Journal of Environmental Radioactivity xxx (2015) 1–10

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

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

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ARTICLE INFO

Article history:
Received 26 November 2014
Received in revised form
12 February 2015
Accepted 18 February 2015
Available online xxx

Keywords:
Marine environment
D-DAT
Dynamic model
Fukushima
Non-human biota

ABSTRACT

The dynamic model D-DAT was developed to study the dynamics of radionuclide uptake and turnover in biota and sediments in the immediate aftermath of the Fukushima accident. This dynamics is determined by the interplay between the residence time of radionuclides in seawater/sediments and the biological half-lives of elimination by 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.

A central element of the model is the inclusion of dynamic transfer of radionuclides to/from sediments by factorising the depletion of radionuclides adsorbed onto suspended particulates, molecular diffusion, pore water mixing and bioturbation, represented by a simple set of differential equations coupled with the biological uptake/turnover processes. In this way, the model is capable of reproducing activity concentration in sediment more realistically.

The model was used to assess the radiological impact of the Fukushima accident on marine biota in the acute phase of the accident. Sediment and biota activity concentrations are within the wide range of actual monitoring data. Activity concentrations in marine biota are thus shown to be better calculated by a dynamic model than with the simpler equilibrium approach based on concentration factors, which tends to overestimate for the acute accident period. Modelled dose rates from external exposure from sediment are also significantly below equilibrium predictions. The model calculations confirm previous studies showing that radioactivity levels in marine biota have been generally below the levels necessary to cause a measurable effect on populations. The model was used in mass-balance mode to calculate total integrated releases of 103, 30 and 3 PBq for ^{131}I, ^{137}Cs and ^{90}Sr, reasonably in line with previous estimates.

1. Introduction

The reactor failures at the Fukushima Dai-chi Nuclear Power Station (FDNPS) resulted in a major release of radionuclides to the surrounding environment. About 80% of the radioactive fallout (mainly ^{131}I and radiocaesium, with smaller quantities of ^{129,129m,132}Te, ^{136}Cs and ^{133}I) occurred over the Pacific ocean (Stohl et al., 2011). The magnitude of the marine release was realised shortly after the accident (Bailly du Bois et al., in press; Buesseler et al., 2011; Garnier-Laplace et al., 2011; IRSN, 2011; Tsumune et al., 2012; Vives i Batlle, 2011). ^{131}I and radiocaesium were the main radionuclides released into the marine environment, although ^{129,129m,132}Te, ^{136}Cs and ^{133}I were also released along with trace amounts of plutonium isotopes (Zheng et al., 2013).

The accident is by now well described (IAEA, 2011; IRSN, 2012; PMJHC, 2011; Povinec et al., 2013; UNSCEAR, 2014). The possible ecological consequences have been discussed (Garnier-Laplace et al., 2011; Kryshev et al., 2012; UNSCEAR, 2014; Vives i Batlle et al., 2014; Vives i Batlle and Vandenhove, 2013) and the general tenor of these studies is that there is no significant potential for effects except possibly for the relatively contaminated area in the vicinity of the discharge point, due to the short duration and the limited space area of the initially high exposures.

The first challenge in an exposure assessment for an accident like Fukushima is establishing radionuclide concentrations in the marine media: seawater, particulate matter and seabed sediments, either by field monitoring or by means of modelling (Aoyama et al., 2012; Choi et al., 2013; Honda et al., 2013). Then, the uptake and turnover of radionuclides by the marine biota need to be

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http://dx.doi.org/10.1016/j.jenvrad.2015.02.023
0265-931X/

determined. However, in the initial release phase, monitoring data are not always available, so it becomes necessary to use models.

Traditional transfer assessment approaches based on concentration factors are not applicable here because organisms are likely at disequilibrium with fluctuating activity concentrations in the environment (Kryshev et al., 2012; Psaltaki et al., in press; Tateda et al., 2013; Vives i Batlle and Vandenhove, 2013). Hence, it is not true to say that the activity concentration in an organism is proportional to the activity concentration in an adjacent volume of water, via a concentration factor (CF), as it is done in routine discharge assessments (Brown et al., 2008; Copplestone et al., 2001; IAEA, 2004; Yu et al., 2004). Likewise, the assumption that radionuclides in the water are in isotopic equilibrium with the sediments via a sediment distribution coefficient (K_d) is also not valid in accidental situations where not only the biota but also the sediments react with a time delay, starting from pre-accident (background) concentrations to much higher levels in a short timescale. In such cases, there are large variations of activity concentration in seawater due to spiked and/or widely fluctuating discharges. For such situations, a dynamic model capable of including exchange kinetics between the marine compartments (seawater, sediment, and biota) is called for.

Recent assessment studies of exposures to humans and non-human biota in Fukushima (Keum et al., 2015; Vives i Batlle et al., 2014) illustrate the above point very well for the case of exposures to biota in the marine environment. In the UNSCEAR study, the dynamic model ECOMOD (Sazykina, 2000) and the D-DAT Dynamic Dose Assessment Tool (Vives Batlle et al., 2008; Watts et al., 2008) were applied to calculate biota exposures at the time of the accident, with consistent results. They were able to approximate better the activity concentrations monitored in the local biota than the equilibrium models could do.

Once activity concentrations in biota are known, the problem becomes one of dose estimation. An international system for the radiological protection of the environment has been developed, which provides the framework for such calculations (Beresford et al., 2007b; Brown et al., 2008; ICRP, 2008, 2009), and the dose conversion coefficients and associated equations for the calculation of dose are the same for both equilibrium and dynamic type of assessments, a prime example of these being given in the methodology embodied by the ERICA assessment tool (Brown et al., 2008).

