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Local scale marine modelling of Fukushima releases. Assessment of water and sediment contamination and sensitivity to water circulation description

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ABSTRACT

The dispersion of ¹³⁷Cs released from Fukushima nuclear power plant to the sea after the March 11th 2011 tsunami has been studied using numerical models. The 3D dispersion model consists of an advection/diffusion equation with terms describing uptake/release reactions between water and seabed sediments. The dispersion model has been fed with daily currents provided by HYCOM and JCOPE2 ocean models. Seabed sediment ¹³⁷Cs patterns obtained using both current data set have been compared. The impact of tides and of atmospheric deposition has been evaluated as well. It has been also found that a 2-step kinetic model (two consecutive reversible reactions) for describing water/sediment interactions produces better results than a 1-step model (one single reversible reaction).

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1. Introduction

After March 11th, 2011, a significant amount of radionuclides were released into the Pacific Ocean from Fukushima nuclear power plant, as a result of the earthquake and tsunami occurred in such date.

A number of studies concerning numerical modelling of ¹³⁷Cs dispersion in the ocean have been published. The work by Nakano and Povinec (2012) deals with dispersion in the global ocean using an annually averaged water velocity field. Radionuclide sources are direct release of contaminated water to the ocean and atmospheric deposition. Dispersion is calculated through a particle-tracking method. Given its coarse resolution (2°), the model cannot give details on radionuclide distribution in the area close to Fukushima, where higher activities are measured. They estimate that the ¹³⁷Cs patch will reach the US coast in about 4–5 years, but specific activities will be low (<3 Bq/m³).

Kawamura et al. (2011) focus on a more regional scale and use a nesting method to study dispersion around Japan with a resolution about 6 km, as well as dispersion in the coastal area of Fukushima with a higher resolution (about 2 km). Dispersion of radionuclides is calculated using a particle-tracking method and two numerical experiments were carried out. In one case only direct release to the ocean is considered; in the other experiment both direct release to the ocean and atmospheric deposition are considered. A reasonable agreement between calculated ¹³⁷Cs concentrations

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and measurements near Fukushima was obtained. These authors also found that direct releases are dominant over atmospheric deposition from the end of March on. Moreover, the effects of atmospheric deposition are more significant in the case of ¹³¹I than for ¹³⁷Cs.

A similar domain to the coastal one described above has been used by Tsumume et al. (2012) with a spatial resolution of 1 km. Hydrodynamics is forced by wind stress and heat and freshwater fluxes provided by a weather forecasting model and tidal effects are also included. Dispersion is calculated in an Eulerian frame. These authors have also found that ¹³⁷Cs coming from direct releases to the ocean is more significant than that originating from atmospheric deposition.

Honda et al. (2012) have simulated ¹³⁷Cs dispersion over a larger scale using a particle-tracking model fed with currents provided by the Japan Coastal Ocean Predictability Experiment 2 (JCOPE2). They have found that high ¹³⁷Cs concentrations measured north of 40° latitude have to be attributed to aeolian input.

A clear agreement between regional models (Honda et al., 2012; Kawamura et al., 2011; Tsumume et al., 2012) is that mesoscale eddies significantly affect offshore transport, leading to complex dispersion patterns. A comparison of the performance of five models in simulating the dispersion of dissolved ¹³⁷Cs is presented in Masumoto et al. (2012).

It is common to all developed models to consider ¹³⁷Cs as a perfectly conservative radionuclide, thus scavenging processes and adsorption on seabed sediments are neglected. Interaction processes between water and sediments should be included in a proper assessment of the aftermath of Fukushima releases since contaminated sediments will act as a long-term delayed source



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of radionuclides previously adsorbed on sediments. The description of water–sediment interactions on the basis of the equilibrium distribution coefficient, k_d , concept is not appropriate in the vicinity of the source and for non-instantaneous releases, since the system water–sediment is not at equilibrium (Periáñez, 2003a). Thus, a kinetic approach will be more adequate. Several kinetic models (involving single or multi-step reversible reactions) could be adopted (Periáñez, 2003b, 2004).

