ISEIS

Journal of Environmental Informatics 36(2) 133-145 (2020)

Journal of Environmental Informatics

www.iseis.org/jei

Improvement of Instantaneous Point Source Model for Simulating Radionuclide Diffusion under Climate Change

A. F. Zhai[1], X. W. Ding[1, 2 *], Y. Zhao[3], W. H. Xiao[3], and B. X. Lu[1]

[1] *Key Laboratory of Regional Energy and Environmental Systems Optimization, Ministry of Education, North China Electric Power University, Beijing, 102206, China*

[2] *Institute for Energy, Environment and Sustainable Communities, University of Regina, Regina, Saskatchewan S4S 7H9, Canada*

[3] *State Key Laboratory of Simulation and Regulation of Water Cycle in River Basin, China Institute of Water Resources and Hydropower Research, Beijing 100038, China*

Received 27 October 2016; revised 4 March 2017; accepted 2 September 2017; published online 20 December 2017

ABSTRACT. Simulation methods have become an important tool to reveal radionuclide migration during accidental radionuclide releases and predict influences of accidents on the marine environment. The instantaneous point source model is a useful method to simulate the large-scale radionuclide diffusion in marine areas. However, the simulation accuracy of this method requires improvement as it didn't take radionuclide decay into account. In this study, an improved instantaneous point source model considering radionuclide decay was proposed on the basis of the original model. Furthermore, the instantaneous point source model and the improved version were used to simulate the concentrations of 131I and 137Cs following the Fukushima Dai-ichi nuclear power plant accident. The results showed that the relative error of 131I concentrations decreased from 200.07% to 34.12% when using the improved model; and improvements in relative errors for 137Cs concentrations were not apparent as the simualtion period was much shorter than its half-life period. Therefore, the improved model can accurately simulate the diffusion process for radionuclides following an accident and provides an efficient decision support tool for risk assessment managers and for use in safety guarantees of nuclear power plants during siting and operational phases.

Keywords: instantaneous point source model, improved instantaneous point source model, radionuclide diffusion, Fukushima Dai-ichi nuclear power plant accident, decay

1. Introduction

With the increase in energy demand, nuclear power has become important for ecomomic prosperity, social development and environmental protection. Today, most nuclear power plants are located in coastal areas because of the convenient cooling water intake, and feasible radioactive effluent discharge (Brown and Milke, 2016). However, there is a huge potential risk for the operation of coastal nuclear power plants, as radionuclides may be directly released into oceans and the marine environment may become seriously polluted under accident conditions (Sudbrock et al., 2016). In recent decades, radionuclide migration and its effects under accident conditions have received worldwide attention, especially after the Fukushima Dai-ichi nuclear power plant accident in 2011 (Taelman et al., 2014). Simulation methods are an important tool to reveal radionuclide migration under accident conditions and predict the influences of accidents on the marine water environment (Wang et al., 2010; Batlle et al., 2016; Zhai et al., 2020; Zheng et al., 2020). Therefore, it is important to develop the mechanisms and simulation models for radionuclide transport in the case of coastal nuclear power plant accidents.

Many studies have focused on various simulation models to assess radionuclide migration following nuclear accident, such as the instantaneous point source model, the Cox ocean model, Hibler ice model, and coupled ice–ocean model (Monte et al., 2014; Kitamura et al., 2015; Lee et al., 2015). A diffusion equation for radionuclides in oceans has been developed that includes an instantaneous point source model and continuous point source model, and the former was widely applied (Nosov et al., 2005). Based on the tide model, a radionuclide simulation model was developed that is suitable for coastal ocean areas (Periáñez et al., 1996). Based on the Cox ocean model and the Hibler ice model, a coupled ice–ocean model was developed to simulate the diffusion of radioactive pollution in the North Pole and other fringe oceans (Preller and Cheng, 1999). Among these models, the instantaneous point source model has advantages of easily acquired parameters alongside acceptable simulation results, and therefore it is especially suitable for estimating the pollution when a large number of radionuclides are instantaneously discharged into a marine area (Harper et al., 2015). In re-

* Corresponding author. Tel.: +86-010-6177-2982; fax: 010-61772982.
E-mail address: binger2000dxw@hotmail.com (X.W. Ding).

ISSN: 1726-2135 print/1684-8799 online
 doi:10.3808/jei.201700380

cent decades, the instantaneous point source model has been used widely in radionuclide migration simulations and has been recommended by the International Atomic Energy Agency (IAEA) (Walan et al., 2014). However, it retains the limitation that radionuclide decay is not taken into account and therefore desired simulation results cannot be obtained on radionuclide migration, especially for radionuclides with short half life periods or simulation periods with long durations. Therefore, it is necessary to improve it for acquiring more accurate results following nuclear power plant accidents in coastal areas (Huang et al., 1997; Maqsood et al., 2005; Liu et al., 2020; Yao et al., 2020). As an extension of the previous efforts, the objective of this study was to develop an improved model to produce more accurate simulations of radionuclide migration by considering radionuclide decay. In this study, the instantaneous point source model was advanced by taking the radionuclide decay characteristics into account. The improved model was then applied to simulate the migration process of radionuclides following the Fukushima Dai-ichi nuclear power plant accident. Finally, trends of the spatiotemporal distribution for ^{131}I and ^{137}Cs were revealed and discussed. The results of this study provide researchers with an accurate and feasible method to reveal diffusion and transport processes of radionuclides in marine areas under accident conditions. Moreover, it may also provide an efficient tool and a decision support for risk assessment managers and those associated with safety guarantee of nuclear power plants during the siting and operational phases.

2. Materials and Methodology

2.1. Instantaneous Point Source Model

Because of its feasibility and reliability, the instantaneous point source model has been widely used in many studies to estimate radioactive pollution when a large number of radionuclides are instantaneously discharged into marine areas. For oceans, horizontal diffusion plays a key role in dispersive mixing of contaminants given that its horizontal scale is much larger than its vertical scale (Regmi, 2017). In general, contaminant concentrations released from an instantaneous point source in radial distribution decrease regularly in shallow water areas. As the diffusion of contaminant was affected by tide and air volume in neritic region and oceanic zone respecttively, the dynamic of ocean current was the major consideration for the diffusion of radionuclides in this research. The instantaneous point source model is outlined as follows:

$$C(r,t)=\frac{1\times10^{-3}Q}{2\pi hp^2t^2}\exp(\frac{-r}{pt}) \tag{1}$$

where C is the contaminant concentration (Bq/L), r is the distance from a research point to the point source (m), t is time (s), Q is instantaneous point source item (Bq), h is water depth (m), and p is horizontal diffusion (m/s). Contaminant concentration in the location of the point source is expressed as:

$$C(0,t)=\frac{1\times10^{-3}Q}{2\pi hp^2t^2} \tag{2}$$

where meanings of parameters are the same as Formula (1).

2.2. Improved Instantaneous Point Source Model

In recent decades, nuclear power plants have flourished and most of these are located in coastal areas (Wang et al., 2017). Once nuclear accidents occur, medium and even high levels of radioactive effluents will be released into oceans (Konstantinova et al., 2015). For radionuclides, the decay characteristic is a major cause for the reduction of its concentrations in marine area after an accident occurs. However, this characteristic is currently disregarded in the instantaneous point source model, and therefore the original model needs to be improved by taking radionuclide decay into account. Many previous isotope-tracing studies have indicated that there is a negative exponential function relationship between the nucleon number of radioactive isotope and time (Blokhintsev and Savin, 2016; de Térамond et al., 2016). The improved instantaneous point source model is identified as:

$$C(r,t)=\frac{1\times10^{-3}Q}{2\pi hp^2t^2}\exp(\frac{-r}{pt})\cdot k_i \tag{3}$$

where i is a radionuclide, k_i is the decay factor of the radionuclide, and meanings of other parameters are as given in Formula (1). And the decay factor k_i is described as:

$$k_i=(1/2)^{\frac{t}{T_i}} \tag{4}$$

where T_i is the half-life for radionuclide i, and meanings of other parameters are the same as presented for Formula (1).

