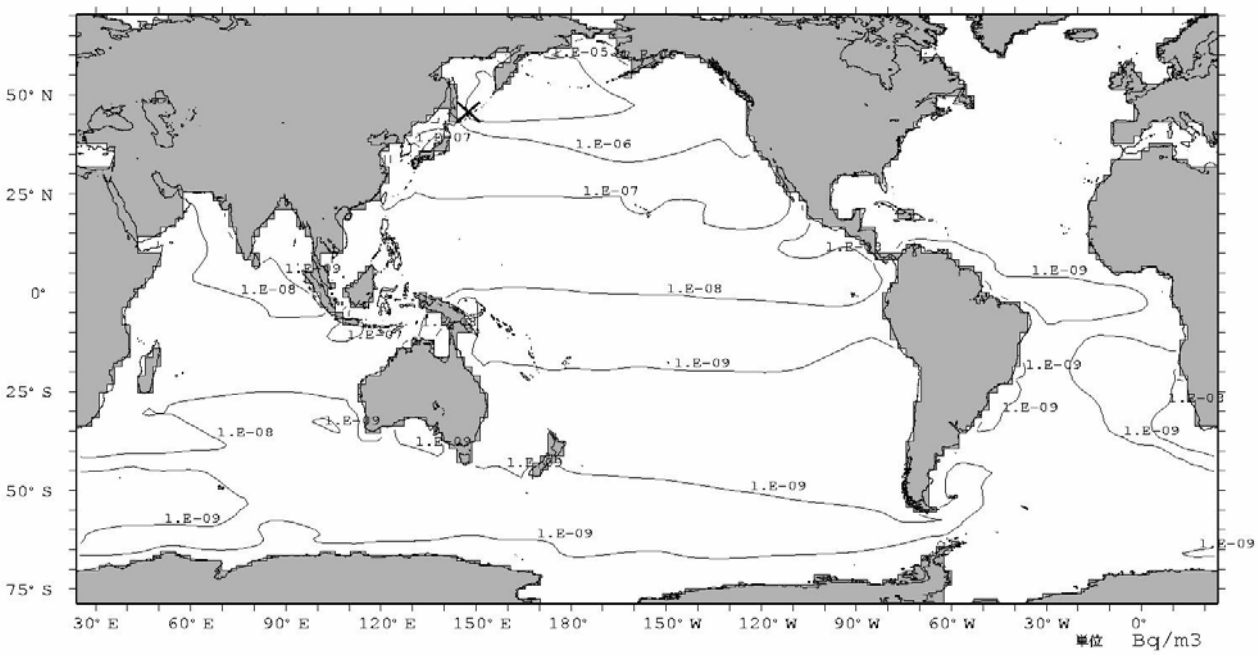

Figure 4. Horizontal distribution of total concentration of 6 radionuclides at 200 years after release from a package at the bottom of 2500m depth near Japan.

general circulation model are averaged with the size of compartments, maximum value is about 5.0 x 10^{-6} Bq m^{-3} in this model. This value in this model is still larger than the one in previous model due to the difference of advection and diffusion processes.

Comparison with fallout concentrations
The distribution of radioactive deposition on the global ocean is estimated from global precipitation data and observed values of annual deposition of radionuclides at Meteorological Research Institute (MRI) in Japan. The distributions of fallout in the global ocean were simulated in considering with input condition from observed deposition rate by this method[13][14]. Simulated results were in good agreements with observed database. Simulated horizontal distribution of ^{239,240}Pu in the surface layer is from 1.0 x 10^{-3} to 1.0 x 10^{-1} Bq m^{-3} in the world ocean in 1993. Simulate concentrations at the hypothetical submergence of one fresh MOX fuel package is far smaller than recent background concentration by fallout. The additional concentration by the release from package is too small to detect in comparison with background concentration.

Result of the effective dose of radiation exposure to the public.
If the package submerges on the seabed at 2500m near Japan, the maximum effective dose in temporal and spatial of radiation exposure to the public was estimated. The effective dose equivalent of radiation exposure to the public is 2.9 x 10^{-8} mSv $year^{-1}$ at 200 years after release of radionuclides which is far less than the effective dose limit (1 mSv $year^{-1}$) by the ICRP recommendation.

Comparison with other irradiated nuclear fuel.
CRIEPI summarized the dose equivalent for public at the submergence of Type B packages, spent fuel (SF), PuO_2 powder, high level wastes (HLW) and fresh mixed oxide (MOX) fuel in the coastal area in IAEA TECDOC[6] and journal[7]. The dose equivalent in the case of spent fuel was maximum and was about 400 times larger than the case of MOX fuel.

CONCLUSIONS

We employed ocean general circulation models to simulate radionuclides concentration in the ocean both for near shore and deep sea areas at the radiological impact assessment at the hypothetical release of radionuclides from a submerged Type B package of irradiated nuclear fuel. These methods represented the fallout concentration to assess the skill of the model in comparison with observed database. More detailed distributions of radionuclides concentration based on the understanding of ocean circulation were achieved by these methods. More realistic information is useful to explain the impact assessment for the public to gain the public acceptance for sea transport of irradiated nuclear fuel.
Simulate concentrations at the hypothetical submergence of one fresh MOX fuel package is far smaller than recent background concentration by fallout both for near shore and deep sea cases. The additional concentration by the release from package is negligible small in comparison with background concentrations. The effective dose of radiation exposure to the public both for near shore and deep sea cases are far less than the effective dose limit (1 mSv $year^{-1}$) by the ICRP recommendation.

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