The objective of this paper is to analyse, using numerical modelling, the dispersion of ¹³⁷Cs in the coastal area around Fukushima, taking into account for the first time water/sediment interactions. Calculated ¹³⁷Cs concentrations in the seabed are compared with measurements. Two kinetic models for those interactions have been tested. We will mainly focus on ¹³⁷Cs distributions in seabed sediments, which have not been studied up to present day. Also, the effect of water circulation description on dispersion patterns was studied: the relevance of tides, with respect to residual circulation (wind and density driven currents), has been assessed. Also, two descriptions of this residual circulation have been used and compared: the provided by HYCOM and JCOPE2 models. We have limited our work to the coastal area around Fukushima since activities are higher here, thus this region is more relevant from the radiological point of view.

The model is presented in the following section. Next, results are presented and discussed.

2. Model description

The dispersion model described below has been developed by the authors and tested in different marine environments (Periáñez, 2003a,b, 2008, 2009, 2012). This code has been modified to import currents from HYCOM and JCOPE2 models. Thus, exactly the same dispersion model is run, the only difference is the source of currents to calculate advective transport.

A brief description of hydrodynamic models is given below. Next, the dispersion model is briefly described as well, since details may be seen in current literature (references cited above).

2.1. Water circulation

2.1.1. Tidal currents

Tides are obtained from a 2D depth-averaged model since other authors have already stated that it is a reasonable approach (Dyke, 2001; Yanagi, 1999). Equations may be seen, for instance, in Periáñez (2005). The solution of these equations provides the water currents at each point in the model domain and for each time step. Currents are treated through standard tidal analysis (Pugh, 1987, Chapter 4) and tidal constants are stored in files that will be read by the dispersion code to calculate the advective transport. The model includes the two main tidal constituents, M_2 and S_2 . Thus, the hydrodynamic equations are solved for each constituent and tidal analysis is also carried out for each constituent separately. A residual transport cannot be produced by the pure harmonic currents given by the tidal analysis, thus tidal residuals have been calculated as well. The procedure may be seen in detail in Periáñez (2012).

Hydrodynamic equations are solved using explicit finite difference schemes, with a second order accuracy scheme for non-linear terms. Boundary conditions consist of specifying water surface elevations, from measured tidal constants, along open boundaries of the domain. Model equations and numerical schemes have been carefully tested in the past (Periáñez, 2008, 2009, 2012).

As an example, tide amplitude and current amplitude for the M_2 tide, calculated on the HYCOM domain (see below), are presented in Fig. 1. A comparison between measured and calculated tidal constants at two points indicated in Fig. 1 is presented in Table 1. It may bee seen that there is a generally good agreement between both set of data. It must be pointed out that, differently to wind and density driven circulation, tides have been calculated by the authors using their own codes.

2.1.2. Residual circulation

A brief description of the main characteristics of the hydrodynamic models from which wind and density driven currents are obtained is given in the following paragraphs.

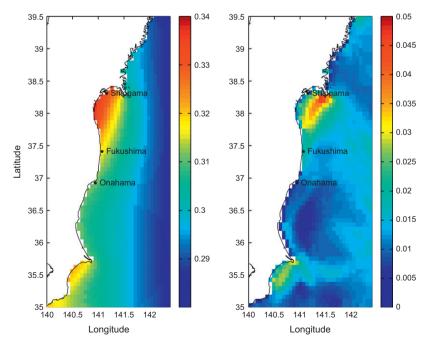


Fig. 1. Maps of computed tide amplitude (m, left panel) and tidal current amplitude (m/s, right panel) for the M₂ constituent and on the HYCOM domain.

Table 1

Observed, index *obs*, and computed, index *comp*, amplitudes (*A*, m) and phases (g, deg) of tidal elevations at several locations indicated in Fig. 1. The source of data is NOAA (1982).

	<i>M</i> ₂				S ₂			
Station	Aobs	g _{obs}	A _{comp}	g _{comp}	Aobs	g _{obs}	A _{comp}	g _{comp}
Onahama Shiogama	0.30 0.35	106 97	0.31 0.33	105 98	0.14 0.16	149 141	0.15 0.16	150 143

HYCOM model

Daily averaged three dimensional current fields have been obtained from the HYCOM (HYbrid Coordinate Ocean Model) in the time frame from March 12th to May 30th. Details may be seen in http://www.hycom.org. The area studied ranges from 140° to 142.96° longitude and from 35.03° to 39.48° latitude. There are 33 vertical levels and spatial resolution is about 7 km.

