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Dynamic modelling of radionuclide uptake by marine biota: application to the Fukushima nuclear power plant accident

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1 Background

The Dynamic Dose Assessment Tool (D-DAT) was developed to study the dynamics of radionuclide uptake and turnover in marine biota (Vives Batlle et al. 2008). This dynamics is determined by the interplay between the residence time of radionuclides in seawater/sediments and the biological half-lives of elimination in the biota. The model calculates time-variable activity concentration of ^{131}I, ^{134}Cs, ^{137}Cs and ^{90}Sr in seabed sediment, fish, crustaceans, molluscs and macroalgae from surrounding activity concentrations in seawater, with which to derive internal and external dose rates.

The model has recently been redeveloped and its capabilities expanded to assess the impact of the Fukushima accident in the local coastal biota (Vives i Batlle and Vandenhove 2014), whereupon it was successfully applied as part of the recent comprehensive UNSCEAR study addressing the environmental impact of the accident (Vives i Batlle et al. 2014). As part of the Marine IRA of COMET, it was agreed to complete development of an advanced version of D-DAT (Task 2 of the WP3 Marine IRA working group) by including dynamic transfer of radionuclides to/from sediments, which factorises depletion of radionuclides adsorbed onto suspended particulates, molecular diffusion, pore water mixing and bioturbation coupled to the differential equations describing the biological uptake/turnover processes. In this way, the model is capable of reproducing activity concentration in sediment more realistically.

The model was then used to recalculate the radiological impact of the Fukushima accident on marine biota in the acute phase of the accident, using extended biokinetic parameters (consideration of both the short- and long-term biological half-life, or $T_{B1/2}$, for radionuclides in biota). Additionally, we performed a retrospective recalculation of the marine source term calculation for ^{131}I, ^{137}Cs and ^{90}Sr. The work was presented orally at ICRER 2014 - 3rd International Conference on Radioecology & Environmental Radioactivity, 7-12 September 2014 (Vives i Batlle 2014). The paper was then selected for publication at the ensuing special issue of the Journal of Environmental Radioactivity (Vives i Batlle 2015), satisfying the stated milestone of producing an article (IRA-Marine-D2: Journal article).

2 Study results

2.1 Data sources and model set-up

As primary source of data for this study we used the UNSCEAR (2014) quality-assured dataset of activity concentration in unfiltered surface seawater, sediment and fish covering the period up to August 2012. To calibrate the model we expanded the existing biokinetic – allometric data for ^{131}I and ^{134,137}Cs from previous work (Vives i Batlle et al. 2007) with ^{90}Sr data from other sources (Boroughs et al. 1956; Casper et al. 2004; Tagami and Uchida 2013; Polikarpov 1965; Phillips and Russo 1978). A new model version was developed in the ModelMaker® 4 environment (Fig. 1) with equations solved using the Gear integration method (Gear 1971).

