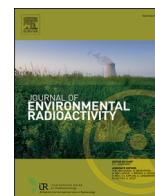




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## Best practices for predictions of radionuclide activity concentrations and total absorbed dose rates to freshwater organisms exposed to uranium mining/milling

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## ABSTRACT

Predictions of radionuclide dose rates to freshwater organisms can be used to evaluate the radiological environmental impacts of releases from uranium mining and milling projects. These predictions help inform decisions on the implementation of mitigation measures. The objective of this study was to identify how dose rate modelling could be improved to reduce uncertainty in predictions to non-human biota. For this purpose, we modelled the activity concentrations of <sup>210</sup>Pb, <sup>210</sup>Po, <sup>226</sup>Ra, <sup>230</sup>Th, and <sup>238</sup>U downstream of uranium mines and mills in northern Saskatchewan, Canada, together with associated weighted absorbed dose rates for a freshwater food chain using measured activity concentrations in water and sediments. Differences in predictions of radionuclide activity concentrations occurred mainly from the different default partition coefficient and concentration ratio values from one model to another and including all or only some <sup>238</sup>U decay daughters in the dose rate assessments. Consequently, we recommend a standardized best-practice approach to calculate weighted absorbed dose rates to freshwater biota whether a facility is at the planning, operating or decommissioned stage. At the initial planning stage, the best-practice approach recommend using conservative site-specific baseline activity concentrations in water, sediments and organisms and predict conservative incremental activity concentrations in these media by selecting concentration ratios based on species similarity and similar water quality conditions to reduce the uncertainty in dose rate calculations. At the operating and decommissioned stages, the best-practice approach recommends relying on measured activity concentrations in water, sediment, fish tissue and whole-body of small organisms to further reduce uncertainty in dose rate estimates. This approach would allow for more realistic but still conservative dose assessments when evaluating impacts from uranium mining projects and making decision on adequate controls of releases.

IAEA, International Atomic Energy Agency.

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## 1. Introduction

The International Atomic Energy Agency (IAEA) recommends member countries to use Radiological Environmental Impact Assessment (REIA) to identify mitigation method to protect the environment against ionizing radiation (IAEA 2010). Canada follows the IAEA recommendation as REIA are central to the safe use of nuclear energy (CNSC 2017). In Canada, a REIA is an environmental risk assessment (ERA) specific to radionuclides. An ERA is a systematic process that identifies, quantifies and characterizes the risk posed by radionuclides released in the environment (Canadian Standard Association Group, 2012). It is used at all stages of the life-cycle of a nuclear facility. The ERA process evolves with the lifecycle of a nuclear facility (CNSC 2017). At the planning stage, an initial ERA for a new facility is predictive as it involves assessing the likelihood of potential future effects. The initial ERA uses the planned radionuclide releases to predict the source terms of gaseous and liquid discharges, the transport of radionuclides through the environment, the total radionuclide activity concentrations in water and sediment that includes baseline activity concentrations, its transfer to representative biota and associated predicted total absorbed dose rates. These initial ERA predictions of radionuclide activity concentrations and associated total absorbed dose rates serve as the basis to evaluate the adequate control on radionuclide releases from the facility. Hence, the ERAs at the planning stage are conservative in nature to provide an adequate margin of safety to account for uncertainties in predictions. As a facility moves through its lifecycle, the ERA is periodically reviewed and revised using the accumulated site knowledge (mainly radionuclide activity concentrations in water, sediment and fish tissue) obtained from the environmental monitoring program, special investigations, incorporation of advances in scientific knowledge and, where available, indigenous traditional knowledge. These “living” ERAs are less uncertain and are used to assess if the initial conservative environmental impact predictions are exceeded or may be exceeded in the future. If exceeded, the ERA process allows for consideration of additional mitigation measures, a process often referred to as adaptive management. In this manner, the initial predictive and conservative ERA evolves through the life of the facility or activity, remaining current and becoming an increasingly more powerful and realistic or less conservative site-specific tool to protect the environment. The Canadian regulatory process hence provides significant monitoring information that can be used to improve total absorbed dose rate modelling.

As initial ERAs are forward-looking, uncertainty in radiological dose rate predictions is at its highest. Initial ERAs use upper-bound predictions to account for the uncertainty. However, overly conservative predictions may raise unnecessary public concern. It is therefore important to obtain realistic worst-case (i.e., conservative) predictions at the planning stage of a project. Such predictions of radiological dose rates to non-human biota rely on adequate knowledge of radionuclide transport and partitioning between water and sediments (Monte et al., 2009; Lofts et al., 2015). Partitioning of radionuclides in freshwater environments depend on their activity concentrations, the quantity of dissolved and particulate phases and the affinity of radionuclides to the different phases. Radionuclides adsorbed to the particulate phase will settle in sediments while those associated with dissolved phases are more likely to remain in water. Radionuclides adsorbed onto the particulate phase will likely be deposited on the surface of sediments. Deposition of particulate radionuclides will be dictated by the hydraulic regime where low flow conditions will favour deposition. Under low flow conditions, the quantity and density of particles will dictate the settling velocity of particles and rate of accumulation of radionuclides in sediments. Under high flow conditions, particulate radionuclides will be transported downstream until reaching lentic systems where they will settle (Chapra 1997). These complex processes contribute to the uncertainty in ERA predictions.

Another source of uncertainty is the prediction of the accumulation of radionuclides by freshwater organisms. It is often simplified in dose

rate assessments by multiplying each radionuclide activity concentration in water by a generic/default conservative concentration ratio (CR) (Howard et al., 2013; Yankovich et al., 2013) or so-called bio-concentration factor (BCF) in metal ecotoxicology (McGeer et al., 2009). A database of CRs was created and made available online (<http://www.wildlifetransferdatabase.org/>) with the objective of providing best estimates of generic CRs for the development of approaches to assess the radiological risks to non-human biota (ICRP 2009; Howard et al., 2013). Although this initiative helped to improve dose assessments to non-human biota, uncertainties remain, and improvements are needed to reduce them (Beresford et al., 2009; Beresford et al., 2010; Bradshaw et al., 2014; Brechignac et al., 2016; Stark et al., 2017). For instance, CRs depend on site-specific water and sediment chemistry, which condition the accumulation of radionuclides via processes similar to those observed for metals (DiToro et al., 1992). For example,  $^{226}\text{Ra}$  accumulation by freshwater bivalves depends on pH and calcium (Jeffree 1991; Jeffree and Simpson 1984), while its strong affinity to dissolved organic matter in water increases its adsorption, possibly reducing its uptake (IAEA 2013). Similarly, the accumulation of  $^{238}\text{U}$  species by aquatic biota is affected by pH and may decrease with increasing concentrations of calcium, alkalinity and dissolved organic carbon in water (Markich, 2002; Sheppard et al., 2005; Fortin et al., 2007; Goulet et al., 2011; van Dam et al., 2012; Goulet et al., 2015). For organisms exposed to sediments, the uptake of metals and radionuclides of metallic elements, may also be affected by the presence of binding phases in sediments such as iron oxides (Tessier et al., 1993), organic matter and sulfidic minerals (DiToro et al. 1992, 2005).

Some uncertainty also arises with the estimation of internal and external dose rates. In particular, external dose is associated with the radionuclides in the surrounding environmental media consisting primarily of water and sediments. The ICRP (2017) recommends a framework for external dose rate calculations. Uncertainties in external dose rate assessments arise from vertical and spatial heterogeneity in sediment activity concentrations and with estimating the time spent in each medium and the positioning of the organisms (in, on or above the contaminated medium). Both internal and external dose rates can then be calculated for predefined organism dimensions or calculated based on the dimensions of a user-defined organism (Ulanovsky and Pröhl, 2006; Ulanovsky et al., 2008; Amato and Italiano 2014; Brown et al., 2016; Ulanovsky 2016). The different dosimetry methods used by the available models add little to the overall uncertainty in estimated dose (Vives i Batlle et al., 2007; 2011).

Finally, another source of uncertainty is the application of a relative biological effectiveness (RBE). For alpha emitters (i.e.,  $^{210}\text{Po}$ ,  $^{226}\text{Ra}$ ,  $^{230}\text{Th}$ ,  $^{234}\text{U}$ , and  $^{238}\text{U}$ ), a radiation weighting factor may be used to account for RBE when estimating internal dose. Kocher and Trabalka (2000) and Chambers et al. (2006) recommended applying an alpha radiation weighting factor from 5 to 20 to account for increased biological effectiveness, the ICRP have recently recommended that a value of 10 should be used (ICRP 2021).