JCOPE2 model

Daily averaged three dimensional currents have also been obtained from the JCOPE2 (Japan Coastal Ocean Predictability Experiment 2) model 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. There are 23 vertical levels and spatial resolution is about 9 km. Details may be seen in http://www.jamstec.go.jp/frcgc/jcope/htdocs/e/

jcope_system_description.html.

As an example, surface currents corresponding to March 12th, produced by both models, may be seen in Fig. 2. The Kuroshio current-flowing to the NE-is clearly appreciated, specially in the larger HYCOM domain. Currents are weak in the area of Fukushima and flow southwards. To the south of Fukushima an anticyclonic eddy may be seen in both domains, although its position is not exactly the same.

It must be commented that, if tides are included in a given simulation, non-linear interactions between tidal constituents and between tides and wind plus density driven flows are removed if a

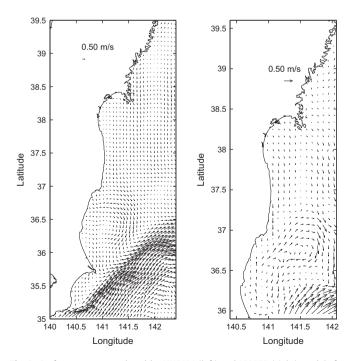


Fig. 2. Surface currents produced by HYCOM (left) and JCOPE2 (right) models for March 12th.

simple addition of hydrodynamic model results is carried out. However, this is a common practice in transport modelling (Proctor et al., 1994; Elliott and Clarke, 1998; Periáñez, 2009).

2.2. Dispersion model

The dispersion model 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. These processes are formulated in a dynamic way, in terms of kinetic transfer coefficients. A detailed description of such formulation may be seen elsewhere (Periáñez, 2008, 2009, 2012).

There has been evidence to suggest that uptake takes place in two stages: fast surface adsorption followed by slow migration of ions to pores and interlattice spacings (Nyffeler et al., 1984; Turner et al., 1992: Turner and Millward, 1994: Ciffrov et al., 2001: El Mrabet et al., 2001). Consequently, two kinetic models have been tested. The 1-step model considers that exchanges of radionuclides between water and sediments are governed by a first-order reversible reaction, being k_1 and k_2 the forward and backward rates respectively. The 2-step model considers that exchanges are governed by two consecutive reversible reactions: surface adsorption is followed by another process that may be a slow diffusion of ions into pores and interlattice spacings, inner complex formation or a transformation such as an oxidation. k_3 and k_4 are forward and backward rates for this second reaction (Fig. 3). Thus, sediments are divided in two phases: a reversible and a slowly reversible fraction. It has been shown that the 2-step model reproduces both the adsorption and release kinetics of ¹³⁷Cs in the Irish Sea, where it is released from Sellafield nuclear fuel reprocessing plant (Periáñez, 2003b).

A detailed formulation of these models may be seen in Periáñez (2003b, 2004), thus will not be repeated here. However, a few comments are made. 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 (Periáñez, 2003a, 2004, 2008, 2009). 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 (Periáñez, 2003a, 2004, 2008, 2009). Assuming spherical particles, the exchange surface is written as (see references cited above):

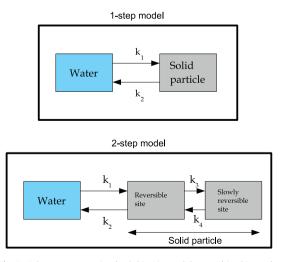


Fig. 3. Scheme representing both kinetic models tested in this study.

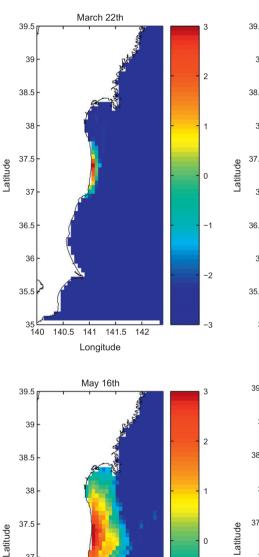
(2)

Table 2

Summary of model runs.