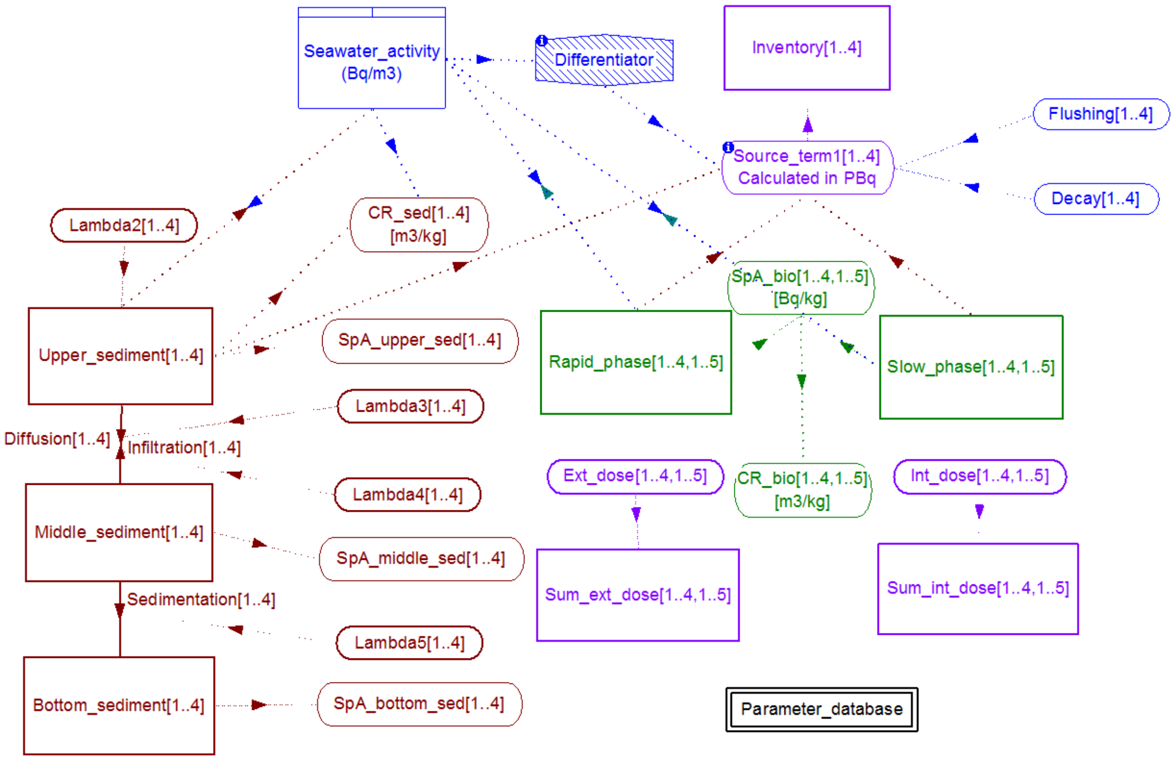

Figure 1. Conceptual representation of D-DAT. Symbols: sub-model (double rectangle) compartment (rectangle), variable (rounded rectangle), flows (solid lines), influences (dotted lines).

2.2 Development of dynamic sediment model and dual $T_{B1/2}$ approach

We applied a sub-model of 3 sediment layers (0.05, 1.95 and 8 m thick) from Simmonds *et al*. (2004) which includes the processes of particle scavenging, molecular diffusion, particle mixing, pore water mixing and sedimentation, as described in Vives i Batlle et al. (2008) (see main parameters in Table 1). We also applied a dual $T_{B1/2}$ approach, as most organisms depurate radionuclides from their bodies via a fast short-term process followed by a longer-term process (Vives i Batlle et al. 2005). The model was further adapted to recalculate the amount of radionuclide discharged in the initial phase of the accident, based on balancing the time derivative of the actual seawater concentrations with (a) the radionuclide fluxes to/from biota and sediment and (b) water flushing, in combination with the available monitored radionuclide concentrations in seawater.

The model can calculate internal and external dose rates, using the ERICA biota dose assessment approach (Brown et al. 2008). The model employs dose rate per unit concentrations (DPUC, in μGy h^{-1} per Bq kg^{-1}) derived from template ERICA simulations for marine biota, using the default parameters (such as occupancy factors) from the ERICA tool. In the latest version of D-DAT, dosimetry and transfer parameters are updated according to the latest version of ERICA and the most recent IAEA publication on transfer parameters to wildlife (IAEA 2014).

Table 1: Oceanographic and sediment-related D-DAT parameters

Parameter	Value	Units	Source
Volume water compartment	3.41E+08	m^3	Periáñez (pers. comm.)
Surface area	6.81E+07	m^2	Periáñez (pers. comm.)
Volume of sediment	3.41E+06	m^3	Periáñez (pers. comm.)
Dynamic viscosity of seawater	1.62E+02	$kg\ m^{-1}\ d^{-1}$	McDonnell and Buesseler (2010)
Sediment bulk density	1.50E+03	$kg\ m^{-3}$	Periáñez (pers. comm.)
Sediment particle density	2.60E+03	$kg\ m^{-3}$	Periáñez (pers. comm.)
Sinking particle velocity	3.94E-02	$m\ d^{-1}$	Calculated (this study)
Particulate depth attenuation coefficient	1.90E-02	m^{-1}	Buesseler (pers. comm.)
Seawater flushing time	2.2 ± 0.3	d	Calculated (this study)
Sedimentation rate	4.89E-05	$kg\ m^{-2}\ d^{-1}$	Honda et al. (2013) - Station S1
Diffusion rate	8.62E-05	$m^2\ d^{-1}$	Simmonds et al. (2004)
Pore-water turnover rate	2.74E-03	d^{-1}	Simmonds et al. (2004) - shallow seas
Sediment reworking rate	1.37E-05	$m\ d^{-1}$	Simmonds et al. (2004) - shallow seas

2.3 *Description of the main results*

The results are fully described in the relevant publications (Vives i Batlle 2015, 2014) whence the following is extracted. Predicted surface sediment activity concentrations near the FDNPS are within the scattered activities reported from actual monitoring measurements. The model also predicts that, following uptake, radioactive decay is the dominant process by which activity concentration in the upper layer changes on the sub-annual timescale of this study. The activity in the upper sediment (< 5 cm) is predicted to rise to 630 Bq kg^{-1} (22 days) and 5400 - 5600 Bq kg^{-1} (40 – 46 d) for ^{131}I and ^{134}Cs – ^{137}Cs, respectively. The monitored data trend of ^{137}Cs sediment concentrations in the vicinity of the FDNPS (< 2 km) confirms this prediction, whilst the simplistic ERICA K_d approach would have given unduly pessimistic values of ~10^5 Bq kg^{-1}.