2.3. Data Processing

The distance from a research point to the point source r is an important factor affecting the radionuclide concentration, and the value can be determined according to the simulation requirements. Time t is another key factor that can be quantified based on the influencing duration of an accident. Instantaneous point source item Q is the total quantity of radionuclide released from an instantaneous point source, which can be acquired from reports of the nuclear power plant and relevant monitoring data. In addition, water depth h can be obtained from the related oceanic administrations. Velocity of horizontal diffusion p is influenced by tide and wind direction and its value is between 0.005 and 0.015 m/s for shallow sea waters (Jankovic et al., 2016). Decay factor of a radionuclide k_i is defined as follows: for a radionuclide, the relationship between the remaining nucleon number and time is:

$$dN=-\lambda\cdot Ndt \tag{5}$$

where N is the remaining nucleon number, λ is the decay constant, and t is the time (s). The half-life period is the period during which the concentration of a radionuclide is reduced to half of its original value. The relationship between the remaining nucleon number and its half-life period is defined as:

$$N = N_0 \cdot (1/2)^{\frac{t}{T}} \quad (6)$$

where N_0 is the initial nucleon number, T is half-life period equaling $ln2/\lambda$, and meanings of other parameters are same as shown in Formula (3). And concentrations of ^{131}I and ^{37}Cs in each research point are processed by ORIGIN.

3. Case Study

3.1. Fukushima Dai-Ichi Nuclear Power Plant Accident

Until now, nuclear accidents have happened in three nuclear plants which were Chernobyl Nuclear Power Plant, Three Mile Island Nuclear Power Plant, and Fukushima Dai-ichi Nuclear Power Plant. Among them, only Fukushima Dai-ichi Nuclear Power Plant was coastal one, the accident of which occurred latest and had a serious influence on marine environment. Therefore, the Fukushima Dai-ichi nuclear power plant accident was chosen as a case study for this research. On 11 March 2011, a strong earthquake occurred off the coast of Japan's main island Honshu and an accompanying large tsunami severely damaged the Fukushima Dai-ichi Nuclear Power Plant (Buesseler et al., 2011; Matsui, 2011). Consequently, a large quantity of radionuclides were released into the environment over an extended period (Prants et al., 2011). Significant releasees occurred on 12 March 2011 with release rates varying considerably over the following week (Takeda et al., 2011). Moreover, ^{131}I and ^{137}Cs were taken as typical radionuclides given that large amounts of these radionuclides were released into marine environment during the Fukushima Dai-ichi accident (Inomata et al., 2015). Specifically, ^{131}I is a short half-life period radionuclide which may accumulate in the thyroid gland for a few weeks when people are exposed to iodine radiation and can harm human health (Andrews et al., 2016). In contrast, ^{137}Cs is a representative long half-life period radionuclide that can remain in the environment over several decades and has an extensive impact on water resources (Tsumune et al., 2012). The radioactive characteristics of ^{137}Cs and ^{131}I are presented in Table 1.

Table 1. Radioactive Characteristics of ^{137}Cs and ^{131}I

Radionuclide	Representativeness	Half-life period	Radiation species
^{131}I	Short half-life period radionuclide	8.3 d	β, γ
^{137}Cs	Long half-life period radionuclide	30.167 a	β, γ

After the accident, the Tokyo Electric Power Company (TEPCO) carried out monitoring among the sea water near Fukushima Dai-ichi nuclear power plant from 21 March 2011. Among the all monitoring points laid out by TEPCO, four were chosen in this case study as research points to simulate the spatiotemporal changes of ^{131}I and ^{137}Cs (Figure 1) based on the distance to the outlets of the two nuclear power stations as well as the direction of seawater movement.

The research points were positioned as follows: the first research point was about 330 m south from the outlet for Units 1 ~ 4 of the Fukushima Dai-ichi Nuclear Power Plant; the second research point was about 30 m north from the outlet for Units 5 and 6; the third was near the north outlet of Fukushima Daini Nuclear Power Plant; and the fourth was located near the Iwasawa coast.

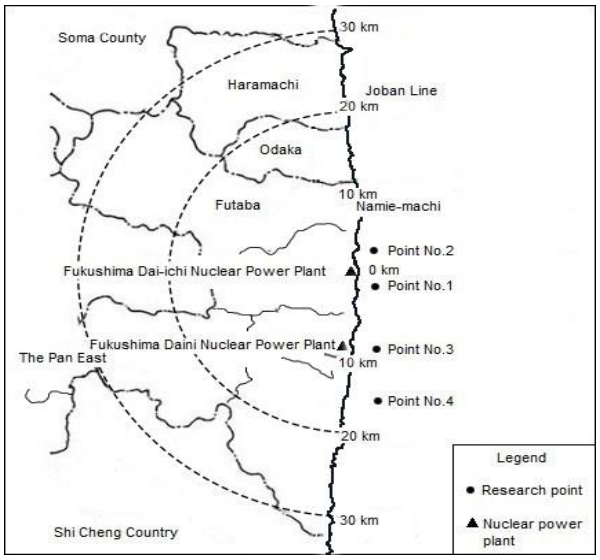


Figure 1. Locations of research points for the Fukushima Dai-ichi nuclear power plant accident.

3.2. Parameter Calibration

^{131}I and ^{137}Cs were transferred by atmospheric circulation after released into the atmospheric environment following Fukushima Dai-ichi nuclear power plant accident, and subsided on the ocean surface. However, the instantaneous point source model had an insufficient simulation accuracy for diffusion process of short half-life period radionuclides under this accident condition. According to the information released by TEPCO and other agencies, a large number of high-level radioactive effluents were also leaked from Unit 2 to the nearby marine area between 2 and 6 April 2011 (TEPCO, 2011; du Bois et al., 2012). It has been reported that there was no subsequent continuous leakage of high level radioactive effluents after 6 April 2011. Therefore, radionuclides discharged in those days could be regarded as an instantaneous point source, and thus 7–24 April 2011 was selected as the simulation period.

Moreover, the total amount of radionuclides released to the sea in the Fukushima Dai-ichi nuclear power plant accident (instantaneous point source item Q) was estimated based on leaked radioactive effluents and radiological environmental monitoring data of the surrounding sea area reported by TEPCO during April 2011. The leakage rate of radioactive effluents was calculated by the following dynamic formula:

$$U = S \cdot v = S\frac{L}{t} = S\frac{L}{\sqrt{\frac{2h}{g}}} \quad (7)$$

where U is the leakage rate of radioactive effluents (m^3/s), S is the sectional area of effluents (m^2), v is the velocity of effluents (m/s), L is splashing horizontal distance of effluents leaked from cracks near the inlet pipe of Unit 2 (m), t is time (s), h is the height of cracks (m), and g is gravitational acceleration (m/s^2).

Specifically, radioactive effluents were leaked from circular cracks with approximate diameters of 3 cm, and sectional area of effluents was obtained as 7.07×10^{-4} m^2. Moreover, L was measured as 0.65 m, h was measured as 0.75 m, and the value of g was 9.8 m/s^2. According to Formula (7), U was estimated as 1.17×10^{-3} m^3/s. Therefore, the total amount of radioactive effluents leaked from the Fukushima Dai-ichi nuclear power plant during 2 ~ 6 April 2011 was 516 m^3. According to monitoring data of radioactive effluents sampled in the vertical shaft of Unit 2 and supplied by TEPCO for 2 April 2011, the concentrations of ^{131}I and ^{137}Cs were 5.40×10^6 and 1.80×10^6 Bq/cm^3, respectively. Therefore, the total amounts of ^{131}I and ^{137}Cs were 2.78×10^{15} and 0.93×10^{15} Bq respectively on the basis of the calculated leakage rate of radioactive effluents and its monitoring values of radionuclide concentration. The total amounts of radionuclides discharged into the sea are given in Table 2.

As horizontal diffusion played a major role in pollutant dispersion in the shallow seas and the ocean surface, radioactive effluents were mixed evenly in the direction of water depth after being discharged into the sea. In this study, the leakage point in the vertical shaft of Unit 2 was chosen as the point source. Radial distances from four research points to the point source were calculated based on longitude and latitude (Table 3).