Run	Hydrodynamics	Kinetic model	Tides	
1	НҮСОМ	1-step	No	
2	HYCOM	1-step	Yes	
3	HYCOM	2-step	No	
4	JCOPE2	2-step	No	

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



where *R* is particle radius, *p* is 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 modelling works cited above. Real particles are not spheres, but with this approach it is possible to obtain an analytical expression for the exchange

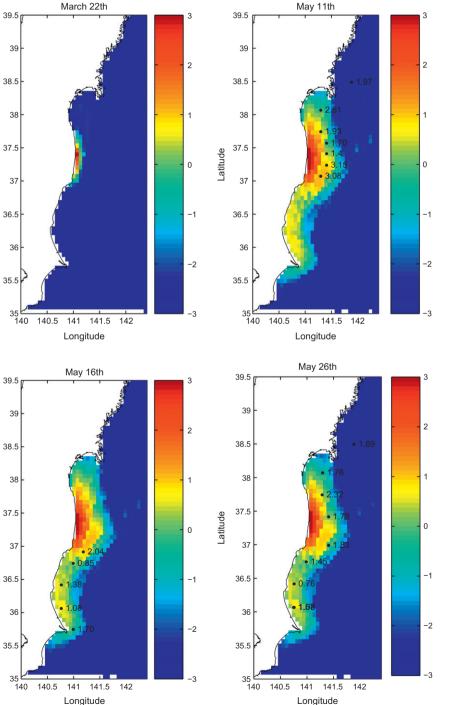


Fig. 4. Calculated (using HYCOM circulation) and measured ¹³⁷Cs concentrations in seabed sediments (Bq/kg). Logarithms of calculated and measured concentrations are drawn.

surface (Duursma and Carroll, 1996). Full model equations in a 3D form may be seen in Periáñez (2009). Numerical solution is carried out using second order accuracy finite difference schemes (Periáñez, 2005).

3. Results

Some parameters are required to simulate ¹³⁷Cs dispersion. Rates k_2 , k_3 and k_4 are taken from previous works dealing with dispersion of this radionuclide (Periáñez, 2004, 2008). Although it is true that kinetic rates are site-specific, there is not information about them in Japan Pacific Ocean coastal waters. Thus, representative values already used in the English Channel and Western Mediterranean have been used as a first order approximation. As discussed before (Periáñez, 2003a, 2004, 2008, 2009), the exchange velocity γ can be deduced from rates mentioned above and the radionuclide distribution coefficient k_d . The mean value of the measured Cs distribution coefficient is 2.1×10^3 (Honda et al., 2012), comparable to the IAEA (2004) recommended value. We have fixed $k_d = 2 \times 10^3$ to deduce γ following the procedure described in such references. The distribution of fine sediments in the seabed, described by parameter f, has been reconstructed from information in Saito (1989). It has been fixed L = 0.05 m. This parameter typically ranges from 0.035 m (Periáñez, 2008) to 0.10 m (Periáñez, 2000, 2003b, 2004, 2009). Also, φ = 0.1 (Periáñez, 2000, 2003b, 2004, 2009). A representative value *R* = 10 µm has been used for the mean particle size, very similar to the used in Periáñez (2000).

To avoid uncertainties in the source term (liquid releases of 137 Cs to the sea) the measured concentration of this radionuclide in the release point (Bailly du Bois et al., in press) has been defined as a "boundary condition" in the grid cell where such point is located in each computational mesh. The same approach was adopted by Kawamura et al. (2011). These authors assumed that the observed concentration at the outlet extended over an area of 1.5 km² in front of the plant. We used the same approach. Since the grid cell surface is larger than 1.5 km² in both domains, a correction factor was introduced to assure that the total activity in the release grid cell is the same as if the discharge was homogeneous over an 1.5 km² area. Given the shallow waters in the outlet point, it was also assumed ¹³⁷Cs concentration here is vertically homogeneous.