While there are several papers published comparing predictions of radiological dose to biota using the different assessment models developed over the last 20 years (e.g. Vives i Batlle et al. 2007; Vives i Batlle et al., 2011; Beresford et al., 2010; Stark et al., 2015; Johansen et al., 2012; Yankovich et al., 2010a,b), no such comparison has been conducted for releases to the aquatic environment from uranium mines and mills. Given that assessments for uranium industry facilities are being conducted in a number of countries (e.g. Beaugelin-Seiller et al., 2008; Camus et al., 1999; Thomas and Liber 2001; Bird et al., 2003) an evaluation of the available modelling approaches would be useful.

The objectives of this study were to compare estimates obtained by different models of predicted dose rates for freshwater organisms exposed to operating and decommissioned uranium mines and mills; to identify the main uncertainties in dose rates predictions; and to provide recommendations on how to reduce uncertainty and conservatism when modelling dose rates to non-human biota at different stages of the

uranium life-cycle.

## 2. Methods

### 2.1. Overview of models used

The initiative presented in this paper was undertaken within the second phase of the Environmental Modelling for Radiation Safety (EMRAS) program under the auspice of the International Atomic Energy Agency from 2009–11. Under this program, a group of international experts participated in a radionuclide accumulation and dose modelling exercise using information collected downstream from uranium mines and mills in northern Saskatchewan, Canada. All models/approaches and their application are summarized within [Table 1](#). The models applied by participants included the software code RESRAD-BIOTA implementing the USDOE Graded approach ([USDOE, 2004](#)), which has since been updated in 2019 ([USDOE, 2019](#)) (The 2004 approach was used in this exercise); the ERICA Tool ([Brown et al., 2008](#)) initially developed by a European Union funded consortium ([Larsson 2008](#)),

**Table 1**

Summary of participants and the models/approaches they applied when predicting activity concentrations (Exercise I), dose rates (Exercise II) and dose rates using the recommended standardized best approach to reference benthic invertebrates and fish downstream of operating and decommissioned uranium mines and mills.

Model/ Approach	Transfer/ dosimetry	Participants (country)	Exercise	References(s)
RESRAD-BIOTA	Y/Y	Argonne National Laboratory (ANL) (the United States of America) Australian Nuclear Science and Technology Organization (ANSTO) (Australia) University of Salford (United Kingdom, UK)	I, II  I  I	<a href="#">USDOE (2004)</a>
ERICA Tool	Y/Y	ANSTO (Australia) UK Centre for Ecology & Hydrology (UK CEH) (UK) Environmental Agency (UK EA) Jožef Stefan Institute (JSI) (Slovenia) Norwegian Radiation Protection Authority (DSA) (Norway) Belgian Nuclear Research Centre (SCK-CEN) (Belgium)	I I <sup>a</sup> , II  I, II, III  II  I <sup>b</sup>  I, II, III	<a href="#">Brown et al. (2008)</a> <a href="#">Larsson (2008)</a>
R&D 128	Y/Y	England and Wales Environment Agency (EA) (UK)	I	<a href="#">Coppelstone et al. (2001)</a>
K-BIOTA	Y/Y	Korea Atomic Energy Research Institute (KAERI) (S. Korea)	I, II, III	<a href="#">Keum et al. (2011)</a>
EDEN	N/Y	Institut de Radioprotection et de Sûreté Nucléaire (IRSN) (France)	II	<a href="#">Beaugelin-Seiller et al. (2006)</a>
Hokus	Y/Y	State Office for Nuclear Safety (SÚJB) (Czech Republic)	I, II	

<sup>a</sup> Provided <sup>210</sup>Po predictions in Fingernail Clam and <sup>210</sup>Po and <sup>226</sup>Ra in White Sucker and Lake Whitefish only.

<sup>b</sup> Provided <sup>210</sup>Po and <sup>210</sup>Pb in Fingernail Clam and White Sucker and Lake Whitefish only.

updated in 2016 ([Brown et al., 2016](#)) and recently update in July 2021 (the 2008 version was used in this paper); R&D 128 ([Coppelstone et al., 2001; 2003](#)) the model developed by the England and Wales Environment Agency; the Elementary Dose Evaluation for Natural Environment (EDEN), a dosimetry tool that calculates dose to biota from exposure information, developed by the Nuclear Safety and Radioprotection Institute of France ([Beaugelin-Seiller et al., 2006](#)); K-BIOTA developed by the Korea Atomic Energy Research Institute ([Keum et al., 2011](#)); and, Hokus developed by State Office of Nuclear Safety of the Czech Republic. To differentiate between participants using the same model, the model and participant are identified as, for instance, RESRAD (ANSTO) denoting that participants from the Australian Nuclear Science and Technology Organization (ANSTO) applied the RESRAD-BIOTA model.

### 2.2. Data sources and quality assurance

At first, a dataset was created by selecting 6 non-affected areas (i.e. reference sites) and 19 affected areas (i.e. exposure sites) associated with former uranium mines and mills in Northern Saskatchewan, Canada. As a legal requirement from the Canadian Nuclear Safety Commission who acts as the regulating body, environmental monitoring programs exist at these sites to monitor the health of freshwater ecosystems and were the source of the data for both exercise I & II (See [Tables 2 and 4](#)). Water, sediment and biota were collected by third party contractors with a vast experience in environmental monitoring in Northern Saskatchewan. Collected water, sediment and biota were analysed by the Saskatchewan Research Council, an accredited laboratory which used quality assurance and quality control to ensure reliability of the radionuclide activity concentration data. Example of quality assurance and quality control were field blanks, use of water, sediment and tissue standards during digestions and during radionuclide analyses.

### 2.3. Exercise I: predicting activity concentrations in organisms

For exercise I, two reference sites (Ref 1–2) and 11 exposure sites (Exposure 1–11) were selected. The first step of modelling the dose rates to non-human biota was to predict the partitioning of radionuclides between water and sediments in the receiving environment where they are not known. Most approaches use partition coefficients ( $K_d$  in  $L \cdot kg^{-1}$  dry mass) to describe the partitioning of radionuclides between sediment and water:

$$K_d = \frac{[RN]_{sed}}{[RN]_{water}} \quad (1)$$

Where  $[RN]_{water}$  and  $[RN]_{sed}$  are the total activity concentrations of a radionuclide in water ( $Bq \cdot L^{-1}$ ) and sediment ( $Bq \cdot kg^{-1}$  dry mass). The second step was to estimate the activity concentrations of radionuclides in organisms where they are not known. The applied approaches use equilibrium concentration ratios ( $CR_{wo-water}$ ) to predict radionuclide activity concentrations in the aquatic organisms considered here from media activity concentrations ([IAEA 2014](#)) where  $CR_{wo-water}$  is defined as:

$$CR_{wo-water} = \frac{[RN]_{wo}}{[RN]_{water}} \quad (2)$$

Where  $[RN]_{wo}$  and  $[RN]_{water}$  are radionuclide activity concentrations in whole organism body ( $Bq \cdot kg^{-1}$  fresh mass) and in (nominally) filtered water ( $Bq \cdot L^{-1}$ ).

For Exercise I, modellers predicted activity concentrations in non-human biota using site-specific water and sediment radionuclide activity concentrations ([Table 2](#)). When radionuclide activity concentrations were not available in water but were given for sediments or vice versa, Equation (1) was used to calculate the radionuclide activity concentration in a given media using the arithmetic mean partition coefficients for the exposed sites ([Table 2](#)) or, if site-specific data were not available,

**Table 2**

Arithmetic mean concentrations of radionuclides in water and sediments in reference (Ref) and exposed (Exp) sites used in the modelling exercises I, II and the standardized best practice.