Table 2. Total Amounts of Radionuclides Discharged into the Sea from 2 to 6 April 2011

Type	Estimated quantity (Bq)
Instantaneous point source item	4.70×10^{15}
^{131}I	2.78×10^{15}
^{137}Cs	0.93×10^{15}

Table 3. Radial Distances from Four Research Points to the Point Source

Research point	Location	longitude and latitude	Radial distance
1	330 m south from the outlet for Units 1 ~ 4 of the Fukushima Dai-ichi Nuclear Power Plant	141°01'57"E 37°23'33"N	330 m
2	30 m north from the outlet for Units 5 and 6	141°02'10"E 37°25'21"N	927 m
3	Near the north outlet of Fukushima Daini Nuclear Power Plant	141°01'16"N 37°18'10"E	10000 m
4	Near the Iwasawa coast	141°01'16"N 37°15'22"E	16000 m

In addition, h was the water depth where the concentration of contamination mixed evenly. Furthermore, its value was set as water depth and depth of mixing layer in neritic region and oceanic zone respectively. In this study, the value of h was taken as 10 m since four research points were located in neritic region and water depths of those points were all about 10m. Velocities of seawater in neritic region and oceanic zone were both large due to the tsunami caused by earthquake, recommended velocity of horizontal diffusion p ranged from 0.005 to 0.015 m/s, and therefore appropriate value of p in this study was set as 0.015 m/s. According to Formula (6) and Table 1, k_i values for ^{131}I and ^{137}Cs were calculated as shown in Table 4.

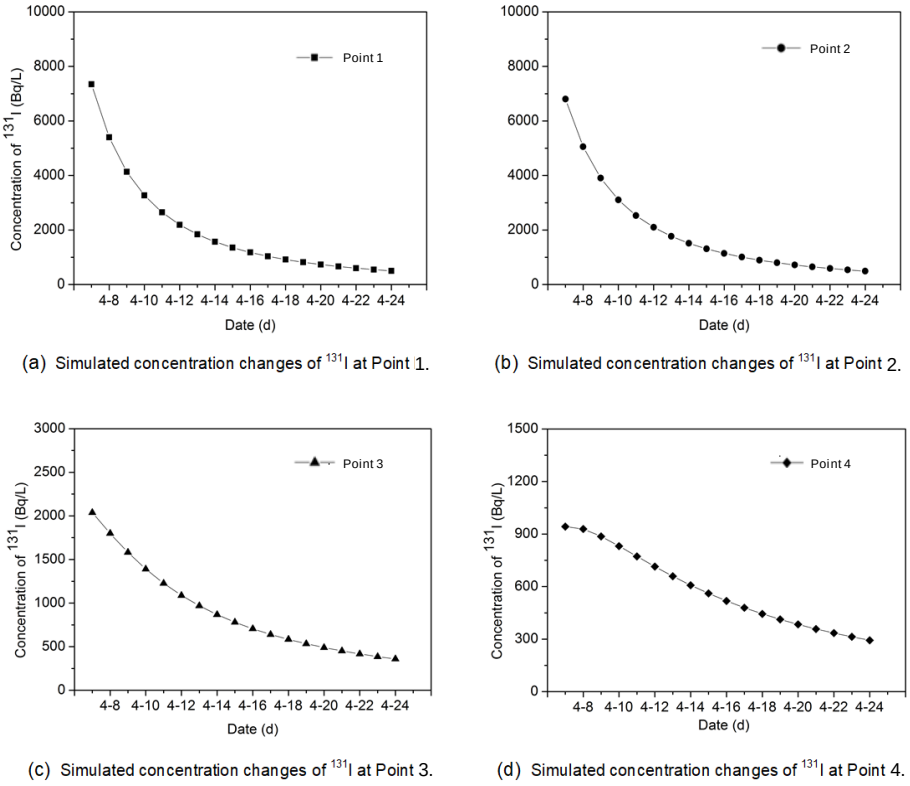
Table 4. Values of the Decay Factor k_i of ^{131}I and ^{137}Cs

Time		Decay factor k_i	
		^{131}I	^{137}Cs
April 7	-	1.00000	1.00000
April 8	1d	0.91988	0.99994
April 9	2d	0.84618	0.99987
April 10	3d	0.77838	0.99981
April 11	4d	0.71602	0.99974
April 12	5d	0.65865	0.99968
April 13	6d	0.60588	0.99962
April 14	7d	0.55734	0.99955
April 15	8d	0.51268	0.99949
April 16	9d	0.47161	0.99943
April 17	10d	0.43382	0.99936
April 18	11d	0.39907	0.99930
April 19	12d	0.36709	0.99924
April 20	13d	0.33768	0.99917
April 21	14d	0.31063	0.99911
April 22	15d	0.28574	0.99905
April 23	16d	0.26285	0.99898
April 24	17d	0.24179	0.99892

4. Results and Discussion

4.1. Simulation Results and Precision Analysis of the Instantaneous Point Source Model

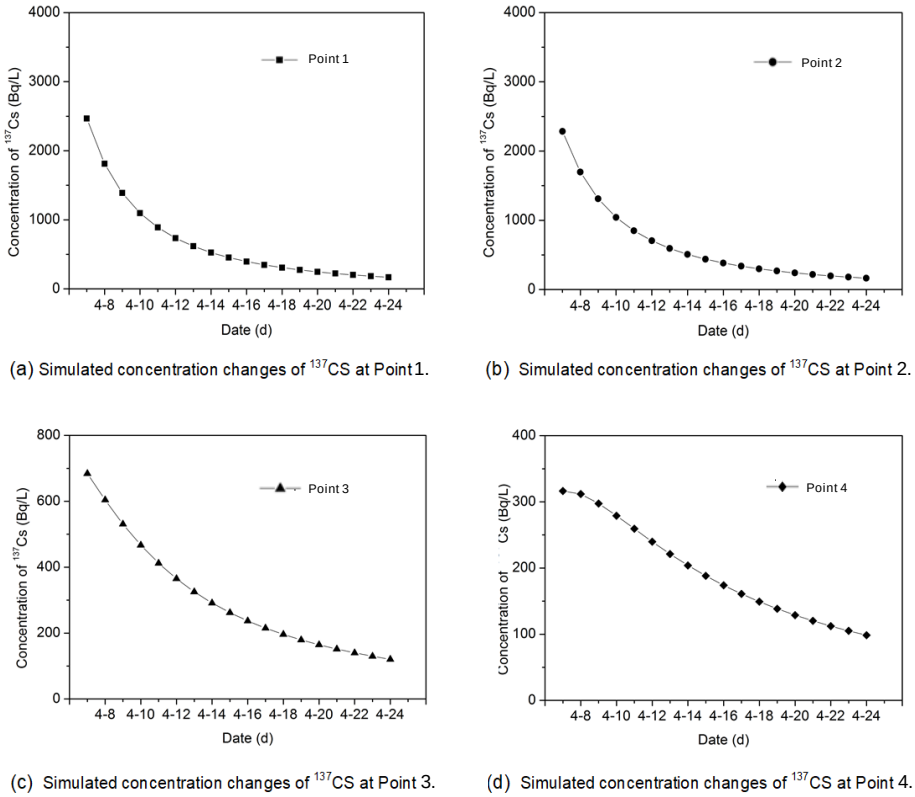
According to the instantaneous point source model and parameters mentioned above, the diffusion and decay processes of ^{131}I and ^{137}Cs for the research points from 7 to 24 April 2011 under the accident conditions were simulated with numerical analysis software. This study was carried out based on the following assumptions: horizontal diffusion played a major role in pollutant transportation in shallow sea area and ocean surface, as the length of shallow sea area in horizontal direction was much larger than that in vertical one; the velocity of water flow for each research point was constant given the ocean surface was calm; the concentrations of radionuclides did not change in the vertical direction in the sea under the condition that contaminants were mixed completely in the direction; diffusion of radioactive effluents was not affected by ocean currents, temperature and depth of ocean, or atmospheric deposition when the external environment was relatively stable; and adsorption and intake of radionuclides by marine plants and animals could be ignored since the research duration was relatively short. The concentration changes of ^{131}I and ^{137}Cs at the four



(a) Simulated concentration changes of ^{131}I at Point 1. (b) Simulated concentration changes of ^{131}I at Point 2.

(c) Simulated concentration changes of ^{131}I at Point 3. (d) Simulated concentration changes of ^{131}I at Point 4.

Figure 2. Simulated concentration changes of ^{131}I at the study points 1 to 4 using the instantaneous point source model.



(a) Simulated concentration changes of ^{137}CS at Point1. (b) Simulated concentration changes of ^{137}CS at Point 2.

(c) Simulated concentration changes of ^{137}CS at Point 3. (d) Simulated concentration changes of ^{137}CS at Point 4.