An estimation of atmospheric deposition of ¹³⁷Cs on the sea surface is also included from atmospheric dispersion modelling results in Honda et al. (2012).

A number of numerical experiments have been carried out, which are summarized in Table 2. The most relevant results will be commented.

Essentially the same results (differences could not be appreciated) were obtained from runs 1 and 2. From this comparison it could be deduced that tides do not play a significant role in dispersion, compared with wind and density driven circulation. Indeed, it may be seen in Fig. 1 that tidal currents for the main constituent are below 5 cm/s in most of the domain. A comparison of results from experiments 1 and 3 highlighted that a 2-step model produces ¹³⁷Cs concentrations in the seabed in better agreement with observations than a 1-step model. This is not a surprising result, as has already been found in numerical modelling of Sellafield releases in the Irish Sea (Periáñez, 2003b) and in laboratory experiments (Ciffroy et al., 2001). Dissolved ¹³⁷Cs concentrations in surface water are not affected by the kinetic model used to describe water/sediment interactions in the seabed. The present model does not improve former ones with respect to dissolved ¹³⁷Cs in surface water, as will be also discussed below. However,

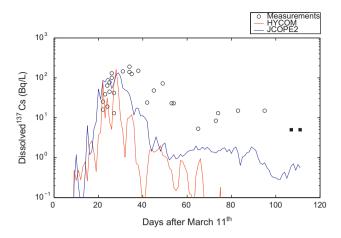


Fig. 5. Measured and calculated dissolved ¹³⁷Cs concentrations in surface water 15 km offshore Fukushima. Black squares indicate measurements below the 5 Bq/L detection limit.

our objective is simulating, for the first time, contamination of seabed sediments.

From these experiments it was concluded that tides may be neglected in comparison with residual circulation. Also, it seems that a 2-step model is producing better results than a 1-step model. Now, experiments 3 and 4 are compared in detail to assess differences purely due to the description of residual circulation (HYCOM or JCOPE2).

Measured and computed ¹³⁷Cs concentration in seabed sediments produced by the dispersion model fed with HYCOM currents may be seen in Fig. 4. Soon after the tsunami (March 22th), ¹³⁷Cs in sediments remains close to the release point. By the middle of April (not shown) a very steady distribution is obtained in sediments, and this remains essentially the same during May, as can be seen in Fig. 4. However, the model generally underestimates concentrations by approximately one order of magnitude. Also, contamination of the seabed extends along a relatively narrow coastal band, differently to experimental results.

Measured and calculated dissolved ¹³⁷Cs concentrations in surface water, 15 km offshore Fukushima, may be seen in Fig. 5. Maximum concentrations, which are reached by the beginning of April, are very well reproduced by the model. However, there is a clear underestimation afterwards. The model predicts a too fast dispersion of ¹³⁷Cs away from this area. It is interesting to note that all models previously applied to predict dissolved ¹³⁷Cs dispersion fail to simulate the relatively high concentrations measured some km offshore Fukushima (Masumoto et al., 2012).

Results for sediments, if water circulation produced by JCOPE2 model is used, may be seen in Fig. 6. In this case, seabed contamination reaches areas far from Fukushima, in agreement with observations (see for instance maps of May 26th and June 10th). A essentially steady distribution in sediments is not reached until the beginning of June, thus later than with HYCOM circulation. Finally, it may be seen that calculated concentrations in the sediment are higher than those of Fig. 4, and generally agree (at least by order of magnitude) with observations.

Results for dissolved ¹³⁷Cs in surface water 15 km offshore Fukushima are presented in Fig. 5. As with HYCOM circulation, the initial peak is very well reproduced by the model. A too fast decrease in concentrations is again produced, although results seem to improve with respect to those obtained with HYCOM circulation.

