Numerical Modeling of the Releases of ^{90}Sr from Fukushima to the Ocean: An Evaluation of the Source Term

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ABSTRACT: A numerical model consisting of a 3D advection/diffusion equation, including uptake/release reactions between water and sediments described in a dynamic way, has been applied to simulate the marine releases of ^{90}Sr from the Fukushima power plant after the March 2011 tsunami. This is a relevant issue since ^{90}Sr releases are still occurring. The model used here had been successfully applied to simulate ^{137}Cs releases. Assuming that the temporal trend of ^{90}Sr releases was the same as for ^{137}Cs during the four months after the accident simulated here, the source term could be evaluated, resulting in a total release of 80 TBq of ^{90}Sr until the end of June, which is in the lower range of previous estimates. Computed vertical profiles of ^{90}Sr in the water column have been compared with measured ones. The ^{90}Sr inventories within the model domain have also been calculated for the water column and for bed sediments. Maximum dissolved inventory (obtained for April 10th, 2011) within the model domain results in about 58 TBq. Inventories in bed sediments are 3 orders of magnitude lower than in the water column due to the low reactivity of this radionuclide. ^{90}Sr/^{137}Cs ratios in the ocean have also been calculated and compared with measured values, showing both spatial and temporal variations.
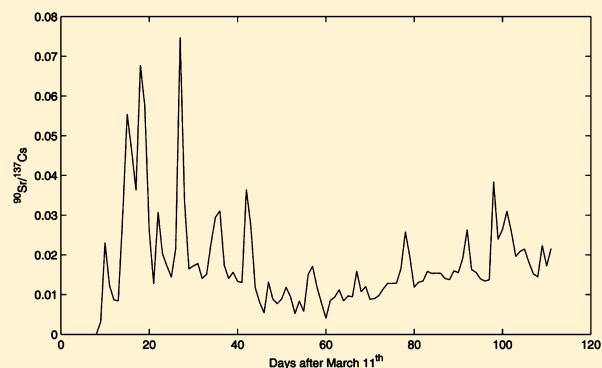
1. INTRODUCTION

As a consequence of the March 11th 2011 tsunami in Japan, significant amounts of anthropogenic radionuclides were released to the atmosphere and the ocean. Extensive measurements of concentrations of Cs isotopes (^{137}Cs and ^{134}Cs) in Japan coastal waters were made relatively soon after the accident (see for instance ref 1). From these sets of measurements, and also using numerical dispersion models, the ^{137}Cs source term to the marine environment due to the direct discharge of coolant waters could be evaluated.[2−5]

The geochemical behavior of Sr is similar to that of Ca. Thus, ^{90}Sr, which is generated in nuclear reactors, is a significant radionuclide from a radiological point of view, since it is accumulated in bones. Nevertheless, few measurements of ^{90}Sr have been made in marine waters off Fukushima after the 2011 tsunami[6,7] in comparison with those carried out for Cs isotopes. This is

partially due to the complexity of the involved radiochemical procedures.[7] These measurements have provided ^{90}Sr concentrations 2 orders of magnitude larger than the background levels (1.2 Bq/m^3). Also, they have pointed out that direct releases to the sea have been much more significant than atmospheric releases (and eventual deposition on land and ocean) for this radionuclide.

The objective of this paper consists of analyzing for the first time, using numerical modeling, the dispersion of ^{90}Sr in the coastal area around Fukushima. Indeed, several papers have already been published in which ^{137}Cs[3,8−14] and ^{131}I[4] dispersion

in the sea is studied by means of numerical models. But there are not equivalent studies for the case of ^{90}Sr.

The model used in this paper has been recently applied with success to simulate ^{137}Cs releases[15] and also to investigate the fate of potential direct releases of plutonium to the sea.[16] It consists of a 3D advection/diffusion equation with terms that describe the interactions of dissolved radionuclides with sediments. Uptake/release processes are described in a dynamic way, using kinetic transfer coefficients. Water circulation was obtained from the JCOPE2 (Japan Coastal Ocean Predictability Experiment 2) hydrodynamic model.[17]

Calculated ^{90}Sr concentrations in the water column have been compared with measured concentrations. This has been done for surface samples and also for several vertical profiles. The model has allowed to carry out an evaluation of the total release of ^{90}Sr to the marine environment until the end of June 2011, which has been compared with previous estimates. These were based on the ^{137}Cs release and on values for the ^{90}Sr/^{137}Cs ratio in the effluent. Inventories of ^{90}Sr in the water column and bed sediments have been evaluated as well.

This work may constitute a baseline for future assessments on the discharges of ^{90}Sr to the sea, since these have continued months after the tsunami. For instance, an accidental discharge has been reported in December 2011.[6,7] The model shall allow future comparisons of calculated ^{90}Sr concentrations with new measurements and, hence, reconstruction of the source term after June 2011.