Figure 3. Simulated concentration changes of ^{137}Cs at the study points 1 to 4 using the instantaneous point source model.

Table 5. The Maximum Values of Simulated Concentrations for ^{131}I and ^{137}Cs at each Research Point

Radionuclide / Research point	^{131}I			^{137}Cs		
	Maximum value	Time	Times of exceeding standard	Maximum value	Time	Times of exceeding standard
1	7.47×10^3 Bq/L	7 April 2011	187	2.53×10^3 Bq/L	7 April 2011	28
2	6.91×10^3 Bq/L	7 April 2011	173	2.21×10^3Bq/L	7 April 2011	25
3	2.05×10^3 Bq/L	7 April 2011	51	6.95×10^2 Bq/L	7 April 2011	8
4	9.02×10^2 Bq/L	7 April 2011	23	3.10×10^2 Bq/L	7 April 2011	3

research points from 7 ~ 24 April 2011 were simulated and are presented in Figures 2 and 3, respectively. The instantaneous point source item Q of ^{131}I and ^{137}Cs resulted in the difference between in the half-life period between ^{131}I and ^{137}Cs.

Figures 2 and 3 show that concentrations of ^{131}I and ^{137}Cs decreased gradually with time at the four research points. In the initial days of the simulation period, the ^{131}I and ^{137}Cs concentrations were relatively high because of the previous discharge of the instantaneous point source item. In particular, the maximum value of concentration for ^{131}I was 7.47×10^3 Bq/L (7 April 2011, Point 1), which was 185 times greater than the regulatory limit in Japan, while that of ^{137}Cs was 2.53×10^3 Bq/L, exceeding the related limit by 28 times (7 April 2011, Point 1). Moreover, the maximum values of simulated concentrations for ^{131}I and ^{137}Cs at each research point were calculated as presented in Table 5.

As Points 1 and 2 were nearto the cracks of high level radioactive effluents in unit 2 of Fukushima Dai-ichi Nuclear Power Plant, concentrations of ^{131}I and ^{137}Cs in the two research points were both 10^3 order of magnitude, which far exceeded the water quality standard. The ^{131}I and ^{137}Cs concentrations reached the peak at points 1 and 2 on 7 April 2011. Moreover, the values of ^{131}I and ^{137}Cs were 6.91×10^3 Bq/L and 2.21×10^3 Bq/L respectively at Point 2, while those at Point 1 were greater than these values. At the same time, ^{131}I and ^{137}Cs concentrations at Points 3 and 4 were at the 10^2 order of magnitude, which is lower than the values of Points 1 and 2 as these two points were more distant from the Fukushima Dai-ichi Nuclear Power Plant. Figures 2 and 3 indicated that the concentrations of ^{131}I were much greater than those of ^{137}Cs at each research point, and the reason was the discharge amount of ^{131}I was larger than that of ^{137}Cs. Because of the diffusion and decay effects in the ocean, the radionuclide concentrations decreased considerably within one week at all research points. The comparison between monitoring concentrations and the results simulated by the instantaneous point source model for ^{131}I and ^{137}Cs concentrations at Points 1 to 4 is given in Figures 4 and 5, respectively.

The relative error (R_e) was used for accuracy analysis, and its computational formula was given as:

$$R_e = \frac{c_i - c_0}{c_0} \times 100\% \quad (8)$$

where R_e is the relative error of model simulation (%), C_i is the simulation concentration (Bq/L), and C_0 is the monitored concentration (Bq/L). If R_e is a positive value, the simulated concentration is relatively high, and vice versa. The model simulation results were considered in agreement with the monitoring values when R_e was 0. From Figures 4 and 5, relative errors of radionuclides between simulation and monitoring values in each research point were calculated as shown in Table 6.

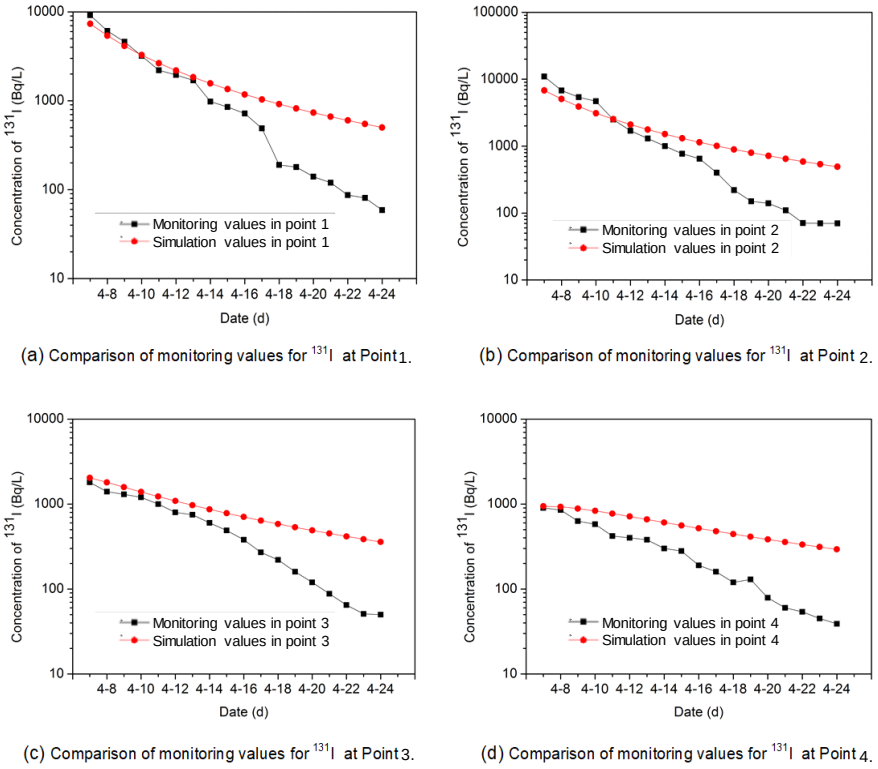
Figures 4 and 5 and Table 6 indicated that the maximum relative error between simulation and monitoring concentrations for ^{131}I at all research points during the simulation period was 678.71%. This was because that the simulated concentrations were higher than the monitored equivalents. The relative error increased over time, and reached its maximum value of 678.71% on the last day of the simulation period (April 23). Moreover, the average relative error of simulation for all research points was 200.07%, which indicated that it was difficult to obtain the desired simulation results for radionuclides with a short half-life period such as ^{131}I through instantaneous point source model during this simulation period. For ^{137}Cs, the relative errors during the whole simulation period were relatively small in each research point, and the average relative error was 34.09%. This showed that the instantaneous point source model had an acceptable accuracy for radionuclides with long half-life periods (such as ^{137}Cs) during this short simulation period.

In conclusion, the relative errors of concentration simulation for radionuclides with short half-life periods were larger than those with long half-life periods when simulating using the original model. This was because the decay of radionuclides was not taken into consideration during the diffusion clides was not taken into consideration during the diffusion process in the model.

4.2. Simulation Results and Precision Analysis of the Improved Instantaneous Point Source Model

Based on the improved instantaneous point source model and determined parameters, the spatial-temporal changes of ^{131}I and ^{137}Cs concentrations were calculated by ORIGIN at each research point from 7 ~ 24 April 2011 under the accident conditions. The initial assumptions were in accordance with those mentioned in Section 4.1.