Site	Water (mBq·L <sup>-1</sup> )					Sediment (Bq·kg <sup>-1</sup> dm)					Partition coefficient (L·kg <sup>-1</sup> dm)				
	<sup>210</sup> Pb	<sup>210</sup> Po	<sup>226</sup> Ra	<sup>230</sup> Th	<sup>238</sup> U	<sup>210</sup> Pb	<sup>210</sup> Po	<sup>226</sup> Ra	<sup>230</sup> Th	<sup>238</sup> U	<sup>210</sup> Pb	<sup>210</sup> Po	<sup>226</sup> Ra	<sup>230</sup> Th	<sup>238</sup> U
Ref 1	20	7.5	6.5	6.5	150	82	82	36	24	20	4100	11,000	5500	370	1300
Ref 2	20	5	5	1	10	130	138	36	74	62	6500	28,000	7200	74,000	13,000
Ref 3	–	–	–	–	8.55	–	–	–	–	–	–	–	–	–	–
Ref 4	–	–	–	–	–	626	394	86	–	23	–	–	–	–	–
Ref 5	–	–	7.0	–	2	–	–	73	–	100	–	–	11,000	–	41,000
Ref 6	20	5.0	5.0	10	10	396	410	60	43	238	20,000	82,000	12,000	4300	24,000
<b>Arithmetic mean</b>											<b>10,200</b>	<b>40,300</b>	<b>8900</b>	<b>26,200</b>	<b>19,800</b>
Exp 1	50	10	6	15	13	260	210	160	720	940	4700	21,000	27,000	48,000	70,000
Exp 2	20	–	15	10	2000	253	–	860	–	1610	–	–	57,000	–	830
Exp 3	50	30	100	10.0	7200	30,600	29,800	5480	1740	465,000	610,000	990,000	55,000	170,000	65,000
Exp 4	200	–	100	–	5500	12,700	–	6130	–	43,500	60,000	–	61,000	–	7900
Exp 5	130	50	2150	35.0	6000	47,600	47,800	45,400	23,200	26,600	380,000	960,000	21,000	660,000	4400
Exp 6	50	–	45	–	1900	–	–	3430	–	2130	–	–	76,000	–	1100
Exp 7	20	–	120	–	2400	4730	–	1520	11	2450	240,000	–	13,000	–	1000
Exp 8	150	–	39	10	1900	1750	–	971	7270	18,300	12,000	–	25,000	73,000	9800
Exp 9	80	40	790	110	130	25,000	18,700	24,000	35,000	1010	310,000	470,000	30,000	320,000	7600
Exp 10	40	30	140	65	1600	13,000	13,300	16,300	47,300	1170	330,000	440,000	110,000	730,000	73,000
Exp 11	20	5	20	10	2700	280	240	300	410	2500	14,000	48,000	15,000	41,000	930
Exp 12	–	–	6	–	–	350	358	350	28	95	–	–	–	–	–
Exp 13	–	–	–	–	–	155	163	35	25	14	–	–	–	–	–
Exp 14	–	–	–	–	–	88	230	88	47	524	–	–	–	–	–
Exp 15	–	–	–	–	–	662	372	116	–	144	–	–	–	–	–
Exp 16	–	–	–	–	2500	452	358	160	–	6590	–	–	–	–	–
Exp 17	–	–	–	–	–	65	117	61	13	14	–	–	–	–	–
Exp 18	60	–	53	–	2050	–	–	–	–	–	–	–	–	–	–
Exp 19	–	–	1500	–	9000	8660	8370	10,100	–	18,000	–	–	–	–	–
<b>Arithmetic mean</b>											<b>217,800</b>	<b>488,100</b>	<b>44,500</b>	<b>291,700</b>	<b>21,900</b>

using default parameters provided by the different models (Table 3). The modellers then estimated the activity concentrations of <sup>210</sup>Pb, <sup>210</sup>Po, <sup>226</sup>Ra, <sup>230</sup>Th, and <sup>238</sup>U for a simplified freshwater benthic food chain comprised of small freshwater Fingernail Clams (e.g., *Pisidium* sp.) and benthic foraging fish (e.g., Lake Whitefish: *Coregonus clupeaformis*). Fingernail Clams were selected as representative of benthic invertebrates since they are believed to be sensitive to exposure from uranium mine/mill effluents and appear to be exposed to higher internal dose rates (Kilgour et al., 2018; Doering et al., 2019). Lake Whitefish were selected as they feed on Fingernail Clams (Pothoven 2009) and are exposed to both water and sediments.

The levels of radionuclides in biota were estimated with Equation (2) using water concentrations (Table 2) and then multiplied by the model default CRs (Table 3). The modelled predictions of activity concentrations in fish were compared to measured values only at reference site 2 and exposure sites 1, 4, 8 and 10 since radionuclide activity concentrations in benthic fish (i.e. Lake Whitefish) were available at these sites. Measured values were available for Lake Whitefish but were not available for Fingernail Clams (Table 4). This comparison provided an indication of the conservativeness of the modelled predictions. Since radionuclide measurements were available for both fish flesh and bone, we calculated whole organism body activity concentrations of radionuclides in fish using the following equation:

$$[RN]_{wo} = 0.85 \times [RN]_{flesh} + 0.15 \times [RN]_{bone} \quad (3)$$

where  $[RN]_{wo}$ ,  $[RN]_{flesh}$  and  $[RN]_{bone}$  are the activity concentrations of radionuclides in fish whole organism body, flesh and bones, respectively. The coefficients preceding  $[RN]_{flesh}$  and  $[RN]_{bone}$  are the proportions of flesh and bone to total fish body mass, respectively. The coefficient for bone proportion comes from the ERICA Tool (version 1.2.1) help file, based on a report that the skeleton accounted for 16% of the total body mass of six Channel Catfish (*Ictalurus punctatus*) reared in the laboratory (Cameron 1975). The coefficient preceding  $[RN]_{flesh}$  was obtained by assuming that the remaining fish mass was attributed to

muscle tissue.

#### 2.4. Exercise II: dose rate estimation

In this second exercise, participants predicted activity concentrations in organisms and then calculated dose rates to freshwater organisms. For exercise II, we selected two new reference sites (reference sites 3 & 4), selected five decommissioned exposure sites from exercise I (i.e. exposure sites 1, 2, 3, 8, 9) and added 8 exposure sites downstream of operating mine sites. This was done to have a more comprehensive database that reflected both operating and decommissioned mining and milling sites in Canada. Modellers calculated total radiation dose rates from internal and external exposure to radionuclides for each freshwater organism using the following equations (Brown et al., 2008):

$$\sum_{RN} D_{int,RN} = \sum_{RN} (DC_{int,RN} \times [RN]_{wo}) \quad (4)$$

$$\sum_{RN} D_{ext,RN} = \sum_{RN} DC_{ext,RN} \times [OF_{water} \times [RN]_{water} + OF_{sed} \times [RN]_{sediment}] \quad (5)$$

Where:

$D_{int, RN}$  = internal radiation dose rate for the radionuclide RN ( $\mu\text{Gy}\cdot\text{h}^{-1}$ )

$D_{ext, RN}$  = external radiation dose rate for the radionuclide RN ( $\mu\text{Gy}\cdot\text{h}^{-1}$ )

$DC_{int, RN}$  = dose coefficient for the radionuclide RN in whole organism body ( $\mu\text{Gy}\cdot\text{h}^{-1}/\text{Bq}^{-1}\cdot\text{kg}$ )

$DC_{ext, RN}$  = external dose coefficient for a given radionuclide RN in water or sediment ( $\mu\text{Gy}\cdot\text{h}^{-1}/\text{Bq}^{-1}\cdot\text{kg}$  (or  $\text{Bq}\cdot\text{L}^{-1}$ )

$OF_w$  = fraction of time spent in water

$OF_{sed}$  = fraction of time spent in the sediments

$[RN]_{wo}$  = whole organism body activity concentration for the radionuclide RN ( $\text{Bq}\cdot\text{kg}^{-1}$  fresh mass)

**Table 3**

Models (organization) and parameters (partition coefficient  $K_d$  and concentration ratio CR) used in predicting activity concentrations in organisms (Exercise I).