Generally speaking, it seems that currents provided by JCOPE2 model produce ¹³⁷Cs concentrations in surface water and seabed sediments in better agreement with observations than currents

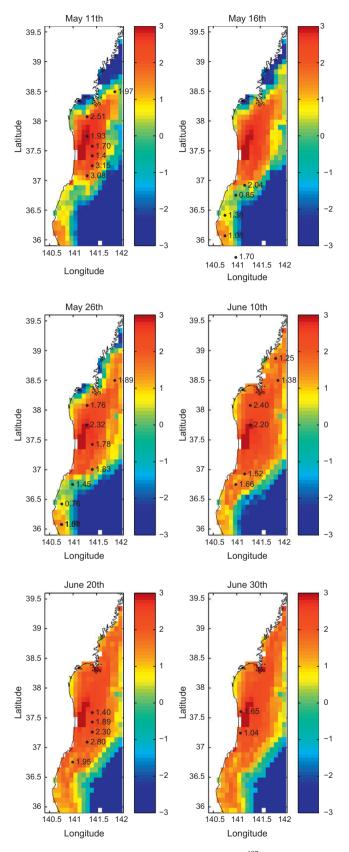


Fig. 6. Calculated (using JCOPE2 circulation) and measured ¹³⁷Cs concentrations in seabed sediments (Bq/kg). Logarithms of calculated and measured concentrations are drawn.

produced by HYCOM model. Also, a 2-step kinetic model seems to work better than a 1-step model. With both HYCOM and JCOPE2

currents there is a too fast decrease in dissolved surface ¹³⁷Cs concentrations. The reason could maybe be related to the existence of an eddy, too small to be adequately reproduced by circulation models, which could trap dissolved ¹³⁷Cs near the coast. But of course this cannot be confirmed. It is worth commenting that the model by Tsumume et al. (2012) also underestimates, by one order of magnitude, ¹³⁷Cs concentrations in water in the coastal area around Fukushima after mid-April.

Another experiment was carried out in the conditions of run 4 in Table 2 but without atmospheric deposition. No significant differences in results were observed. As already noted by Kawamura et al. (2011) and Tsumume et al. (2012), direct releases to the sea are dominant over atmospheric deposition on the surface.

The sediment half-time has been defined (Periáñez, 2003a) as the time in which the radionuclide content in the sediment decreases by a factor 2. It is known that a contaminated sediment may act as a long-term delayed source of previously released contaminants (Cook et al., 1997). Consequently, it is relevant to have estimations of the sediment half-time. The total inventory of ¹³⁷Cs over the JCOPE2 domain in bed sediments has been evaluated each time step. From numerical fitting of the temporal evolution of such inventory (once it has started to decrease, i.e., 40 days after March 12th) to an exponential decay function, it was obtained that sediment half-time is 167 days. This number is of the same order as ¹³⁷Cs sediment halving time in the English Channel obtained with a 2-step kinetic model (Periáñez, 2004).

4. Conclusions

A three dimensional dispersion model has been used to evaluate ¹³⁷Cs dispersion in the coastal area around Fukushima nuclear power plant. The model includes uptake/release reactions between water and seabed sediments, which are described in a dynamic way using kinetic transfer coefficients. Two kinetic models have been tested: a 1-step model consisting of a single reversible reaction and a 2-step model consisting of two consecutive reversible reactions. All simulations carried out before considered ¹³⁷Cs as a perfectly conservative radionuclide.

Tides have been calculated using a 2D depth averaged model. Three dimensional wind and density driven circulation has been obtained from the output of two well-known ocean models: HY-COM and JCOPE2. Exactly the same dispersion model is run, but with different water currents. Daily averaged values from both models are used.

Several conclusions are obtained from the numerical experiments which have been carried out:

- Tides do not play a significant role in transport and mixing in Fukushima coastal area, since essentially the same results are obtained in simulations carried out with and without them.
- Accordingly to previous simulations, direct releases to the sea dominate over atmospheric deposition on the sea surface.
- A 2-step kinetic model produces results in better agreement with observations than a 1-step model. The same conclusion was obtained from past simulations in the Irish Sea.
- In general, the dispersion model produces better results if it is fed with JCOPE2 currents than with HYCOM currents. With HYCOM circulation, seabed contamination remains close to the coast, which is not in agreement with measurements. Also, concentration levels in sediments are generally underestimated.
- Dissolved surface ¹³⁷Cs concentrations offshore Fukushima are underestimated after the initial activity peak. Maybe this could be attributed to the low spatial resolution of circulation models.

Acknowledgements

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