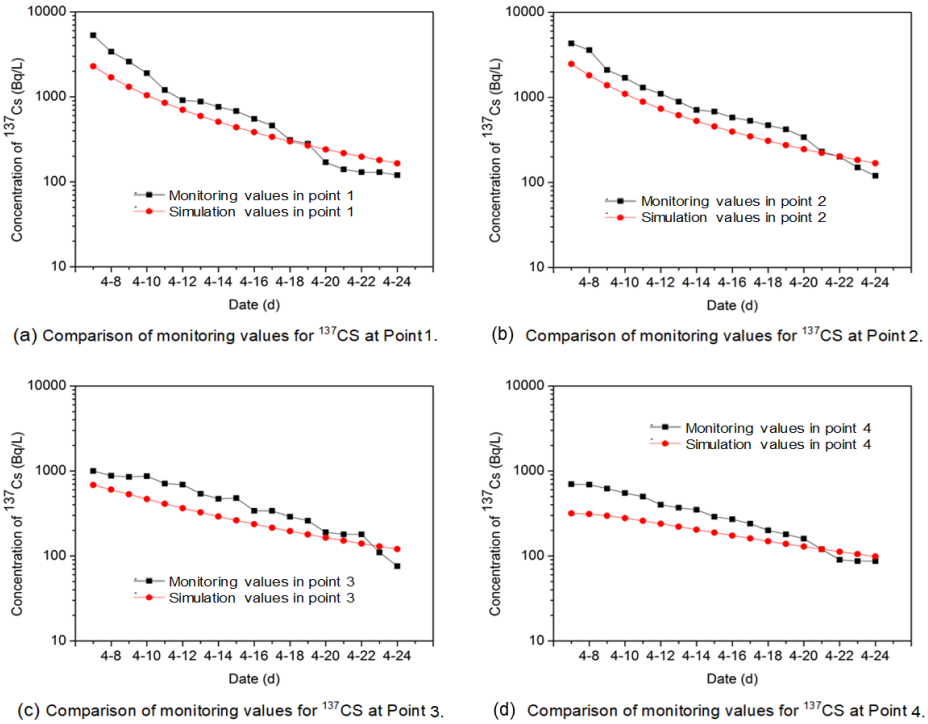
The simulation results indicated that concentrations of ^{131}I and ^{137}Cs at each research point decreased over time (Figures 6 and 7). Owing to the effects of migration and decay, concentrations of radionuclides decreased considerably at each research point within one week. Compared with the ^{137}Cs concentrations, the ^{131}I concentrations were much higher within one half-



(a) Comparison of monitoring values for ^{131}I at Point 1. (b) Comparison of monitoring values for ^{131}I at Point 2.

(c) Comparison of monitoring values for ^{131}I at Point 3. (d) Comparison of monitoring values for ^{131}I at Point 4.

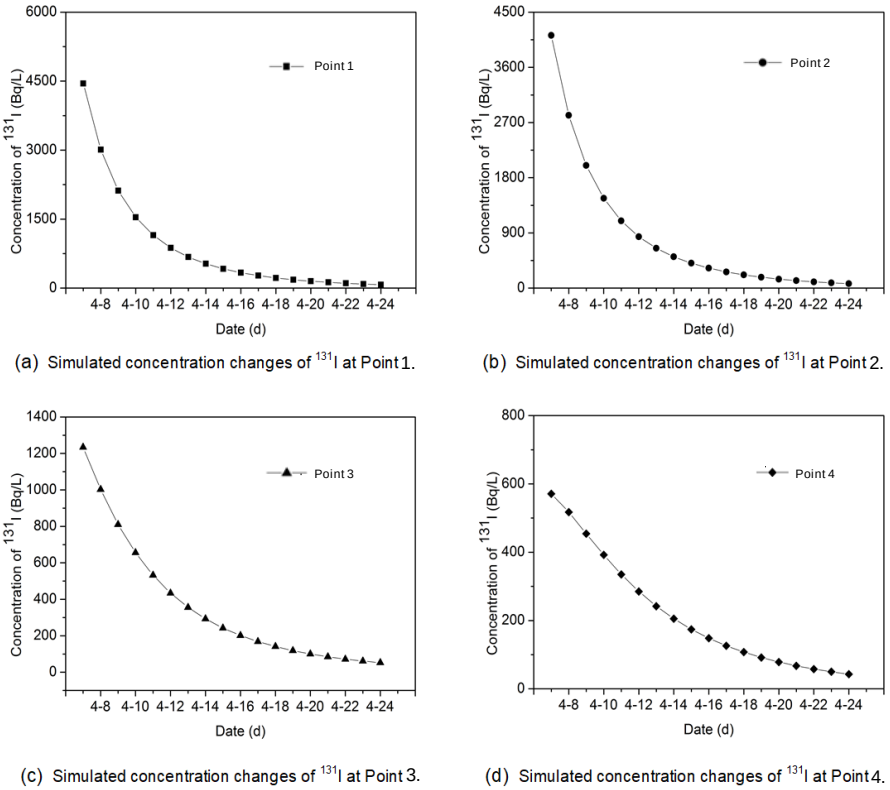
Figure 4. Comparison of monitoring values for ^{131}I at research points 1 to 4 with those simulated using the instantaneous point source model.



(a) Comparison of monitoring values for ^{137}CS at Point 1. (b) Comparison of monitoring values for ^{137}CS at Point 2.

(c) Comparison of monitoring values for ^{137}CS at Point 3. (d) Comparison of monitoring values for ^{137}CS at Point 4.

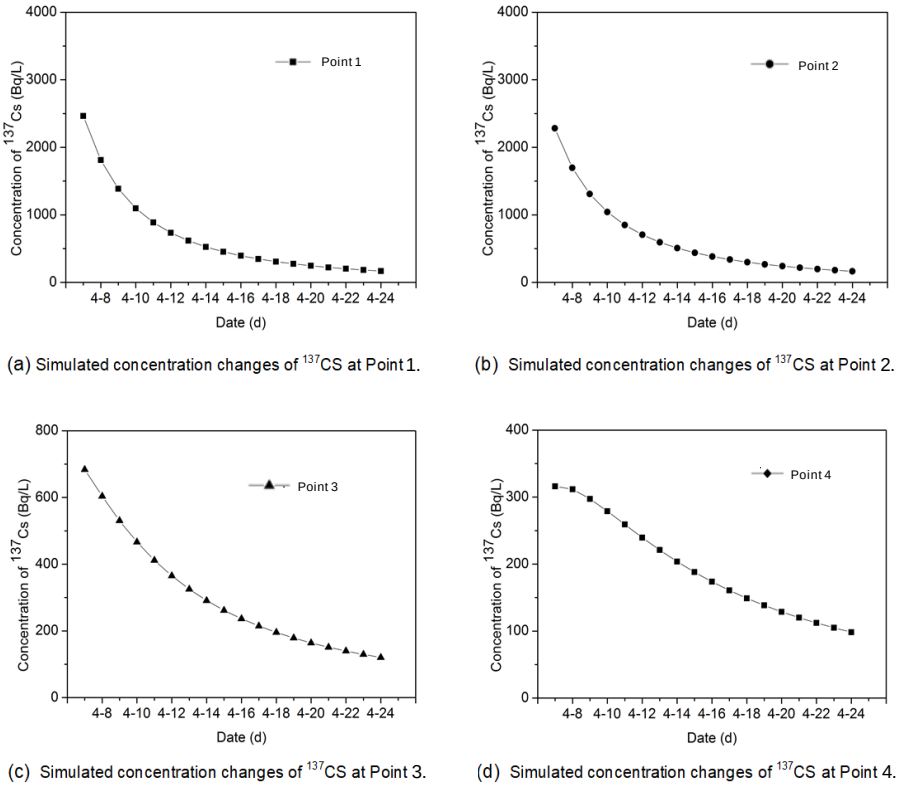
Figure 5. Comparison of monitoring values for ^{137}Cs at research points 1 to 4 with those simulated using the instantaneous point source model.



(a) Simulated concentration changes of ^{131}I at Point 1. (b) Simulated concentration changes of ^{131}I at Point 2.

(c) Simulated concentration changes of ^{131}I at Point 3. (d) Simulated concentration changes of ^{131}I at Point 4.

Figure 6. Simulated concentration changes of ^{131}I at research points 1 to 4 using the improved model.



(a) Simulated concentration changes of ^{137}CS at Point 1. (b) Simulated concentration changes of ^{137}CS at Point 2.

(c) Simulated concentration changes of ^{137}CS at Point 3. (d) Simulated concentration changes of ^{137}CS at Point 4.

Figure 7. Simulated concentration changes of ^{137}Cs at research points 1 to 4 using the improved model.

Table 6. Accuracy Analysis of Simulation Results for ^{131}I and ^{137}Cs Based on the Instantaneous Point Source Model (unit: %)

Research point	^{131}I			^{137}Cs		
	Maximum Relative Error	Minimum Relative Error	Average relative error	Maximum Relative Error	Minimum Relative Error	Average relative error
1	749.66	11.36	213.23	-49.67	-16.79	30.33
2	655.67	14.79	172.47	-56.91	-10.37	35.77
3	657.27	13.22	189.80	-58.49	-13.44	34.32
4	652.23	19.56	224.78	-60.29	-13.21	35.94
Average	678.71	14.73	200.07	-27.10	-13.45	34.09

life period (8.3 days), and there was a continuous decrease of ^{131}I concentration. Subsequently, ^{131}I and ^{137}Cs concentrations were both within the 10^2 order of magnitude at all research points. Two half-life periods of ^{131}I later, the ^{131}I concentrations were less than those of ^{137}Cs at all research points. Moreover, ^{131}I had the most serious impact on the nearby marine environment in the first half-life period and the impacts subsequently greatly reduced. In addition, the ^{137}Cs concentrations also decreased during the simulation period even though the range of this decrease was much smaller than that of ^{131}I. The effects of ^{137}Cs on water bodies could be not fully eliminated in a short period because of its long half-life and its leaching and release in vegetation and soil.