The model is very briefly described in section 2, since it has already been published.[15] Assumptions made to simulate ^{90}Sr are also described in this section. Next, model results are presented and discussed.

2. MODEL DESCRIPTION

The dispersion model (based on previous work[15]) consists of a 3D advection/diffusion equation with terms describing the adsorption/desorption reactions between the deepest water layer, in contact with the seabed, and bed sediments. Suspended matter particles, able to scavenge elements from the water column to the seabed, are not considered due to the extremely low reactivity of Sr.[18]

Uptake/release processes are formulated in a dynamic way, in terms of kinetic transfer coefficients. A detailed description of such formulation may be seen elsewhere.[19−22] The kinetic model considers that exchanges of radionuclides between water and sediments are governed by a first-order reversible reaction, where k_1 and k_2 are the forward and backward rates, respectively.

The adsorption process is a surface phenomenon that depends on the surface of particles per water volume unit into the grid cell. This quantity has been denoted as the exchange surface.[19−22] Thus:

$$k_1 = \chi S \tag{1}$$

where S is the exchange surface (dimensions $[L]^{-1}$) and χ is a parameter with the dimensions of a velocity. It is denoted as the exchange velocity. Assuming spherical particles, the exchange surface is written as (see references cited above):

$$S = \frac{3Lf(1-p)\phi}{RH} \tag{2}$$

where R is the particle radius, p is the sediment porosity, and ϕ is a correction factor that takes into account that part of the sediment particle surface may be hidden by other sediment

particles. L is the sediment mixing depth (thickness of the sediment layer which interacts with water above it), H is the thickness of the deepest water layer (in contact with the sediment), and f gives the fraction of fine (muddy) sediment particles. This is required since the transfer of radionuclides to sediments is essentially due to such small particles. This formulation has been successfully used in all modeling works cited above. Full model equations in a 3D form may be seen elsewhere.[20] A numerical solution is carried out using second-order accuracy finite difference schemes.

Water currents are required to solve the advective terms in the dispersion equation mentioned above. Daily averaged three-dimensional wind and density driven currents have been obtained from the JCOPE2 model[17] in the time frame from March 12th to June 30th. The area studied extends from 140.46° to 142.04° longitude and from 35.96° to 39.54° latitude (Figure 1).
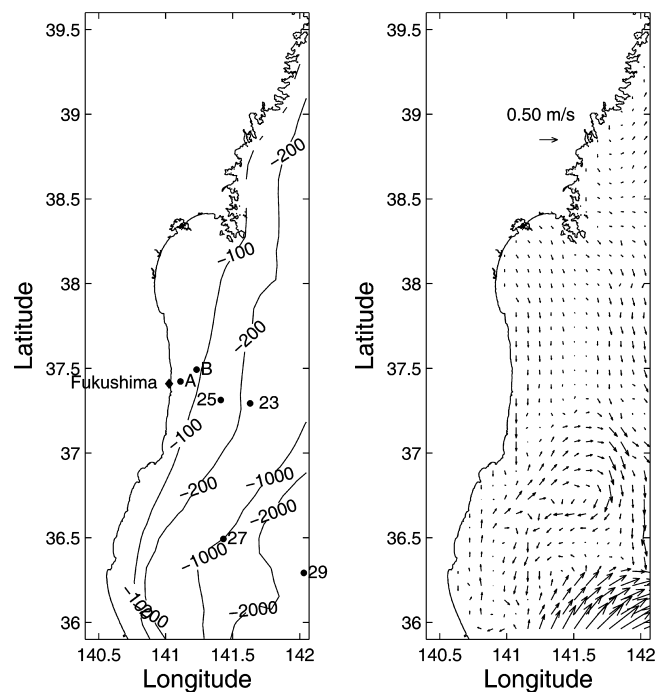

Figure 1. Left: model domain. Bathymetric contours are given in m, and sampling stations are indicated. Right: surface currents produced by the JCOPE2 model for March 12th.

There are 23 vertical levels, and the spatial resolution is 5 min of arc.[17] It was found that tides do not have a significant effect in dispersion patterns;[15] thus they are not included. As an example, surface currents corresponding to March 12th may be seen in Figure 1. The Kuroshio current—flowing to the NE—is clearly appreciated. Currents are weak in the area of Fukushima and flow southwards. To the south of Fukushima an anticyclonic eddy can also be identified.