Model (organization)	Radionuclides	$K_d$ (L·kg <sup>-1</sup> dm)	CR (L·kg <sup>-1</sup> fm whole body)	
			Fish (all species)	Fingernail Clam
ERICA (ANSTO)	<sup>210</sup> Pb	100,000	300	1700
	<sup>210</sup> Po	20,000,000	240	38,000
	<sup>226</sup> Ra	15,200	80	1500
	<sup>230</sup> Th	18,400,000	110	100
	<sup>238</sup> U	50	30	180
ERICA (CEH)	<sup>210</sup> Pb	nc <sup>a</sup>	Nc	nc
	<sup>210</sup> Po	20,000,000	4210	13,200
	<sup>226</sup> Ra	15,200	98.4	nc
	<sup>230</sup> Th	nc	nc	nc
	<sup>238</sup> U	nc	nc	nc
ERICA (EA UK)	<sup>210</sup> Pb	100,000	790	140
	<sup>210</sup> Po	20,000,000	4900	130,000
	<sup>226</sup> Ra	15,200	73	8600
	<sup>230</sup> Th	18,400,000	4900	31,000
	<sup>238</sup> U	50	63	31,000
ERICA (DAS)	<sup>210</sup> Pb	100,000	344	533
	<sup>210</sup> Po	20,000,000	7520	1735
	<sup>226</sup> Ra	15,200	nc	nc
	<sup>230</sup> Th	18,400,000	nc	nc
	<sup>238</sup> U	50	nc	nc
ERICA (SCK CEN)	<sup>210</sup> Pb	244,000	370	1400
	<sup>210</sup> Po	420,000	240	38,000
	<sup>226</sup> Ra	41,900	80	1500
	<sup>230</sup> Th	352,000	110	100
	<sup>238</sup> U	20,300	30	180
Hokus (SUJB)	<sup>210</sup> Pb	200,000	300	1700
	<sup>210</sup> Po	30	240	38,000
	<sup>226</sup> Ra	70	80	1500
	<sup>230</sup> Th	60,000	110	100
	<sup>238</sup> U	50	30	180
K-BIOTA (KAERI)	<sup>210</sup> Pb	100,000	380	1700
	<sup>210</sup> Po	20,000,000	240	38,000
	<sup>226</sup> Ra	15,200	287	1500
	<sup>230</sup> Th	18,400,000	200	100
	<sup>238</sup> U	50	17	180
RESRAD-BIOTA (ANL)	<sup>210</sup> Pb	20,000	300	1700
	<sup>210</sup> Po	30	240	38,000
	<sup>226</sup> Ra	70	80	1500
	<sup>230</sup> Th	60,000	110	100
	<sup>238</sup> U	50	30	180
RESRAD-BIOTA (ANSTO)	<sup>210</sup> Pb	20,000	300	1700
	<sup>210</sup> Po	30	500	38,000
	<sup>226</sup> Ra	70	3200	1500
	<sup>230</sup> Th	60,000	80	100
	<sup>238</sup> U	50	1000	1000
RESRAD-BIOTA (U of Salford)	<sup>210</sup> Pb	20,000	300	300
	<sup>210</sup> Po	30	500	38,000
	<sup>226</sup> Ra	70	80	1500
	<sup>230</sup> Th	60,000	110	80
	<sup>238</sup> U	50	30	1000
R&D 128	<sup>210</sup> Pb	1000	10,900	5230
	<sup>210</sup> Po	2700	50	102,000
	<sup>226</sup> Ra	50	70	1370
	<sup>230</sup> Th	50	70	1370
	<sup>238</sup> U	100,000	10	1800

<sup>a</sup> nc exercise not completed.

$[RN]_{sed}$  = radionuclide activity concentration for the radionuclide RN in sediments (Bq·kg<sup>-1</sup> fresh mass)

$[RN]_{water}$  = radionuclide activity concentration for the radionuclide RN in water (Bq·L<sup>-1</sup>)

For organisms that are present at the sediment-water interface,  $OF_w$  and  $OF_{sed}$  values were taken from Table 5. Modellers calculated weighted absorbed dose rates applying in effect equations (4) and (5) to Fingernail Clam, large benthic foraging fish (White Sucker and Lake Whitefish) and pelagic foraging fish (Northern Pike, *Esox lusius*) and

Lake Trout (*Salvelinus namaycush*). For fish, modellers used site-specific radionuclide activity concentrations in the flesh and bones when available (Table 4) to estimate whole organism activity concentrations using Equation (3). When site-specific activity concentrations for flesh and bone were not available, modellers had to estimate whole organism radionuclide activity concentrations using concentration ratios. They had to specify which concentration ratios they used. The organism geometry, mass and occupancy information required for the calculation of external dose rates were provided to the modellers for the three organism types (Table 5). Modellers used default dose coefficients from the model applied, making, if necessary, a correspondence with an organism present in the model database. Modellers also assumed decay chain daughters (generally a default assumption in the models was used) and applied radiation weighting factors (Table 6).

### 3. Results and discussion

#### 3.1. Variability in partition coefficients

In Exercise I, partition coefficients ( $K_d$ ) were used to estimate water concentrations when values were not available. Modellers were given freedom in the choice of  $K_d$  by either using the site-specific values provided in Table 2 or by using the default  $K_d$  included in their models (Table 3). Fig. 1 provides the range of  $K_d$  values used by modellers for each radionuclide across the selected sites. Partition coefficients for a given radionuclide varied by several orders of magnitude. The <sup>210</sup>Pb  $K_d$  values varied from the default 1000 L kg<sup>-1</sup> dry mass (dm) of the R&D 128 model (Table 3) to 218,000 L kg<sup>-1</sup> dm obtained by calculating the mean activity concentrations in sediments and water from all exposed sites (Table 2). At reference sites, the mean  $K_d$  value of 10,200 L kg<sup>-1</sup> dm was lower than at exposure sites (Table 2). For <sup>210</sup>Po, the  $K_d$  values ranged from 30 L kg<sup>-1</sup> dm in the RESRAD-BIOTA application to 20,000,000 L kg<sup>-1</sup> dm in the ERICA Tool (Table 3). The mean <sup>210</sup>Po  $K_d$  values at reference and exposed sites were 40,333 L kg<sup>-1</sup> dm and 488,000 L kg<sup>-1</sup> dm, respectively (Table 2). For <sup>226</sup>Ra and <sup>230</sup>Th,  $K_d$  values ranged from 50 L kg<sup>-1</sup> dm in the R&D 128 application to 18,400,000 L kg<sup>-1</sup> dm in the ERICA Tool (Table 3). The mean <sup>226</sup>Ra  $K_d$  at reference sites and exposed sites were 8900 L kg<sup>-1</sup> dm and 44,500 L kg<sup>-1</sup> dm, respectively (Table 2). For <sup>230</sup>Th,  $K_d$  values at reference and exposed sites were 26,200 L kg<sup>-1</sup> dm and 291,700 L kg<sup>-1</sup> dm, respectively (Table 2). Finally, the ERICA Tool (SCK) used the <sup>238</sup>U average  $K_d$  value of 21,900 L kg<sup>-1</sup> dm (Table 2). In contrast, the R&D128 used a default value of 100,000 L kg<sup>-1</sup> dm and Hokus a value of 34,000 L kg<sup>-1</sup> dm. These values were three orders of magnitude above the default value of 50 L kg<sup>-1</sup> dm suggested in the RESRAD-BIOTA and the ERICA Tool (Table 3). In contrast to other radionuclides, the mean  $K_d$  values for <sup>238</sup>U at reference sites and exposure sites were more or less similar with higher variability at exposed sites (Table 2). For other radionuclides, the  $K_d$  at reference sites was lower than the mean  $K_d$  at exposure sites (Table 2).

#### 3.2. Variability in concentration ratios

Variability in the concentration ratios (CRs) applied was a key source of uncertainty in predicting activity concentrations in freshwater biota, when water activity concentrations were provided at a given site (Fig. 2; Table 2). Similar to partition coefficients, CRs varied in the different models (Fig. 2). In addition, there was also variation in concentrations within species. Using Fingernail Clams as an example of benthic invertebrates, <sup>210</sup>Pb CRs ranged from 140 L kg<sup>-1</sup> in ERICA Tool (UK) to 5230 L kg<sup>-1</sup> fm in the R&D128 (Fig. 2, lower panel; Table 3). <sup>210</sup>Po had the highest CR compared to other radionuclides, with values between 1700 L kg<sup>-1</sup> fm (ERICA Tool (DSA)) and 102,000 L kg<sup>-1</sup> fm (R&D 128). For <sup>226</sup>Ra, most CRs were around 1500 L kg<sup>-1</sup> fm. For <sup>230</sup>Th, most modellers used a CR of 100 L kg<sup>-1</sup> fm, however, the R&D 128 used a CR of 1370 L kg<sup>-1</sup> fm. Finally, <sup>238</sup>U values ranged from 120 to 1800 L kg<sup>-1</sup>

**Table 4**

Arithmetic mean  $\pm$  standard deviation of calculated radionuclide activity concentrations (Bq·kg<sup>-1</sup> fresh mass) in whole body Northern Pike, Lake Trout, Lake Whitefish, Longnose Sucker and White Sucker using Equation (3) at sites considered in the modelling exercises I, II and standardized best practice.