Furthermore, the simulated values of the improved instantaneous point source model for ^{131}I and ^{137}Cs at Points 1 to 4 were compared with the monitoring data (Figures 8 and 9). The monitoring data in Figures 8 and 9 came from relevant reports on radioactive levels of surrounding seas which were published by TEPCO, Ministry of Education, Culture, Sports, Science and Technology (MEXT), and IAEA (IAEA, 2011; MEXT, 2011; TEPCO, 2011). Meanwhile, the simulation values were based on the simulation of the improved instantaneous point source model.

According to Formula (8), the relative errors between the simulated and monitoring values at the research points were calculated (Table 7). The minimum and average relative errors for ^{131}I at all research points during the simulation period were 7.63 and 34.12% respectively, which indicated that acceptable simulation results could be obtained for radionuclides with a short half-life using the improved instantaneous point source model. Moreover, for ^{137}Cs, the relative errors during the simulation period were relatively small at all research points, and the average relative error was 33.44%. This illustrated that the improved instantaneous point source model also had an acceptable simulation accuracy for long half-life radionuclides.

In conclusion, simulated results from the improved instantaneous point source model, in which radionuclide decay was taken into account, were acceptable for both short and long half-life radionuclides.

4.3. Accuracy Comparison between the Original and Improved Models

The average relative errors for simulation of ^{131}I and ^{137}Cs concentrations at research points 1 to 4 were compared between the original and improved models (Table 8).

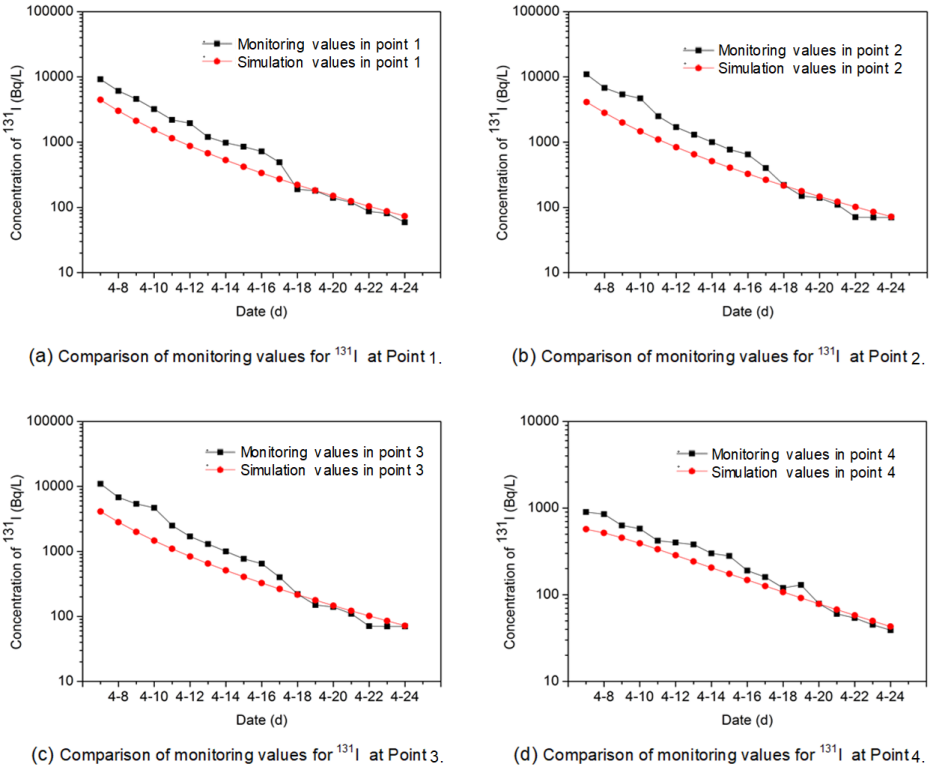
Through the model improvement, the relative errors of ^{131}I concentrations at research points 1 to 4 substantially reduced, and the simulation accuracy of ^{131}I enhanced considerably. Relative errors reduced by 178.24, 133.94, 155.88, and 195.73% for research points 1 to 4, respectively, and the average value decreased by 165.95%. This indicates that the improved instantaneous point source model could reflect the decay and diffusion of ^{131}I in sea water. Through considering the decay process of radionuclides, the accuracy of the simulation model was substantially improved for radionuclides with short half-life periods such as ^{131}I. However, the improvement was small for ^{137}Cs average relative errors, which reduced from 34.09 to 33.44%. This was because the simulation period was merely 18 days (7 ~ 24 April 2011), which was much shorter than the half-life period of ^{137}Cs (30.167 years) and the decay of ^{137}Cs over such a short time could therefore be neglected. If the simulation period was much longer (e.g. the half-life period of ^{137}Cs), the simulation accuracy increase would be obvious as the influence of radionuclide decay on radioactive concentration could be adequately reflected by the improvement model. This indicated that the improved instantaneous point source model was also suitable to simulate the transport and diffusion of radionuclides with long half-life periods such as ^{137}Cs in shallow marine areas, considering its acceptable simulation precision.

In conclusion, a good improvement in accuracy was found following application of the improved instantaneous point source model relative to the original model. Compared with the original model, the improved instantaneous point source model could more objectively reflect the decay and diffusion mechanism of radionuclides in ocean, and support a more precise simulation of spatiotemporal changes for radionuclides in oceans following nuclear power plant accidents.

5. Conclusions

In this study, an improved instantaneous point source model was developed for the simulation of radionuclide transport and diffusion in oceans following nuclear power plant accidents based on the additional consideration of the decay process of radionuclides. The original and improved models were applied to simulate the concentration changes of ^{131}I and ^{137}Cs with time over a period after the Fukushima Dai-ichi nuclear power plant accident (7 ~ 24 April 2011) at four research points. The simulation results from the original model were compared with those of the improved model.

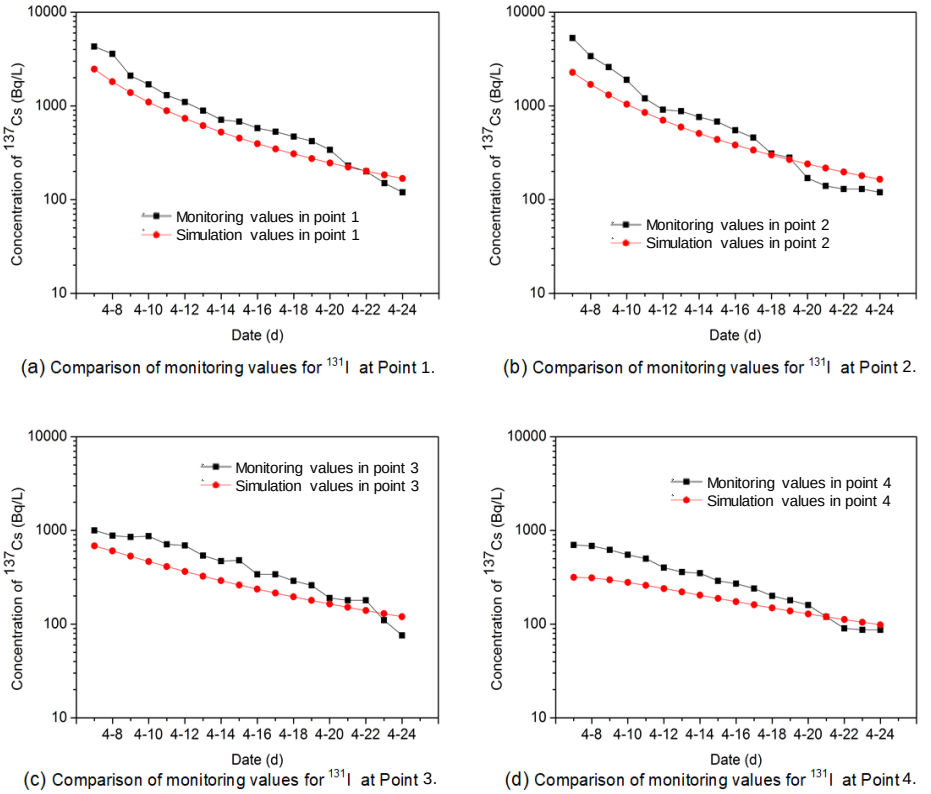
The simulated concentrations from the original model for



(a) Comparison of monitoring values for ^{131}I at Point 1. (b) Comparison of monitoring values for ^{131}I at Point 2.