The following assumptions are made to simulate ^{90}Sr dispersion in coastal waters off Fukushima:

1. The same temporal trend of direct releases as for ^{137}Cs is considered. This approach was already used to simulate the fate of potential plutonium releases.[16] It has also been applied to discuss measured ^{90}Sr concentrations in these waters.[7]
2. Releases of ^{90}Sr to the atmosphere, and subsequent deposition on the sea surface, are negligible. This is due to the low volatility of this radionuclide.[6,7] Thus, the deposition
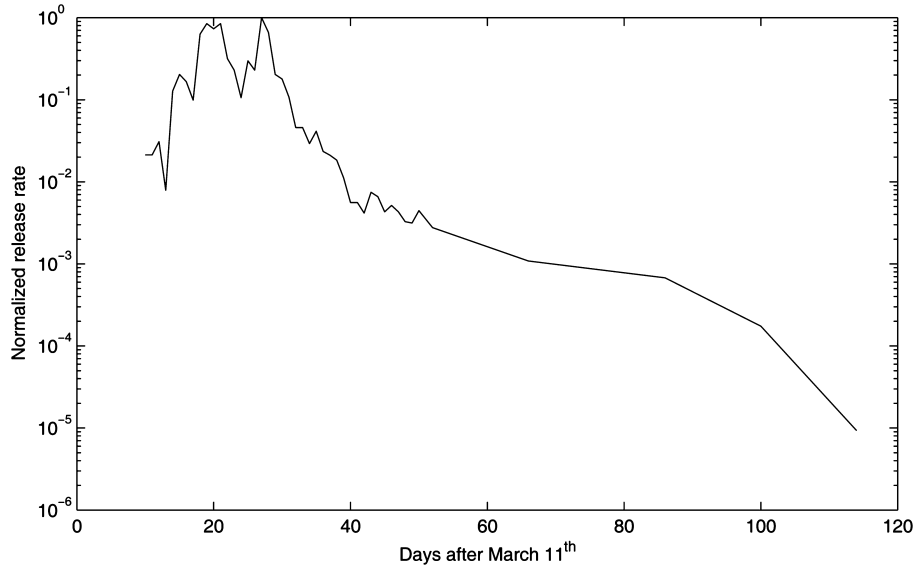
Figure 2. Normalized ^{137}Cs release rates obtained from the source term used in Min et al.[9] simulations. The ^{90}Sr release is obtained multiplying these rates by factor F (see text).
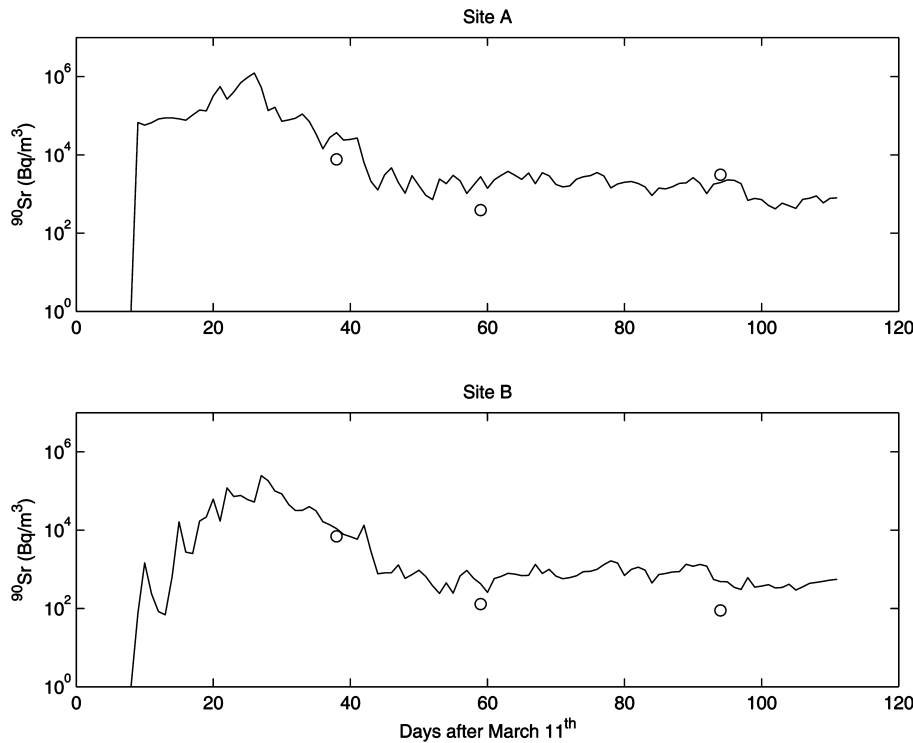

Figure 3. Calculated (lines) and measured (points) ^{90}Sr concentrations in surface water in sites indicated in Figure 1.

of ^{90}Sr from the atmosphere is negligible compared to the direct releases. The same was found in local-scale simulations of ^{137}Cs dispersion.[15]