Although there are no direct monitoring data in the FDNPS zone with which to validate the acute period, model predictions are consistent with data from coastal stations < 50 km from Fukushima Dai-ichi for ^{137}I in macroalgae and molluscs (Vives i Batlle and Vandenhove 2014). Long-term predicted ^{137}Cs activity concentrations match the benthic fish samples collected near the FDNPS in days 440 – 520 post- accident, for which an exponential trend $A(Bq\ m^{-3}) = 252.7e^{-0.166T}$(days) is observed, which can be back-extrapolated to the relevant time. Where a single $T_{B1/2}$ model calibrated with the long-term $T_{B1/2}$ would have overshot these concentrations, the dual $T_{B1/2}$ model can therefore be calibrated to predict correctly both acute and long-term biota ^{137}Cs concentrations.

We calculated the source term influx of radioactivity to sea necessary to sustain the activity concentrations observed in the FDNPS vicinity. This requires the flushing time of water from this coastal region. We fitted the ^{137}Cs data from the chronic period (T> 50 d) to a double release exponential and used the rate constants derived to calculate the flushing time by a standard method (Choi and Lee 2004; Periáñez 2012), giving 2.2 ± 0.3 days. With this information, the model gives integrated releases of 103, 30 and 3 PBq for ^{131}I, ^{137}Cs and ^{90}Sr.

The UNSCEAR report puts these estimates in the ranges 70 – 120 and 8 – 14 PBq for ^{131}I and ^{137}Cs (no data for ^{90}Sr), hence our model gives a reasonable prediction.

Biota dose rates calculated with D-DAT approximate those estimated previously (UNSCEAR 2014) when the model is used in single $T_{B1/2}$ mode. The main differences with the new model are the dual $T_{B1/2}$ approach and modelling the sediment pathway (potentially increasing peak dose rates by a factor of 5). For benthic fish, the calculated external dose for ^{131}I and radiocaesium from sediment only exceeds 50% of the total (sediment + seawater) after 30 – 35 d, supporting previous suggestions that, during the acute phase, sediments were not sufficiently contaminated to give significant exposures. However, in the long term it is proper to factorise sediment hold-up. After 50 days, external exposure from sediment exceeds that from seawater (for benthic biota), though internal exposure still dominates till 90 days (at that time, predicted dose rates are <1 μGy h^{-1}). .

2.4 Summary

Up to recently, most marine radioecology studies have addressed chronic release situations, and most radioecological models for radionuclide transfer to marine biota are for equilibrium situations. However, we have found that in a post-accidental situation, the need to use dynamic modelling is incontrovertible. The importance of integrating a dynamic representation of sediment uptake is also demonstrated, highlighting its potential for source-term reconstruction and a better understanding of long-term processes. The D-DAT model is a well-proven practical tool which can address these issues.

3 Current progress

The study as envisioned is complete and the goal of producing an article (IRA-Marine-D2: Journal article) has been accomplished in the form of (a) oral presentation and extended abstract at the ICRER conference and (b) publication as full journal article in the Journal of Environmental radioactivity. The article, which is already available online (http://dx.doi.org/10.1016/j.jenvrad.2015.02.023), explicitly acknowledges the link to the COMET project.

Work on D-DAT continues as part of the COMET-FRAME project, as this project proceeds in full syntony with the COMET Marine IRA. The model is being further updated to include additional radionuclides (^{129}I, ^{236}U), which have so far been poorly studied in relation to the Fukushima accident, yet they are important as tracers of the water masses, particularly in the mixing region or “perturbed area” in between sea currents. We are also planning to use local parameters derived from the FRAME sea cruises and new data from recent publications for the better calibration and validation of the D-DAT dual compartment kinetic exchange with multiple $T_{B1/2}$s.

Future avenues of development being considered are the addition of new organisms (e.g. plankton) and addressing any remaining data gaps in the biokinetic data by allometric analysis of the biokinetic database for marine biota that we are developing as part of the

IAEA MODARIA Working Group 8. In the longer term, we plan to migrate D-DAT from its current ModelMaker platform to FORTRAN code, producing a simple user interface for the model using Visual Basic, thereby rendering the model more friendly for non-specialised users.

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