In the present paper, the D-DAT model has been redeveloped into an operational tool capable of calculating transfer to biota using biokinetic parameters for these radionuclides (short- and long-term biological half-lives), furthermore taking into account the dynamics of transfer to and from sediments, which improves calculation of external dose components – an important issue for benthic organisms and for retrofitting the likely radionuclide releases to the local marine environment.

2. Materials and methods

The primary sources of data were activity concentration in unfiltered surface seawater, sediment and fish covering the period up to August 2012, obtained from Japanese sources such as TEPCO (TEPCO, 2011) and MEXT (MEXT, 2011, 2013), complemented with published data (Buesseler and Aoyama, 2012; Buesseler et al., 2012; Honda et al., 2012). These were combined in the UNSCEAR quality-assured dataset of activity in unfiltered seawater, sediment and fish up to August 2012. We used this dataset with authorisation from UNSCEAR,[1] filtering the data based on a radial distance to the source point (FDNPS) of less than 5 km. Biota data were filtered to include species of fish only. Values reported as "less than" were excluded, leaving some 650 seawater (mostly from the Dai-ichi discharge channels), 20 sediment and 75 biota data points. For seawater, ^{134,137}Cs were monitored quite continuously over the first year. However, ^{90}Sr and ^{131}I measurements are available only up to 20 and 112 days, respectively, imposing a time limit on model simulations.

Initial inspection of the seawater activity concentration data reveals a period of acute discharges declining to about 10^7 Bq m^{-3} by 9 April 2011 (T = 29 days) (UNSCEAR, 2014), followed by a relatively fast exponential decrease between 35 and 40 days to levels generally below 2×10^5 Bq m^{-3}, and a sustained period of slower exponential decrease between 40 and 350 days. The sediment data (mainly ^{137}Cs) are scattered in space, with no obvious time trend except when isolating the few data points < 2 km from the FDNPS which removes data for $T < 125$ d but improves an exponential fit of the 16 remaining data: $A_S = 7.34 \times 10^3 \exp(-5.85 \times 10^{-3}T)$, $r^2 = 0.56$ with an associated clearance half-time of 118 days. A similar situation is apparent for the biota (mostly ^{137}Cs mostly for benthic fish), ranging 5–1300 Bq m^{-3} and for which weak decreasing trends are barely distinguishable. Some more clarity can be obtained by separating the data according to individual organism groups, as shown in Fig. 1. These data seem consistent with ecological half-lives ranging by a factor of 3: From 70 to 90 days from macroalgae and zooplankton to 200–220 days for crustacean and pelagic fish, with differences attributable to differences in occupancy in the water column and differences in trophic level. With the exception of polychaete (spurious correlation) and benthic fish (large amount of data with broad geographical dispersion), such information is broadly compatible with previous radiocaesium ecological half-lives for 16 Species in Marine Biota from the Fukushima area (Iwata et al., 2013).

The D-DAT dynamic model was initially developed in Excel for application to marine discharges from the Sellafield site in Cumbria, UK (Vives i Batlle, 2012), and subsequently adapted in 2011 for Fukushima studies. The basics of D-DAT have been previously described both in terms of its functionality (Vives Batlle et al., 2008; Watts et al., 2008) and its application to Fukushima (Vives i Batlle et al., 2014; Vives i Batlle and Vandenhove, 2013). The associated biokinetic – allometric data for ^{131}I and ^{134,137}Cs is also from previous work (Vives i Batlle et al., 2007) and was used in the recent UNSCEAR study (UNSCEAR, 2014), and then supplemented with ^{90}Sr data from other sources (Baptist et al., 1970; Boroughs et al., 1956; Casper et al., 2004; Freitas et al., 1988; Phillips and Russo, 1978; Polikarpov, 1965; Seixas and Pierce, 2005; Tagami and Uchida, 2013). A new model version was developed in a ModelMaker® 4 environment (Adamatzky, 2001; Citra, 1997; Rigas, 2000) (Fig. 2) with equations solved using the Gear integration method (Gear, 1971).

For the transfer to and from biota we applied a dual biological half-life ($T_{B1/2}$) approach, in essence a 3-compartment model (seawater + slow and fast biological compartments) reflecting the fact that in the most general case organisms depurate much of the radionuclide activity from seawater in their bodies via a short-term fast process followed by a longer-term slow process with short- and long-term biological half-lives $T^a_{B1/2}$ and $T^b_{B1/2}$, respectively, as well as radioactive decay (Vives i Batlle et al., 2005). The general solution is of the form:

$$q_i(t) = \frac{f_i}{\gamma\delta} + \frac{(q_i\gamma^2 - d_i\gamma + f_i)}{\gamma(\gamma-\delta)}e^{-\gamma t} + \frac{\left(q_i\delta^2 - d_i\delta + f_i\right)}{\delta(\delta-\gamma)}e^{-\delta t}, \quad i = 1,2,3$$

where q_1 is the activity in seawater (Bq m^{-3}), q_2 and q_3 represent the activity retained by the organism (Bq kg^{-1}, fresh mass) and the

[1] This study is fully independent from the preceding UNSCEAR study and should not be misinterpreted as a UNSCEAR position.

Please cite this article in press as: Vives i Batlle, J., Dynamic modelling of radionuclide uptake by marine biota: application to the Fukushima nuclear power plant accident, Journal of Environmental Radioactivity (2015), http://dx.doi.org/10.1016/j.jenvrad.2015.02.023