Model parameter values for previous ^{137}Cs and plutonium applications have already been described.[15,16] In particular, values for the kinetic rates in the water/sediment interaction model are required. As widely described before,[19−22] the same value may be used for k_2 for different elements since the main parameter controlling pollutant geochemical behavior is the exchange velocity, χ. The exchange velocity may be deduced

from k_2 and the radionuclide equilibrium distribution coefficient k_d since the following relation holds (see the same references):

$$k_d = \frac{\chi}{k_2} \frac{3}{\rho R}$$

(3)

where ρ and R are particle density and radius, respectively. The value $k_2 = 1.16 \times 10^{-5}$ s^{-1} has been fixed. It has been successfully used in earlier simulations[19] for Cs and Pu, heavy metals,[20] and in Fukushima for Cs and Pu.[15,16] It was measured for Cs by Nyffeler et al.[23]
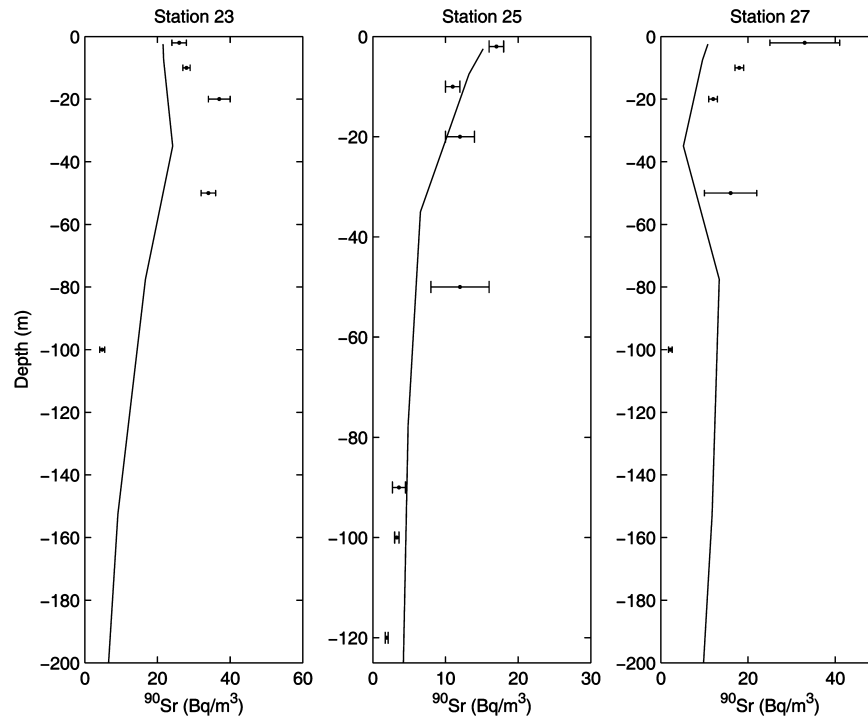
Figure 4. Calculated (lines) and measured (points) ^{90}Sr concentration vertical profiles at stations indicated in Figure 1.
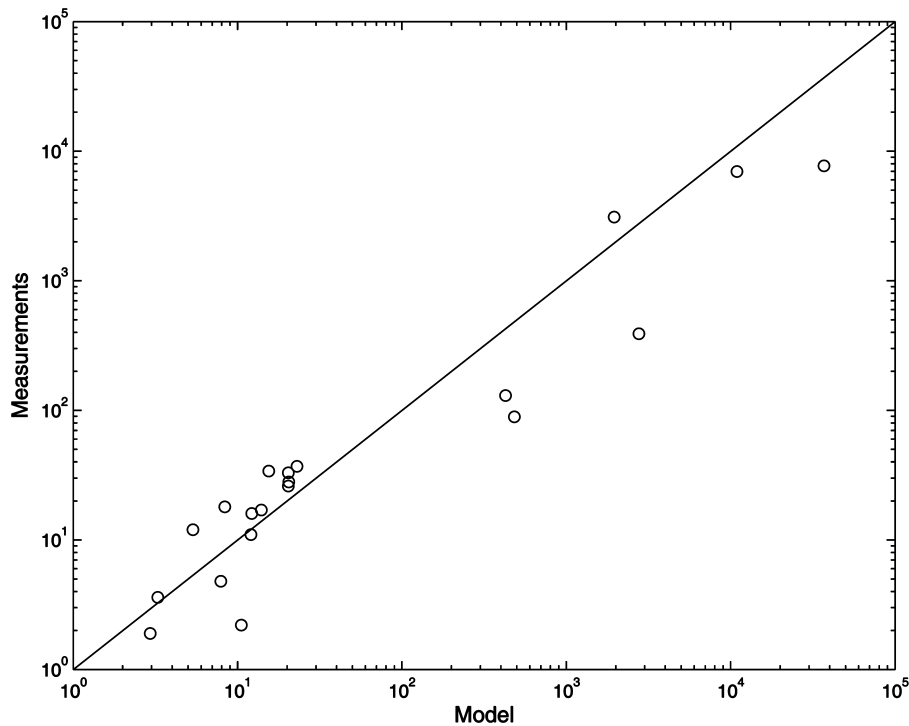

Figure 5. Measured vs calculated ^{90}Sr concentrations (Bq/m^3) for all samples. A line with the unit slope is also drawn.

