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Inter-comparison of dynamic models for radionuclide transfer to marine biota in a Fukushima accident scenario☆

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ABSTRACT

We report an inter-comparison of eight models designed to predict the radiological exposure of radionuclides in marine biota. The models were required to simulate dynamically the uptake and turnover of radionuclides by marine organisms.

Model predictions of radionuclide uptake and turnover using kinetic calculations based on biological half-life ($T_{B1/2}$) and/or more complex metabolic modelling approaches were used to predict activity concentrations and, consequently, dose rates of ^{90}Sr, ^{131}I and ^{137}Cs to fish, crustaceans, macroalgae and molluscs under circumstances where the water concentrations are changing with time. For comparison, the ERICA Tool, a model commonly used in environmental assessment, and which uses equilibrium concentration ratios, was also used. As input to the models we used hydrodynamic forecasts of water and sediment activity concentrations using a simulated scenario reflecting the Fukushima accident releases.

Although model variability is important, the intercomparison gives logical results, in that the dynamic models predict consistently a pattern of delayed rise of activity concentration in biota and slow decline instead of the instantaneous equilibrium with the activity concentration in seawater predicted by the ERICA Tool. The differences between ERICA and the dynamic models increase the shorter the $T_{B1/2}$ becomes; however, there is significant variability between models, underpinned by parameter and methodological differences between them.

The need to validate the dynamic models used in this intercomparison has been highlighted, particularly in regards to optimisation of the model biokinetic parameters.

1. Introduction

Radiological protection of the environment (i.e. wildlife) is still relatively novel and exposure assessment methodologies for non-human biota are being continually improved. It is generally

☆ This paper is dedicated to the memory of our departed colleague and friend Rudie Heling.

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accepted that prediction of the uptake of radionuclides from the surrounding environmental media by organisms is a major source of uncertainty (Beresford et al., 2008).

The development of assessment approaches has focused on chronic exposure scenarios and, for aquatic biota, the majority of radiological assessment models assume that the activity concentration in an organism of mass M (i.e. A_O, in Bq kg^{-1} expressed on a fresh mass (f.m.) basis) is proportional to the activity concentration (A_W, in Bq L^{-1}) in an adjacent volume V of water *via* a whole organism concentration ratio, or CR_{wo} (in L kg^{-1} f.m.) (IAEA, 2014). The ERICA Tool (Brown et al., 2008) is an example of a model which represents the uptake of radionuclides from environmental media by these simple CR_{wo}s. These methodologies are unlikely to assess reliably situations outside of equilibrium.

The truth is that, in reality, instantaneous equilibrium between biota and the medium does not exist. This is because biota accumulates radionuclides with a 'time delay' relative to variations of activity concentration in seawater. In its simplest formulation, the dynamics of the process are determined by a balance between the residence time of the radionuclide in the water in the presence of efficient hydraulic dilution, and the biological half-life ($T_{B1/2}$) of an organism. For a single component biological half-life, the activity concentrations in biota (A_O, Bq kg^{-1}) and water (A_W, Bq m^{-3}) can be represented by a simple model with two rate constants; k_W for uptake and k_O for elimination: $\frac{dA_O}{dt} = k_W A_W \frac{V}{M} - (k_O + \lambda)A_O$; $\frac{dA_W}{dt} = -(k_W + \lambda)A_W + k_O \frac{M}{V} A_O$.

Where $k_O = \frac{\ln 2}{T_{B1/2}}$, $k_W = ((k_O + \lambda)M/V)CR_{wo}$ and λ is the radionuclide decay constant (Vives i Batlle, 2012). This type of model can be simplified by assuming that the water concentration does not depend on the exchange from an aquatic organism (because the amount of radioactivity in the organism is much smaller than in the surrounding volume of water, V) – hence $dA_W/dt \approx 0$, and that the organism uptake rate does not change with time (i.e. ignoring the effect of organism growth).

Other dynamic models exist that are more complex and can, for example, model uptake by higher organisms *via* food (Brown et al., 2004; Keum et al., 2015; Maderich et al., 2014), requiring two additional parameters: assimilation efficiency and ingestion rate. Furthermore, some models consider organism growth processes requiring information on metabolism (Sazykina, 2000) and other models include more complex food web modelling (Heling et al., 2002).