Site	Species	[ <sup>210</sup> Pb]	[ <sup>226</sup> Ra]	[ <sup>230</sup> Th]	[ <sup>210</sup> Po]	[ <sup>238</sup> U]
Ref 2 <sup>a</sup>	Lake Whitefish ( <i>Coregonus clupeaformis</i> )	1.3 $\pm$ 0.8	0.4 $\pm$ 0.2	0.5 $\pm$ 0.2	0.8 $\pm$ 0.4	0.6 $\pm$ 0.7
Ref 4	Northern Pike ( <i>Esox lucius</i> )		0.4 $\pm$ 0.1		1.8 $\pm$ 0.7	
	Lake Whitefish ( <i>Coregonus clupeaformis</i> )		0.4 $\pm$ 0.1		1.1 $\pm$ 0.4	
	Arithmetic mean	<b>1.3 <math>\pm</math> 0.8</b>	<b>0.4 <math>\pm</math> 0.1</b>	<b>0.5 <math>\pm</math> 0.2</b>	<b>1.2 <math>\pm</math> 0.5</b>	<b>0.6 <math>\pm</math> 0.7</b>
Exp 1 <sup>a</sup>	Lake Whitefish ( <i>Coregonus clupeaformis</i> )	1.2	0.4 $\pm$ 0.3	0.6 $\pm$ 0.2	1.0 $\pm$ 0.4	2.4 $\pm$ 2.6
	Northern Pike ( <i>Esox lucius</i> )		0.1 $\pm$ 0.1	0.4 $\pm$ 0.2	1.4 $\pm$ 0.8	0.1 $\pm$ 0.1
Exp 3	Northern Pike ( <i>Esox lucius</i> )		15.4 $\pm$ 5.3		40.5 $\pm$ 18.2	9.3 $\pm$ 1.9
Exp 4 <sup>a</sup>	Lake Whitefish ( <i>Coregonus clupeaformis</i> )	1.2 $\pm$ 0.5	5.4 $\pm$ 1.2			20.5 $\pm$ 9.1
Exp 7	Lake Trout ( <i>Salvelinus namaycush</i> )	4.8 $\pm$ 1.7	1.4 $\pm$ 0.7			5.6 $\pm$ 4.4
	White Sucker ( <i>Catostomus commersoni</i> )	3.0 $\pm$ 1.6	8.2 $\pm$ 4.3			15.2 $\pm$ 2.3
Exp 8 <sup>a</sup>	Lake Whitefish ( <i>Coregonus clupeaformis</i> )	2.7 $\pm$ 1.5	1.6 $\pm$ 0.7			22 $\pm$ 28
Exp 9	Northern Pike ( <i>Esox lucius</i> )	1.6 $\pm$ 1.3	3.0 $\pm$ 1.1	1.9 $\pm$ 2.9	95.2 $\pm$ 68.3	0.05 $\pm$ 0.003
Exp 10 <sup>a</sup>	Lake Whitefish ( <i>Coregonus clupeaformis</i> )	6.3 $\pm$ 5.8	12.2 $\pm$ 9.6	1.9 $\pm$ 1.6	10.8 $\pm$ 8.3	5.3 $\pm$ 2.5
Exp 13	Northern Pike ( <i>Esox lucius</i> )		0.2		1.4 $\pm$ 0.6	
	Lake Whitefish ( <i>Coregonus clupeaformis</i> )					0.7 $\pm$ 0.6
Exp 14	Northern Pike ( <i>Esox lucius</i> )		0.8 $\pm$ 0.3		1.3 $\pm$ 0.2	0.1
	Longnose sucker ( <i>Catostomus catostomus</i> )		0.4 $\pm$ 0.2		1.0 $\pm$ 0.3	0.1
Exp 15	Northern Pike ( <i>Esox lucius</i> )		0.1	1.0 $\pm$ 0.1	2.1 $\pm$ 0.8	
	Lake Whitefish ( <i>Coregonus clupeaformis</i> )				1.0 $\pm$ 0.4	
Exp 16	White Sucker ( <i>Catostomus commersoni</i> )	4.0	0.7 $\pm$ 0.3	0.6 $\pm$ 0.2	1.3 $\pm$ 0.5	2.9 $\pm$ 1.1
Exp 17	Northern Pike ( <i>Esox lucius</i> )		0.2		1.0 $\pm$ 0.4	0.5 $\pm$ 0.4
	White Sucker ( <i>Catostomus commersoni</i> )				1.1 $\pm$ 0.3	0.1 $\pm$ 0.2
Exp 19	Northern Pike ( <i>Esox lucius</i> )	9.0	36.8 $\pm$ 12.3		67.7 $\pm$ 23.7	7.9 $\pm$ 3.2
	White Sucker ( <i>Catostomus commersoni</i> )		24.1 $\pm$ 5.5		13.1 $\pm$ 3.6	4.7 $\pm$ 1.3
	Arithmetic mean	<b>3.8</b>	<b>6.5</b>	<b>1.1</b>	<b>16.0</b>	<b>5.7</b>

<sup>a</sup> Data used to verify activity concentration predictions in Lake Whitefish during exercise I.

**Table 5**

Geometry and mass of aquatic animals under consideration during Exercise II.

Animal (species)	Geometry (cm) Length x height x width	Mass (g fm)	Occupancy in water column/in sediment (%)
Pelagic fish (e.g., Northern Pike & Lake Trout) <sup>a</sup>	50 $\times$ 15 $\times$ 10	1200	75/25
Benthic Fish - Large (White Sucker & Lake Whitefish) <sup>a</sup>	45 $\times$ 15 $\times$ 10	1191	30/70
Benthic Invertebrates (Fingernail Clams) <sup>b</sup>	2.5 $\times$ 1.5 $\times$ 1	1.6	0/100

<sup>a</sup> Data on fish size taken from industry environmental monitoring reports.

<sup>b</sup> From Kilgour and Mackie (1991) and Funk and Reckendorfer (2008).

fm; the highest value was selected in the R&D 128 applications.

Looking at the values of CRs for fish (Fig. 2; upper panel), <sup>210</sup>Pb CRs ranged from 300 to 10,900 L kg<sup>-1</sup> fm, from 50 to 7520 L kg<sup>-1</sup> fm for <sup>210</sup>Po, and from 44 to 3200 L kg<sup>-1</sup> fm for <sup>226</sup>Ra. For <sup>230</sup>Th, default CR values ranged from 70 to 200 L kg<sup>-1</sup> fm. Finally, for <sup>238</sup>U, CRs varied from 4 to 1000 L kg<sup>-1</sup> fm. RESRAD-BIOTA (ANSTO) used a value of 1000 L kg<sup>-1</sup> fm (i.e. default value), which was considerably higher than values used by all other modellers which were all  $\leq$ 30 L·kg<sup>-1</sup> fm. Default values are considered conservative and appropriate for screening assessments at the project planning stage while site-specific values should be chosen for more detailed assessments (e.g. Tier II and III) typical of assessment during operating and site closure phases.

### 3.3. Predicted activity concentrations in fish

Predicted activity concentrations for radionuclides in fish were within an order of magnitude for <sup>230</sup>Th and all were within two orders of magnitude of the measured data (Fig. 3). There was comparatively little variation between the predictions of the various models because of the relatively similar water concentrations (1–110 mBq L<sup>-1</sup>; Table 2) and CRs (70–200 L kg<sup>-1</sup> fm; Table 3 and Fig. 2 upper panel) used by

modellers. Modellers under-predicted <sup>230</sup>Th activity concentrations when the measured whole organism body activity concentrations were low, and over-predicted activity concentrations when measured activity concentrations were high (Fig. 3).

For <sup>226</sup>Ra, RESRAD-BIOTA (ANSTO) predicted high activity concentrations because the RESRAD-BIOTA default CR value (3200 L kg<sup>-1</sup> fm) was purposefully applied to demonstrate the effects of using this model as a simple screening-level assessment. The RESRAD-BIOTA default CR is for a generic “aquatic animal” and according to the RESRAD-BIOTA support documentation (USDOE 2004), it was derived from empirical data on freshwater *Gammarus* sp. (amphipod). The default RESRAD-BIOTA CR values purport to be maximum values for the generic organism types and are for application in conservative screening assessments typical of the planning stage of uranium mining and milling projects. Hence as expected, the use of the default CRs predicted high fish whole organism body concentrations. Other models used lower CRs and hence predicted lower <sup>226</sup>Ra accumulation in Lake Whitefish whole organism body, with predicted activity concentrations being close to measured values.