(c) Comparison of monitoring values for ^{131}I at Point 3. (d) Comparison of monitoring values for ^{131}I at Point 4.

Figure 8. Comparison of ^{131}I concentrations measured at research points 1 to 4 and those simulated using the improved model.



(a) Comparison of monitoring values for ^{131}I at Point 1. (b) Comparison of monitoring values for ^{131}I at Point 2.

(c) Comparison of monitoring values for ^{131}I at Point 3. (d) Comparison of monitoring values for ^{131}I at Point 4.

Figure 9. Comparison of ^{137}Cs concentrations measured at research points 1 to 4 and those simulated using the improved model.

Table 7. Analysis of the Accuracy of Simulation Results for ^{131}I and ^{137}Cs Using the Improved Model (unit: %)

Research point	^{131}I			^{137}Cs		
	Maximum Relative Error	Minimum Relative Error	Average relative error	Maximum Relative Error	Minimum Relative Error	Average relative error
1	-55.20	-9.73	34.99	-49.65	-7.83	30.13
2	-68.84	-8.49	38.53	-56.93	-8.56	35.76
3	-52.57	-11.25	33.92	58.46	-13.41	34.29
4	-43.14	-7.63	29.05	-60.27	-11.75	33.95
Average	-54.94	-9.28	34.12	-27.10	-10.39	33.44

Table 8. Comparison of Average Relative Errors for Simulation Results between the Original and Improved Models (unit: %)

Research point	Average relative error (^{131}I)			Average relative error (^{137}Cs)		
	Instantaneous point source model	Improved model	Reduction of average relative error	Instantaneous point source model	Improved model	Reduction of average relative error
1	213.23	34.99	178.24	30.33	30.13	0.20
2	172.47	38.53	133.94	35.77	35.76	0.01
3	189.80	33.92	155.88	34.32	34.29	0.03
4	224.78	29.05	195.73	35.94	33.95	1.99
Average	200.07	34.12	165.95	34.09	33.44	0.56

^{131}I and ^{137}Cs at each research point were also compared with monitoring values. The maximum relative error between simulation and monitoring concentrations for ^{131}I at all research points during the simulation period was 749.66%. Moreover, the average relative error of simulation for all research points was 224.78%. For ^{137}Cs, the relative errors during the simulation period were relatively small at all research points, and the average relative error was 34.09%. The relative errors for ^{131}I were too large to be accepted, which was because its half-life was relatively short (8.3 days). During the simulation period, there was a decay process of ^{131}I, which was not taken into account in the original model. However, in the improved model, the minimum relative error of ^{131}I at all research points during the simulation period was -7.63% and the average relative error of ^{131}I concentrations at all research points reduced to 34.12%. Furthermore, the average relative error of ^{137}Cs was 33.44%. In addition, the average relative errors of ^{131}I and ^{137}Cs reduced from 200.07% and 34.09% to 34.12% and 33.44%, respectively, representing an overall reduction of 165.95% and 0.56%. This considerable decrease in the ^{131}I simulation accuracy was caused by the added consideration of the decay process. Moreover, the simulation accuracy of ^{137}Cs changed only slightly because the simulation period was much shorter than the half-life period of ^{137}Cs (30.167 years).

Acceptable simulation results can be obtained for simulation of the spatiotemporal change of radionuclides in oceans following nuclear power plant accidents through the improved instantaneous point source model. This study is the first attempt to take the decay of radionuclides into account when simulating transport process by diffusion equation for radionuclides in oceans following nuclear power plant accidents. It provides a useful tool for accident emergency response under the conditions that source of pollution was instantaneous point one and that the contaminated areas were ocean, neritic region as well as oceanic zone. In this study, the simulation was based on the instantaneous point source model, which had some limitations the directions of wind and tide were not taken into account. In the future, this concern will be considered further to realize more accurate simulation.

Acknowledgments. This research was supported by the National Key Research and Development Plan (2016YFA0601502, 2016YFC0502-800), the National key scientific and technological projects of the PRC [2014ZX07104005], National Key Research and Development Plan [2016YFA0601502], and the Fundamental Research Funds for the Central Universities of PRC [2017YQ002]. We also thank the editor and the anonymous reviewers for their critical and valuable comments that have led to the improvement in this paper.

References

Andrews, K.W., Roseland, J.M., Gusev, P.A., Palachuvattil, J., Dang, P.T., Savarala, S., Han, F., Pehrsson, P.R., Douglass, L.W., Dwyer, J.T., Betz, J.M., Saldanha, L.G. and Bailey, R.L. (2016). Analytical ingredient content and variability of adult multivitamin/mineral products: national estimates for the Dietary Supplement Ingredient Database. *Am. J. Clin. Nutr.*, 105 (2), 526-539. https://doi.org/10.3945/ajcn.116.134544

Batlle, J.V.I., Beresford, N.A., Beaugelin-Seiller, K., Bezhenar, R., Brown, J., Cheng, J.J., Cujic, M., Dragovic, S., Duffa, C. and Fievet, B. (2016). Inter-comparison of dynamic models for radionuclide transfer to marine biota in a Fukushima accident scenario. *J. Environ. Radioactiv.*, 153, 31-50. https://doi.org/10.1016/j.jenvrad.2015.12.006

Blokhintsev, L.D. and Savin, D.A. (2016). Analytic continuation of scattering data to the region of negative energies for systems that have one and two bound states. *Phys. Atom. Nucl*., 79 (3), 358-369. https://doi.org/10.1134/S1063778816030066

Brown, C. and Milke, M. (2016). Recycling disaster waste: feasibility, method and effectiveness. *Resour. Conserv. Recycl.*, 106, 21-32. https://doi.org/10.1016/j.resconrec.2015.10.021

Buesseler, K., Aoyama, M. and Fukasawa, M. (2011). Impacts of the Fukushima nuclear power plants on marine radioactivity. *Environ. Sci. Technol*., 45 (23), 9931-5. https://doi.org/10.1021/es202816c

de Téramond, G.F. (2016). The spectroscopy and form factors of nucleon resonances from superconformal quantum mechanics and

holographic QCD. *Few-Body. Syst*., 57 (10), 925-932. https://doi.org/10.1007/s00601-016-1129-6

du Bois, P.B., Laguionie, P., Boust, D., Korsakissok, I., Didier, D. and Fiévet, B. (2012). Estimation of marine source-term following Fukushima Dai-ichi accident. *J. Environ. Radioactiv*., 114 (12), 2-9. https://doi.org/10.1016/j.jenvrad.2011.11.015

Guo, J.H., Huang, G.H., Wang, X.Q., Wu, Y.H., Li, Y.P., Zheng, R.B., and Song, L.M. (2020). Evaluating the added values of regional climate modeling over China at different resolutions. *Sci Total Environ.*, 718, 137350. https://doi.org/10.1016/j.scitotenv.2020.137350

Harper, E.M., Diao, Z., Panousi, S., Nuss, P., Eckelman, M.J. and Graedel, T.E. (2015). The criticality of four nuclear energy metals. *Resour. Conserv. Recycl*., 95, 193-201. https://doi.org/10.1016/j.resconrec.2014.12.009

Huang, G. H., Baetz, B. W., Patry, G. G., and Terluk, V. (1997). Capacity planning for an integrated waste management system under uncertainty: a North American case study. *Waste Manag. & Res.*, 15(5), 523-546. https://doi.org/10.1006/wmre.1996.0106

Inomata, Y., Aoyama, M., Tsubono, T., Tsumune, D. and Hirose, K. (2015). Spatial and temporal distributions of Cs-134 and Cs-137 derived from the TEPCO Fukushima Daiichi Nuclear Power Plant accident in the North Pacific Ocean by using optimal interpolation analysis. *Environ. Sci. Process. Impact*., 18 (1), 126-136. https://doi.org/10.1039/C5EM00324E