The Fukushima nuclear accident has refocused strongly the vision for marine radioecology and highlighted the limited knowledge that we have in this area (Vives i Batlle, 2011). This disaster has brought some evidence that a dynamic modelling approach is advantageous compared with traditional equilibrium-based transfer approaches (Psaltaki et al., 2013; UNSCEAR, 2014; Vives i Batlle et al., 2014; Vives i Batlle and Vandenhove, 2014), owing to the relatively slow response of many biota to changing concentrations in seawater. Some models such as BURN-POSEIDON (Maderich et al., 2014), D-DAT (Vives i Batlle et al., 2008) and ECOMOD (Sazykina, 2000) have been applied in a 'dynamic assessment' context, including as part of the recent assessments of the impact of the Fukushima nuclear accident on marine biota in the acute phase (Tateda et al., 2013; Vives i Batlle et al., 2014), closely following initial application of equilibrium models to make predictions (Garnier-Laplace et al., 2011).

Notwithstanding the availability of some models for dynamic situations, the availability of parameterisation data is a problem. There are many knowledge gaps, especially concerning elemental biological half-lives, and there are several types of model in use ranging from simple linear first order kinetic approaches to metabolic and foodchain transfer models. To date, there has been no international comparison of dynamic models for estimating biota exposure. For this reason, we decided to perform the first systematic comparison between such models within the International Atomic Energy Agency (IAEA) MODARIA programme (http://www-ns.iaea.org/projects/modaria/default.asp).

The focus of this study was to compare activity concentrations and exposures to biota calculated by dynamic transfer models; the location chosen for this model simulation was close to the point where radionuclides were released from the Fukushima Nuclear Power Plant to the Pacific Ocean during the reactor accident in March/April 2011. We used seven dynamic models: BURN-POSEIDON, the ANL approach, D-DAT, ECOMOD, the IRSN approach, K-BIOTA-DYN-M and the NRPA marine dynamic model; all models are described and referenced in Section 2.1 below. The predictions of these dynamic models were compared with the output from the equilibrium-based ERICA Tool. The input for the intercomparison was a series of hydrodynamic forecasts or monitoring data (activity concentrations in seawater and sediment) for a site close to the Fukushima nuclear complex for the 110 days after the accident, produced by means of marine dispersion models, as referenced below.

The resultant estimates should be considered as illustrative only, and not as a thorough assessment of exposures and effects at this site close to the Fukushima NPP. Such an evaluation using both model prediction and monitoring measurements can be found elsewhere (Vives i Batlle et al., 2014). The present study is based on model comparisons for a single location in close proximity to the release point, and thus the calculated activity concentrations in water and sediments used in the present study represent only a limited area. This area is not representative of the general region inhabited by populations of biota, since the gradients of the activity concentrations for both water and sediments are very pronounced (UNSCEAR, 2014). This is why the discussion of the results is limited to the numerical differences between the models and does not include an evaluation of the levels of exposures and possible effects on biota.

2. Materials and methods

2.1. Input data for the intercomparison

The inputs to the exercise were the modelled activity concentrations of ^{90}Sr, ^{131}I, and ^{137}Cs in near-surface water (top 1 m; Bq m^{-3}) as well as bottom seawater (Bq m^{-3}) and sediment (Bq kg^{-1}, dry mass – d.m.) given at daily intervals. The period of the simulation was fixed between 11 March and the end of June 2011 (^{90}Sr) and July (other two radionuclides), owing to the different setup of the model employed for ^{90}Sr. The radionuclide concentrations were obtained from a suite of marine dispersion models that have been previously validated and compared (Periáñez et al., 2015). Lagrangian models were used for ^{137}Cs and ^{131}I (Kawamura et al., 2011; Min et al., 2013) and an Eulerian model was applied in the case of ^{90}Sr (Periáñez et al., 2013). Essentially, these models utilise current fields pre-computed by operative three-dimensional hydrodynamic models to solve the transport of radionuclides in the sea. This is determined by advection due to currents and turbulent mixing. Interactions of radionuclides with sediments are described in a dynamic way, in terms of kinetic transfer coefficients. Both direct releases into the Pacific Ocean and deposition from the atmosphere were used as modelling source terms for each radionuclide.

For ^{131}I and ^{137}Cs, the model simulations were based on the source term estimated by the Japan Atomic Energy Agency (JAEA) from measurements made by the Tokyo Electric Power Company (TEPCO) at the point of discharge (Periáñez et al., 2015). In the case of ^{90}Sr, an inverse modelling technique was used to estimate the