For <sup>238</sup>U, modellers tended to under-predict fish whole-body activity concentrations at the less affected sites and over-predict when <sup>238</sup>U was measured to be above 10 Bq·kg<sup>-1</sup> fresh mass (fm) in fish. This can be explained by over-estimating uranium transfer at exposed sites. Uranium bioaccumulation can often be over-estimated since its bioavailability has been reported to be affected by pH, dissolved organic carbon, hardness and alkalinity (Sheppard et al., 2005; Goulet et al., 2011). The purposeful use of default values (RESRAD-BIOTA (ANSTO)), again predicted high accumulation because of the conservative RESRAD-BIOTA default CR of 1000 L kg<sup>-1</sup> fm, compared to values ranging from 4 to 30 L kg<sup>-1</sup> fm in other models.

Most modellers over-predicted <sup>210</sup>Po accumulation in fish whole organism body. The ERICA Tool (DSA) predicted high <sup>210</sup>Po in fish whole organism body because they used a CR value of 7520 L kg<sup>-1</sup> fm as reported by Gjelsvik and Brown (2009). The CRs of most other modellers ranged from 50 to 500 L kg<sup>-1</sup> fm. The ERICA Tool (CEH) and RESRAD-BIOTA (ANSTO) predicted high <sup>210</sup>Po accumulation as they used CR values of 4250 and 500 L kg<sup>-1</sup> fm, respectively. The R&D 128 results consistently predicted lower accumulation as their selected CR

**Table 6**

Models and participating organizations, decay chain equilibrium assumptions used when radionuclides concentrations were not available in water and/or sediment, weighting factors used for dose rates calculations in exercise II.

Model (organization)	decay chain equilibrium assumption	Weighting factors	References
ERICA (SCK)	$^{210}\text{Pb}$ ; $^{210}\text{Bi}$ $^{210}\text{Po}$ : x $^{226}\text{Ra}$ ; $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ $^{230}\text{Th}$ :x $^{238}\text{U}$ , $^{234}\text{U}$	Internal alpha: 10 Internal beta gamma: 1 Internal low beta: 3	Brown et al. (2008)
K-BIOTA (KAERI)	$^{210}\text{Pb}$ ; $^{210}\text{Bi}$ $^{210}\text{Po}$ : x $^{226}\text{Ra}$ ; $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ , $^{218}\text{At}$ $^{230}\text{Th}$ : x $^{238}\text{U}$ , $^{234}\text{U}$	Internal alpha: 10 Internal beta gamma: 1 Internal low beta: 3	Keum et al. (2011)
Hokus (SUJB)	$^{210}\text{Pb}$ : x $^{210}\text{Po}$ : x $^{226}\text{Ra}$ : x $^{230}\text{Th}$ : x $^{238}\text{U}$ : x	Internal alpha: 10 Internal beta gamma: 1 Internal low beta: 1	
RESRAD-BIOTA (ANL)	$^{210}\text{Pb}$ ; $^{210}\text{Bi}$ $^{210}\text{Po}$ : x $^{226}\text{Ra}$ ; $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ , $^{218}\text{At}$ , $^{210}\text{Tl}$ $^{230}\text{Th}$ : x $^{238}\text{U}$ ; $^{234}\text{Th}$ , $^{234}\text{Pa}$ , $^{234\text{m}}\text{Pa}$	Internal alpha: 10 Internal beta gamma: 1 Internal low beta: 1	USDOE (2004)
ERICA (CEH)	$^{210}\text{Pb}$ ; $^{210}\text{Bi}$ $^{210}\text{Po}$ : x $^{226}\text{Ra}$ ; $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ $^{230}\text{Th}$ : x $^{238}\text{U}$ ; $^{234}\text{Th}$ , $^{234}\text{Pa}$ , $^{234\text{m}}\text{Pa}$	Internal alpha: 10 Internal beta gamma: 1 Internal low beta: 3	Brown et al. (2008)
EDEN (IRSN)	$^{210}\text{Pb}$ ; $^{210}\text{Bi}$ $^{210}\text{Po}$ : x $^{226}\text{Ra}$ ; $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ $^{230}\text{Th}$ :x $^{238}\text{U}$ , $^{234}\text{U}$	Alpha: 10 Gamma: 1 Internal low beta: 3	Beaugelin-Seiller et al. (2006)
ERICA (JSI)	$^{210}\text{Pb}$ ; $^{210}\text{Bi}$ $^{210}\text{Po}$ : x $^{226}\text{Ra}$ ; $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ $^{230}\text{Th}$ : x $^{238}\text{U}$ : x	Internal alpha: 10 Internal beta gamma: 1 Internal low beta: 3	Brown et al. (2008)
ERICA (EA)	$^{210}\text{Pb}$ ; $^{210}\text{Bi}$ $^{210}\text{Po}$ : x $^{226}\text{Ra}$ ; $^{222}\text{Rn}$ , $^{218}\text{Po}$ , $^{214}\text{Pb}$ , $^{214}\text{Bi}$ , $^{214}\text{Po}$ $^{230}\text{Th}$ : x $^{238}\text{U}$ ; $^{234}\text{Th}$ , $^{234}\text{U}$	Internal alpha: 10 Internal beta gamma: 1 Internal low beta: 3	Brown et al. (2008)

x – no decay product considered for this radionuclide.

value was  $50 \text{ L kg}^{-1} \text{ fm}$  (Table 3). Modellers using CRs of about  $170\text{--}240 \text{ L kg}^{-1} \text{ fm}$  predicted within an order of magnitude the measured accumulation of  $^{210}\text{Po}$  in Lake Whitefish.

For  $^{210}\text{Pb}$ , most modellers over-predicted accumulation in Lake Whitefish, with some predictions by R&D 128 being three orders of magnitude above the measured data. The R&D 128 model does not have  $^{210}\text{Pb}$  in the default radionuclide list. Therefore, following the recommended approach associated with the R&D 128 methodology (Coppstone et al., 2003), the CR for  $^{137}\text{Cs}$  (of  $10,900 \text{ L kg}^{-1} \text{ fm}$ ) was used. In general, a CR value around  $300 \text{ L kg}^{-1} \text{ fm}$  yielded  $^{210}\text{Pb}$  activity concentrations within one to two orders of magnitude.

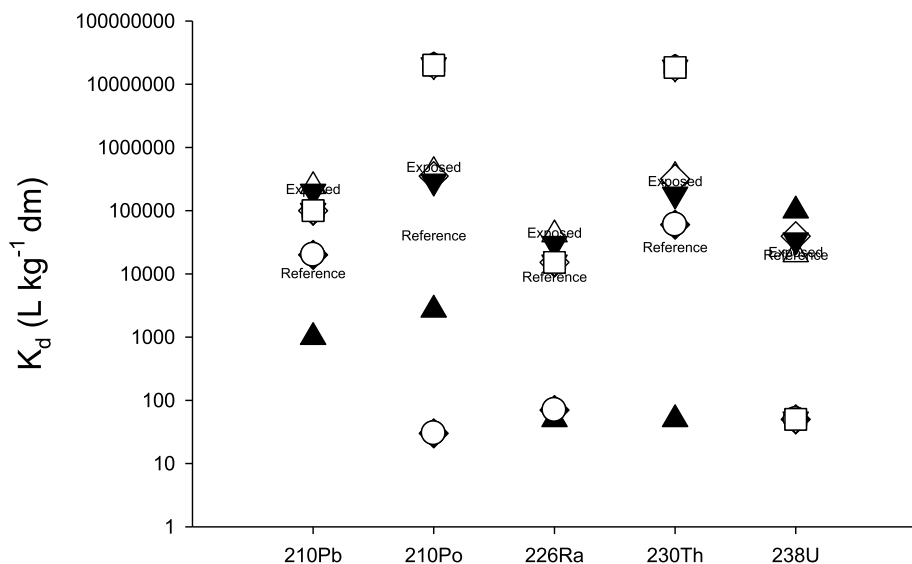
### 3.4. Predicted activity concentrations in benthic invertebrates

In general, predictions for activity concentrations in Fingernail Clams, as representative of the benthic invertebrate community, varied between modellers by over 2–3 orders of magnitude depending on the radionuclide (Fig. 4). The wide variation was due by one modeller (RESRAD-BIOTA UoS) that predicted high accumulation of radionuclides (closed diamond symbols in Fig. 4). This was the result of purposely using sediment concentrations and partition coefficients to obtain water concentrations, and then using default CRs in RESRAD-BIOTA. Note that such an approach is not recommended if water activity concentrations are available (as they were in this case) (e.g., Brown et al., 2008). Hence, we will not discuss the RESRAD-BIOTA UoS results further.