Sr is more conservative than Cs, as indicated by their respective distribution coefficients, k_d's, in marine coastal waters: 8 and 4×10^3 L/kg (recommended values by IAEA[18]). This very low distribution coefficient will imply corresponding low inventories in the bottom sediments. Such IAEA recommended k_d for Sr has been used. Although this parameter is site-specific, there is no information for Japan coastal waters. Moreover, the adsorption of Sr on bed sediments will be

extremely low in any case. This equilibrium k_d is the only new parameter to be specified for this ^{90}Sr application. It must be pointed out that, in spite of using an equilibrium k_d as input parameter, water/sediment interactions are described in a dynamic way.

The temporal evolution of ^{137}Cs direct releases to the sea described in Min et al.[9] has been used. However, these release rates have been normalized to their maximum value.
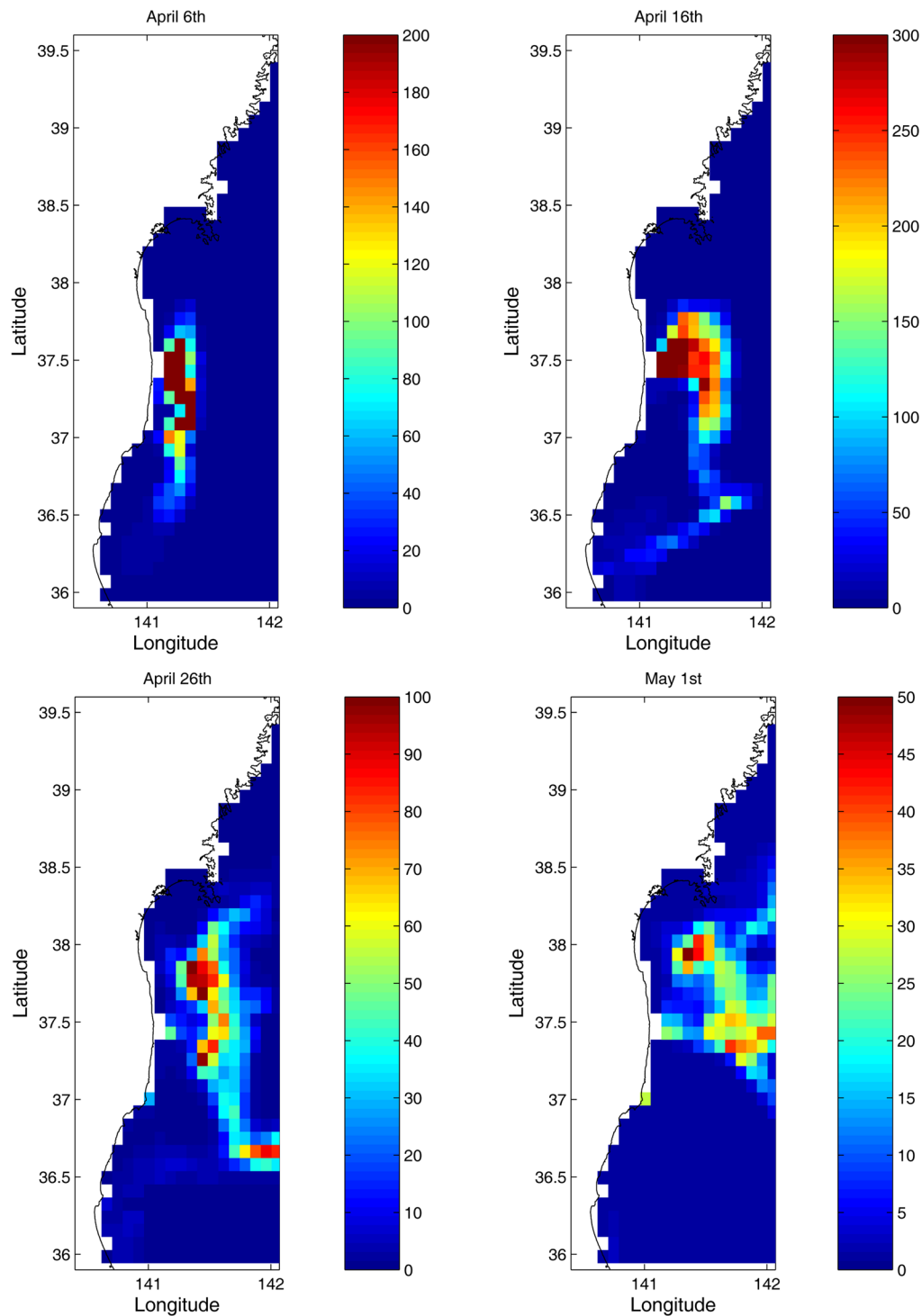
Figure 6. Calculated ^{90}Sr concentrations in surface water (Bq/m^3) at different times after the accident. Note the different scales in color bars.

The normalized release is presented in Figure 2. Releases of ^{137}Cs peaked on April 7th. The actual maximum release rate this date (day 27 in the graphic) is 7.17×10^{14} Bq/h.