International Atomic Energy Agency (IAEA). IAEA International fact finding expert mission of the Fukushima Dai-ichi NPP accident following the great east Japan earthquake and tsunami.2011. https://www.mendeley.com/research-papers/expert-mission-iaea-international-fact-finding-expert-mission-fukushima-daiichi-npp-accident-followi/

Jankovic, I., Maghrebi, M., Fiori, A. and Dagan, G. (2016). When good statistical models of aquifer heterogeneity go right: the impact of aquifer permeability structures on 3d flow and transport. *Adv. Water. Resour*., 100, 199-211. https://doi.org/10.1016/j.advwatres.2016.10.024

Kitamura, A., Kurikami, H., Yamaguchi, M., Oda, Y., Saito, T., Kato, T., Niizato, T., Iijima, K., Sato, H., Yui, M., Machida, M., Yamada, S., Itakura, M., Okumura, M. and Onishi, Y. (2015). Mathematical modeling of radioactive contaminants in the Fukushima environment. *Nucl. Sci. Eng*., 179 (1), 104-118. https://doi.org/10.13182/NSE13-89

Konstantinova, M., Lukšienė, B., Tarasiuk, N. and Maceika, E. (2015). Estimation of Cs isotope accumulation by litter, moss and grass in Lithuania attributed to the Fukushima Daiichi NPP accident. *J. Geochem. Explor*., 174, 159-163. https://doi.org/10.1016/j.gexplo.2015.11.001

Lee, K.H., Kim, K.H., Lee, J.H., Yun, J.Y. and Kim, C.H. (2015). Modeling of long range transport pathways for radionuclides to Korea during the Fukushima Dai-ichi nuclear accident and their association with meteorological circulations. *J. Environ. Radioactiv*., 148, 80-91. https://doi.org/10.1016/j.jenvrad.2015.06.007

Liu, X.C, Huang, G.H., and Chen, J.P. (2020). The development of inexact dual-objective programming for regional energy systems planning in Guang-Fo-Zhao region, China. *J. Cleaner Prod.*, 265, 121351. https://doi.org/10.1016/j.jclepro.2020.121351

Maqsood, I., Huang, G., Huang, Y., and Chen, B. (2005). ITOM: an interval-parameter two-stage optimization model for stochastic planning of water resources systems. *Stoch. Env. Res. and Risk A.*, 19(2), 125-133. https://doi.org/10.1007/s00477-004-0220-6

Matsui, T. (2011). Deciphering the measured ratios of iodine-131 to cesium-137 at the Fukushima reactors. *Prog. Theor. Phys*., 126 (6), 1167-1176. https://doi.org/10.1143/PTP.126.1167

Ministry of Education, Culture, Sports, Science and Technology (MEXT). 2011. http://www.mext.go.jp/a_menu/saigaijyouhou/syousai/1303856.htm and http://www.mext.go.jp/component/a_menu/other/detail/icsFiles/afieldfile/2011/07/29/1306949_072914.pdf.

Monte, L. (2014). Application of the migration models implemented in the decision system MOIRA-PLUS to assess the long term behaviour of Cs-137 in water and fish of the Baltic Sea. *J. Environ. Radioactiv*., 134 (2), 136-144. https://doi.org/10.1016/j.jenvrad.2014.03.009

Nosov, A.V. (2005). Method for using measurements of the contamination of bottom deposits to estimate the source of radioactive substances flowing into a river. *Atom. Energy*., 99 (3), 658-664. https://doi.org/10.1007/s10512-005-0262-y

Periáñez, R., Abril, J.M. and García-León, M. (1996). Modelling the dispersion of non-conservative radionuclides in tidal waters-Part 2: Application to 226 Ra dispersion in an estuarine system. *J. Environ. Radioactiv*., 31 (3), 253-272. https://doi.org/10.1016/0265-931X(95)00050-K

Prants, S.V., Uleysky, M.Y. and Budyansky, M.V. (2011). Numerical simulation of propagation of radioactive pollution in the ocean from the Fukushima Dai-ichi nuclear power plant. *Dokl. Earth. Sci*., 439 (2), 1179-1182. https://doi.org/10.1134/S1028334X11080277

Preller, R.H. and Cheng, A. (1999). Modeling the transport of radioactive contaminants in the arctic. *Mar. Pollut. Bull*., 8 (2), 71-91. https://doi.org/10.1016/S0025-326X(99)80001-0

Regmi, D. (2017). A regularity criterion for two and half dimensional magnetohydrodynamic equations with horizontal dissipation and horizontal magnetic diffusion. *Math. Method. Appl. Sci*., 40, 1497-1504. https://doi.org/10.1002/mma.4073

Sudbrock, F., Schomäcker, K. and Drzezga, A. (2016). The effectiveness of wastewater treatment in nuclear medicine: performance data and radioecological considerations. *J. Environ. Radioactiv*., 166, 202-207. https://doi.org/10.1016/j.jenvrad.2016.04.002

Taelman, S.E., De Meester, S., Schaubroeck, T., Sakshaug, E., Alvarenga, R.A.F. and Dewulf, J. (2014). Accounting for the occupation of the marine environment as a natural resource in life cycle assessment: an exergy based approach. *Resour. Conserv. Recycl*., 91, 1-10. https://doi.org/10.1016/j.resconrec.2014.07.009

Takeda, M., Yamauchi, M., Makino, M., and Owada, T. (2011). Initial effect of the Fukushima accident on atmospheric electricity. *Geophys. Res. Lett*., 38 (15), 98-106. https://doi.org/10.1029/2011GL048511

Tokyo Electric Power Company (TEPCO). Fukushima Dai-ichi Nuclear Power Station Unit 2: Countermeasures to stop the outflow of contaminated water and the water amount flowed out into the sea. http://www.tepco.co.jp/en/press/corp-com/release/11042103-e.html (accessed Apr 21, 2011)

Tsumune, D., Tsubono, T., Aoyama, M. and Hirose, K. (2012). Distribution of oceanic 137Cs from the Fukushima Dai-ichi nuclear power plant simulated numerically by a regional ocean model. *J. Environ. Radioactiv*., 111, 100-108. https://doi.org/10.1016/j.jenvrad.2011.10.007

Walan, P., Davidsson, S., Johansson, S. and Höök, M. (2014). Phosphate rock production and depletion: regional disaggregated modeling and global implications. *Resour. Conserv. Recycl*., 93, 178-187. https://doi.org/10.1016/j.resconrec.2014.10.011

Wang, T.C., Wang, J.R., Lin, H.T., Chen, H.C., Chen, S.W., Shih, C., Chiang, S.C. and Yu, T.Y. (2017). The ultimate response guideline simulation and analysis using TRACE, MAAP5, and FRAPTRAN for the Chinshan nuclear power plant. *Ann. Nucl. Energy*, 103, 402-411. https://doi.org/10.1016/j.anucene.2017.01.036

Weng, S.Q., Huang, G.H., and Li, Y.P. (2010). An integrated scenario-based multi-criteria decision support system for water re- sources management and planning–A case study in the Haihe River Basin. *Expert Syst. with Appl.*, 37(12), 8242-8254. https://doi.org/10.1006/wmre.1996.0106

Yao, Y., Huang, G., An, C.J., Chen, X.J., Zhang, P., Xin, X.Y., Shen, J., and Agnew, J. (2020). Anaerobic digestion of livestock manure in cold regions: Technological advancements and global impacts. *Renew. Sust. Energ. Rev*., 119, 109494. https://doi.org/10.1016/j.

rser.2019.109494

Zhai, M.Y., Huang, G.H., Liu, L.R., Zheng, B.Y., and Guan, Y.R. (2020). Inter-regional carbon flows embodied in electricity transmission: network simulation for energy-carbon nexus. *Renew. Sust. Energ. Rev*., 118, 109511. https://doi.org/10.1016/j.rser.2019.109511

Zheng, S.G., Huang, G.H., Zhou, X., and Zhu, X.H. (2020). Climate-change impacts on electricity demands at a metropolitan scale: A case study of Guangzhou, China. *Appl. Energy*, 261, 114295. https://doi.org/10.1016/j.apenergy.2019.114295