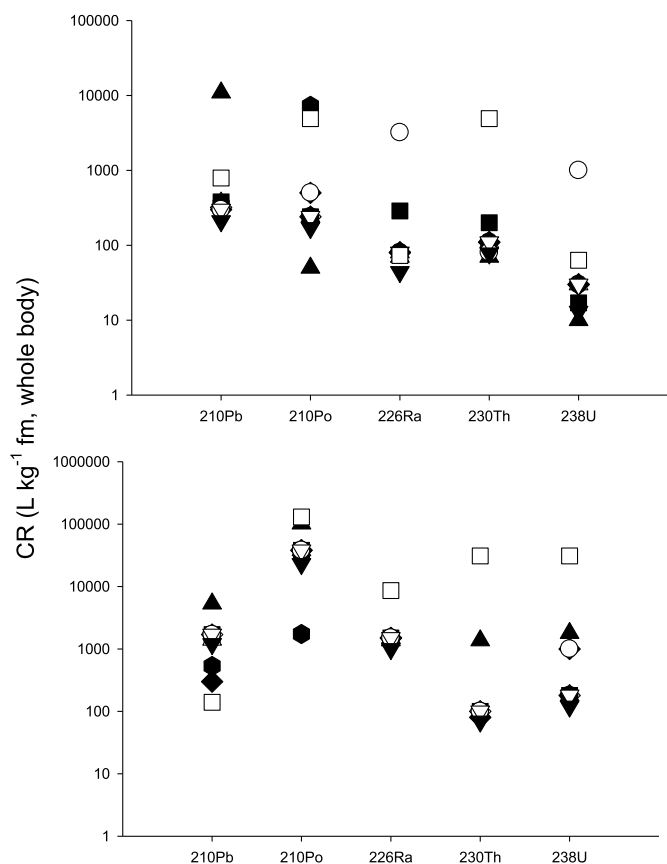
Predictions for  $^{230}\text{Th}$  were similar for most modellers because the CRs used by all modellers were within one order of magnitude (Table 2 and Fig. 2 upper panel). In contrast, predicted levels of  $^{210}\text{Po}$  in Fingernail Clams varied by 2–3 orders of magnitude because of variability in the choice of CRs (Table 2 and Fig. 2 upper panel). No site-specific measured data in benthic invertebrates were available to judge the conservativeness of both the  $^{230}\text{Th}$  and  $^{210}\text{Po}$  predictions in Fingernail Clams.

In reference site 1 near Uranium city, Saskatchewan, modelling predictions of  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  activity concentrations in Fingernail Clams ranged from 10 to 300, 1–10 and 1–10  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$ , respectively. While no site-specific data was available to compare the accumulation of  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$ , there were activity concentrations data in aquatic insects collected by Swanson (1982). While these are different species, the data of Swanson (1982) is used as comparison simply to gauge if the predictions were at least within accumulations seen in other invertebrate species. Swanson (1982) reported that levels of  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  in invertebrate species ranged from 4 to 396  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$ , 1 to 26  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$  and 3 to 58  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$ , respectively in Fredette Lake, an unaffected lake also near Uranium City. This comparison indicates that modellers predicted radionuclides in the same order of magnitude.

In exposed areas, the predicted activity concentrations of  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  in Fingernail Clams ranged from 10 to 1000  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$ , 1 to 1300  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$ , 1 to 10,000  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$ , respectively. Swanson (1982) reported levels of  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  for a wide range of insect larvae, including dipterans (black flies), tricopterans (caddis flies), odonates (dragon and damselflies), red worms (midges) and notonectids (backswimmer) in high exposure areas near Uranium City, Saskatchewan. These measured activity concentrations of  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  ranged from 13 to 1265  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$ , from 2 to 1323  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$  and from 38 to 610  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$ , respectively for organisms collected downstream of lakes partially infilled with tailings. More recently, Wiramanaden et al. (2015) reported  $^{226}\text{Ra}$  activity concentrations in caged oligochaetes ranging from 140 to 2970  $\text{Bq}\cdot\text{kg}^{-1} \text{ dm}$  (circa 35  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$  to 740  $\text{Bq}\cdot\text{kg}^{-1} \text{ fm}$ ; estimated using dry to fresh mass conversion factor presented in IAEA (2014)). Hence, considering that levels of  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  reported by Swanson (1982) and Wiramanaden et al. (2015) were measured downstream of tailings lakes, we suggest that predicted concentrations for Fingernail Clams were broadly similar to the literature data. Whether these predictions are realistic remains to be confirmed as different species accumulates at different rates (IAEA (2014)) and the oligochaetes accumulation data from Wiramanaden et al. (2015) were obtained from 10 days exposure duration and so it is possible that accumulation did not reach equilibrium with sediment and water. In contrast,  $^{238}\text{U}$  was generally over-predicted likely because uranium transfer were over-estimated at exposed sites. As indicated before, Uranium bioaccumulation can often be over-estimated since its bioavailability has been reported to be affected by pH, dissolved organic carbon, hardness and alkalinity (Sheppard et al., 2005; Goulet et al., 2011).



**Fig. 1.** Range of distribution coefficients used for radionuclides monitored near U mines and mills in Canada by the different participants. (●) RESRAD-BIOTA (ANL), (○) RESRAD-BIOTA (ANSTO), (■) K-BIOTA (KAERI), (□) ERICA (UK EA), (▲) R&D 128, (△) ERICA (SCK CEN), (▼) Hokus, (▽) ERICA (ANSTO), (◆) RESRAD-BIOTA (U of S), (◇) ERICA (UK CEH) and (●) ERICA (DAS). Mean distribution coefficients at all exposed sites and at all reference site also shown as “exposed” and “reference”, respectively.



**Fig. 2.** Range of concentration ratios between water and Fingernail Clam (lower panel) and fish (upper panel). The following symbols indicate the model: (●) RESRAD-BIOTA (ANL), (○) RESRAD-BIOTA (ANSTO), (■) K-BIOTA (KAERI), (□) ERICA (UK EA), (▲) R&D 128, (△) ERICA (SCK CEN), (▼) Hokus, (▽) ERICA (ANSTO), (◆) RESRAD-BIOTA (U of S), (◇) ERICA (UK CEH), and (●) ERICA (DAS).

### 3.5. Weighted absorbed dose rate to pelagic fish

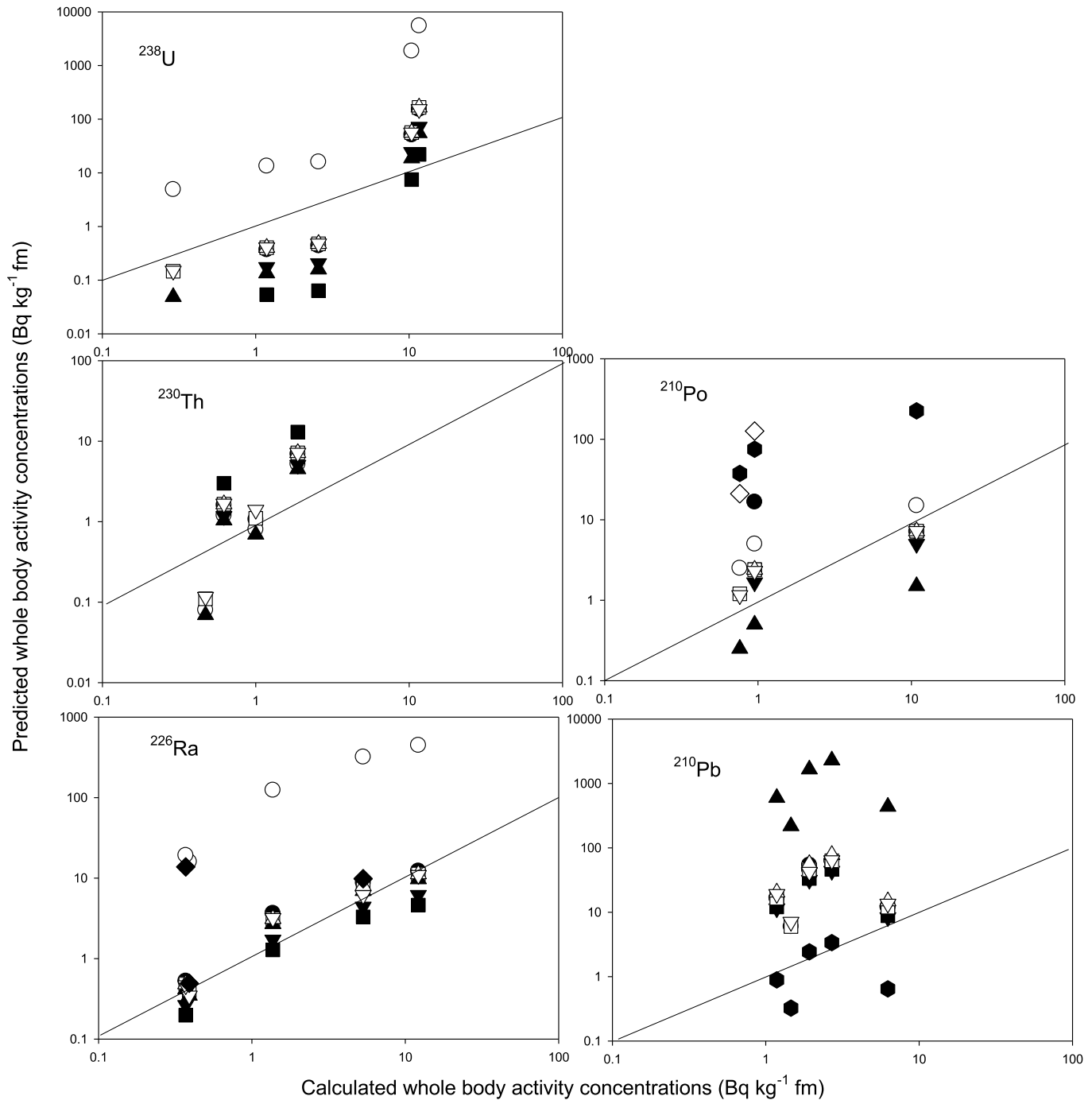
Modellers used calculated whole organism body activity concentrations when available (Table 4). At the reference sites 3 and 4 as well as several exposure sites, the predicted dose rates to pelagic fish ranged