Then, calculated ^{90}Sr concentrations in seawater are multiplied by a constant factor F which would provide the best possible agreement between measured and calculated concentrations. Real ^{90}Sr releases are given by the normalized sequence multiplied by F. This procedure is similar to the previously used to estimate radionuclide release rates from Fukushima to the atmosphere.[24]

3. RESULTS AND DISCUSSION

^{90}Sr concentrations have been measured[6] in surface water near the discharging channel from Fukushima and also 15 km offshore (sites A and B, respectively, in Figure 1). Actually, site A results used in this paper are mean values between measurements in sites 1 and 2 in Povinec's paper,[6] since the spatial resolution of the model does not allow to distinguish both points. Similarly, site B results are mean values between measurements in sites 7 and 8 in ref 6. Also, vertical profiles of ^{90}Sr concentrations in the water column have been reported.[7]
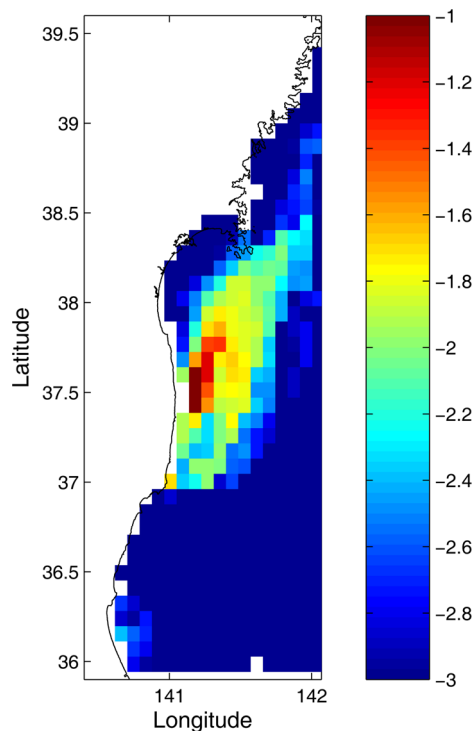
Figure 7. Calculated ^{90}Sr distribution in bed sediments (Bq/kg) for May 26th. Logarithms of calculated concentrations are drawn to appreciate differences.

Sampling locations are identified as points 23, 25, 27, and 29 in Figure 1.

Measurements of ^{90}Sr in surface waters (sites A and B, and surface samples of the vertical profiles) have been used to determine the factor F by which calculated concentrations resulting from the normalized release rates are multiplied to obtain the best possible agreement with measurements. The quadratic distances between measurements and the corresponding calculated concentrations are minimized to obtain F.

A comparison between model calculations and measured concentrations for sites A and B is shown in Figure 3. It can be appreciated that, once F has been determined, predicted ^{90}Sr surface concentrations for both sites are in agreement with measurements. The fact that the temporal trends of ^{90}Sr concentrations in both sites are adequately reproduced by the model supports assumption 1 in section 2.

Casacuberta et al.[7] have measured ^{90}Sr vertical profiles in the water column at some locations indicated in Figure 1 and at specific dates. These measured vertical profiles have been compared with the calculated by the model. Model results are extracted at the same date when samples were collected. Measured and calculated profiles are presented in Figure 4. Model results and measurements are of the same order of magnitude. However, there is an underestimation of measured ^{90}Sr concentrations (stations 23 and 27). This is not surprising since all models previously applied to predict dissolved ^{137}Cs dispersion fail to simulate the relatively high concentrations measured some tens of kilometers offshore Fukushima,[8] and stations 23 and 27 are relatively far from the nuclear plant (Figure 1). Generally speaking, there is a decrease in ^{90}Sr concentrations with depth, although subsurface maxima may be observed both in measurements and model results (stations 23 and 27). Results for station 29 (in the southeast corner of the model domain) are not shown since the model predicts that ^{90}Sr would not be present here at any depth, while a maximum concentration of 85 ± 3 Bq/m^3 was measured at the surface.[7] This may be due to the relatively low spatial resolution of JCOPE2 hydrodynamic model, which is not able to reproduce some features of circulation in this region, like small-scale eddies.

A plot showing the measured ^{90}Sr concentrations vs the calculated ones, for all samples, is presented in Figure 5. In general, the differences between calculated and measured concentrations are smaller than 1 order of magnitude. Actually, the mean value of the ratio between measured and calculated concentrations is 1.0 ± 0.7.

Snapshots of calculated ^{90}Sr concentrations in surface water at some dates are shown in Figure 6. Dispersion patterns are very similar to those previously obtained for ^{137}Cs,[9] since both radionuclides have a similar geochemical behavior and the temporal trend of releases is the same. Radionuclides are mainly transported to the south, although a significant northeast transport is also observed in mid April. The well-known clockwise eddy existing to the south of Fukushima[9] may be appreciated at this
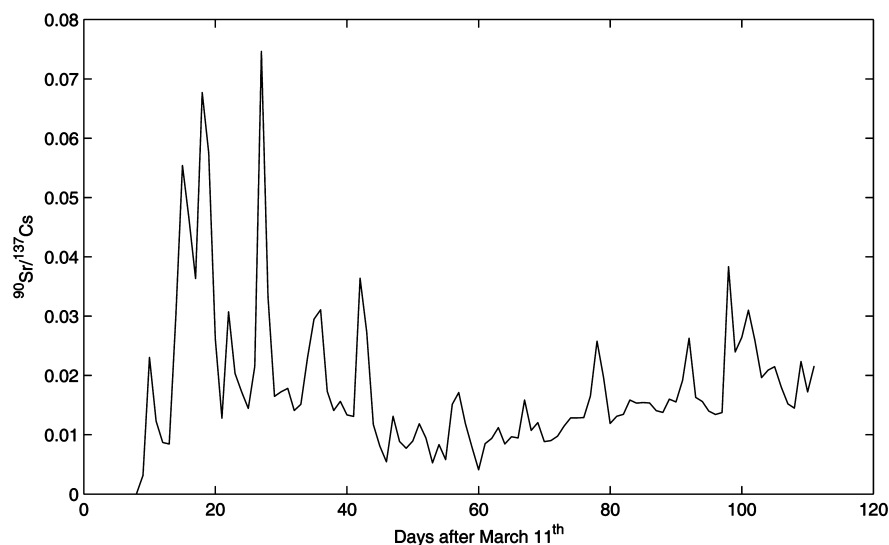

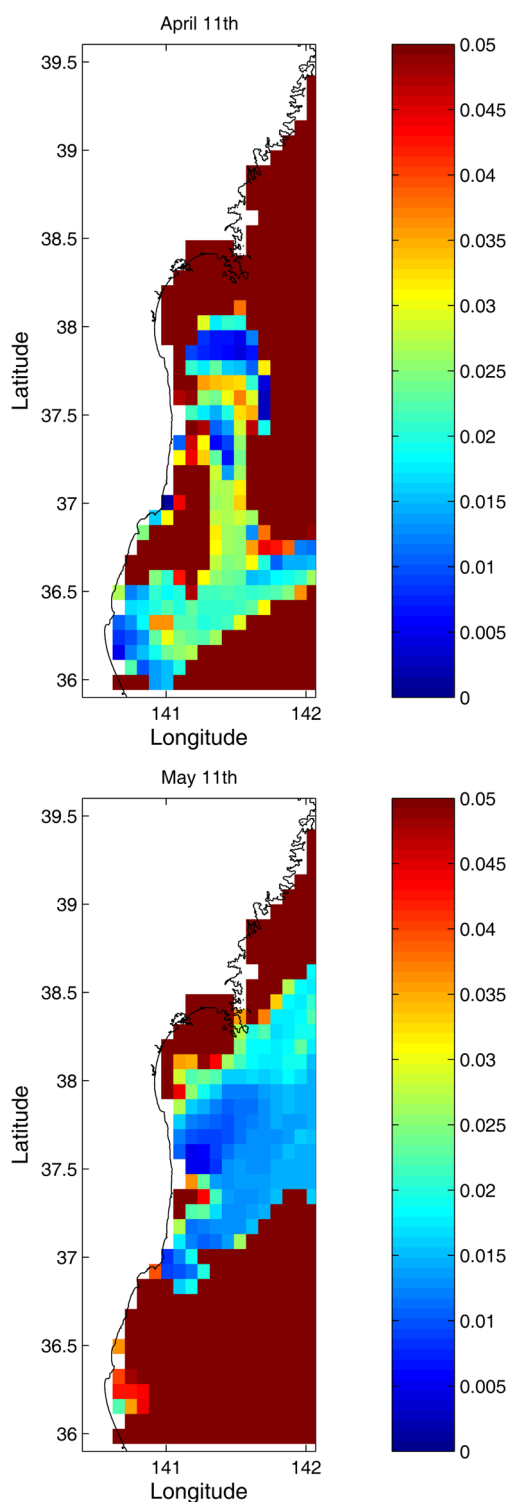**Figure 8.** Calculated ^{90}Sr/^{137}Cs ratios in surface water at 15 km offshore Fukushima.

Figure 9. Calculated ^{90}Sr/^{137}Cs ratios in surface water at two different times.

date as well. By the end of April release rates have been significantly reduced, and the radionuclide patch is slowly transported to the open ocean (May 1st map) with significant southeast and northeast components.[9] Actually, it is known that this is an area of convergence of two main currents (Oyashio and Kuroshio, which flow in such respective directions).

Predicted ^{90}Sr concentrations in bed sediments are extremely low, as it should be expected for such a very conservative

radionuclide. As an example, the predicted distribution for May 26th is presented in Figure 7. Maximum calculated concentrations in bottom sediments are about 0.7 Bq/kg in areas very close to release point in Fukushima.

The ^{90}Sr/^{137}Cs ratios in surface water for the model domain have been also calculated. ^{137}Cs concentrations have been obtained from previous simulations,[15] using the same model. As an example, the temporal evolution, until the end of June, of these ratios in a point 15 km offshore Fukushima (surface water) is presented in Figure 8. Background (pre-Fukushima) concentrations of both radionuclides are not included here. It may be seen that they present some variability. By the end of April (around day 50), the ratio is compatible with the mean value found by Povinec et al.[6] (0.01) for this month. Next there is an increase toward the mean value found in Casacuberta et al.[7] in June: 0.0265 ± 0.0006. Actually, the mean value for the whole sequence in Figure 8 is 0.018 ± 0.012. In June, the mean value of the sequence is 0.019 ± 0.006, in good agreement with reported values for this month.[7] ^{90}Sr releases are directly proportional to the ^{137}Cs ones (through factor F). Thus, variations in activity ratios must be related to the different geochemical behavior of both radionuclides: Cs is more reactive than Sr[18] and thus will be more adsorbed by sediments. The different adsorption is particularly enhanced in the shallow waters where direct releases occur and during weak current episodes (since water−sediment contact time increases).

As an additional example, maps of ^{90}Sr/^{137}Cs ratios in surface water at two different times are presented in Figure 9, now considering the pre-Fukushima background. In areas not affected by Fukushima releases the ^{90}Sr/^{137}Cs ratio is compatible with the atmospheric fallout characteristic value (0.63).[7] A ^{90}Sr/^{137}Cs ratio departing from 0.63 would be indicative that the water would have been affected by the releases from Fukushima Dai-ichi NPP.

Inventories of ^{90}Sr within the model domain are evaluated along the simulation time. The maximum inventory in the water column is obtained for April 10th, resulting in 58.4 TBq. By mid June, this figure is reduced to 32.5 TBq due to advection of radionuclides off the model domain and lower release rates. Casacuberta et al.[7] have estimated, for June, an inventory of 53 TBq (although their calculation domain is larger than the present model domain). This figure was obtained from the ^{137}Cs inventory estimated from measurements[1] and a established ^{90}Sr/^{137}Cs ratio of 0.0265 for waters off Fukushima. Inventories in bed sediments are 3 orders of magnitude smaller than in the water column: 0.031 and 0.015 TBq for April 10th (maximum inventory) and the middle of June, respectively. There are no reported data which could be used to test these model results.

Finally, the total ^{90}Sr release from Fukushima into the Pacific Ocean (from the accident until the end of June) has been calculated using the model, resulting in 80 TBq. Previous estimates, based on the assessed ^{137}Cs releases and a ^{90}Sr/^{137}Cs ratio of 0.0265,[7] yielded ^{90}Sr releases until June in the range of 90 and 900 TBq. The upper limit, however, is based on the ^{137}Cs release estimate by Bailly du Bois et al.,[2] which is significantly biased high as already justified in detail.[10]

Povinec et al.[6] estimated a ^{90}Sr release of about 40 TBq. This figure is based on a ^{90}Sr/^{137}Cs ratio of 0.01 (observed in April) and a ^{137}Cs release of 4 PBq, based on models.[3,4] Depending on the selected value for the ^{90}Sr/^{137}Cs ratio [equal to that in stagnant water produced in turbine buildings and surrounding areas during cooling of reactors (0.08); or equal to the mean value in surface water offshore Fukushima from April 2011

to February 2012 (0.24)], reported that ^{90}Sr releases could be as high as 1 PBq.[6] All of these previous estimates are summarized in Table 1.

Table 1. Previous Estimates of ^{90}Sr Releases Based on ^{90}Sr/^{137}Cs Ratios and ^{137}Cs Releases[a]

^{137}Cs release (PBq)	^{90}Sr/^{137}Cs ratio	^{90}Sr release (TBq)
3.5[3]	0.0265[7]	93
34[2]	0.0265[7]	901
4.0[4]	0.01[6]	40
4.0[4]	0.24[6]	960
4.0[4]	0.08[6]	320
	model estimation:	80

[a]References are given after each value.

If the mean ^{90}Sr/^{137}Cs ratio calculated by the model is used (0.018), then the ^{90}Sr releases would range from some 60 to 600 TBq (considering that ^{137}Cs release ranges from some 3.5[3] to 34[2] PBq).

Our estimation (80 TBq) is in the lower range of previous estimates mentioned above (40 TBq to 1 PBq, Table 1). While these are based on ^{137}Cs releases and a somewhat uncertain value for the ^{90}Sr/^{137}Cs ratio, our estimation is based on measured ^{90}Sr concentrations in surface waters offshore Fukushima through a modeling exercise.

It would be required to have more data on ^{90}Sr after June 2011, especially since discharges continued until present, with the particularity that, while Cs in the released water is being extracted to some extent, this is not the case for Sr and it could be of relevance on the long-term. Then, this work provides with a baseline for the first months after the accident that shall allow future comparisons and reconstruction of the historical releases.

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