from 0.001 to 100  $\mu\text{Gy h}^{-1}$  (Fig. 5). Variation in predicted absorbed dose rates was often high for a given site. Often, this wide variation was the result of some modellers only considering measured radionuclides and others estimating all radionuclides from the  $^{238}\text{U}$  decay chain assuming secular equilibrium when daughter radionuclides were not measured. In particular, the wide variability in dose predictions can be explained by modellers considering  $^{210}\text{Po}$  and  $^{230}\text{Th}$  along with  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  while other modellers only considered  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  because these were the only available measurements in fish tissue, sediment or water. Omitting to consider  $^{210}\text{Po}$  and  $^{230}\text{Th}$  and not assuming secular equilibrium with all parent radionuclides (i.e.  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$ ,  $^{226}\text{Ra}$ ,  $^{230}\text{Th}$  and  $^{238}\text{U}$ ) led some modellers to estimate comparatively low dose rates at some sites. Despite the variability mentioned above, the calculated total absorbed dose rates were within two orders of magnitude at several exposed sites. This relatively narrow range can be explained by using calculated whole organism body radionuclide activity concentrations using measured in fish muscle and bones (Table 4) and by using whole organism body radionuclide CRs from similar sites provided in Table 3 with water activity concentrations for radionuclides when radionuclide activity concentrations were not measured in fish tissue.

### 3.6. Weighted absorbed dose rate to benthic fish

Benthic fish are exposed to radionuclides in sediments because they live near the river/lake bottoms where they typically forage for food. Hence, their radionuclide exposure will differ from pelagic fish. Modellers predicted the absorbed dose rates to benthic fish and used activity concentrations in whole organism body (Table 4) of benthic fish when available because at this stage a realistic (as possible) assessment was required. The predicted dose rates to benthic fish were the same order as those for pelagic fish ranging from 0.006 up to just below 400  $\mu\text{Gy h}^{-1}$  (Fig. 6).

At exposed sites, the predicted dose rates were generally higher in benthic fish (Fig. 6) than in pelagic fish (Fig. 5) because benthic fish are more exposed to radionuclide activity concentrations in sediments than pelagic fish. Chosen input parameters presented in Table 5 indicates that pelagic fish are exposed to sediment 25% of the time while benthic fish are exposed to sediment 70% of the time as they forage for food. Wide variations in dose predictions happened because some modellers considered  $^{210}\text{Po}$  and  $^{230}\text{Th}$  along with  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  while others only considered  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  because these were the only available measurements in fish flesh and bones, sediment or water and hence predicted lower absorbed dose rates.

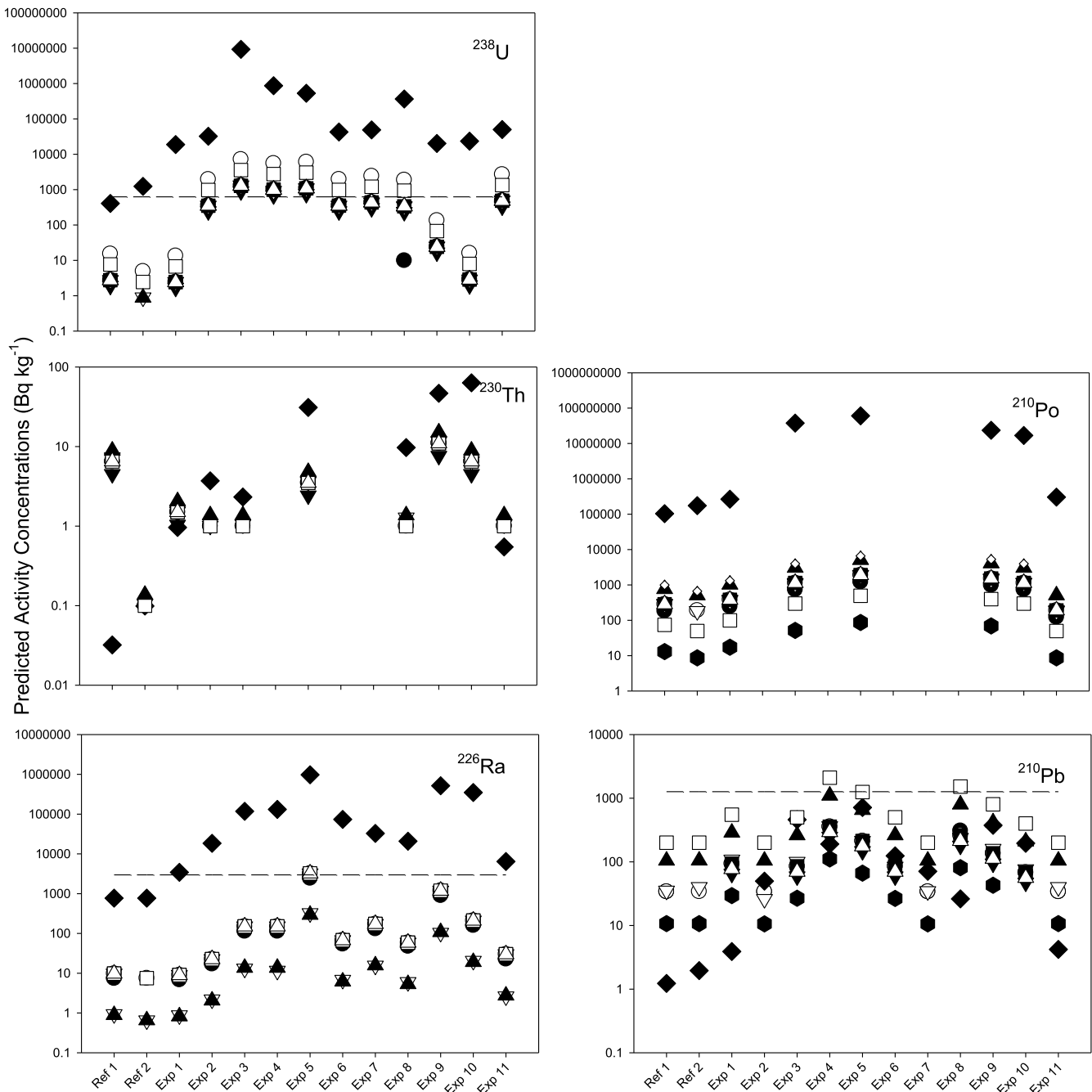


**Fig. 3.** Predicted accumulation of radionuclides in comparison to calculated accumulation based on measured radionuclide activity concentrations in flesh and bones of Lake Whitefish (Data from Table 4). Results from (●) RESRAD-BIOTA (ANL), (○) RESRAD-BIOTA (ANSTO), (■) K-BIOTA, (□) ERICA (UK EA), (▲) R&D 128, (△) ERICA (SCK CEN), (▼) Hokus (SUJB), (▽) ERICA (ANSTO), (◇) ERICA (UK CEH), (●) ERICA (DAS). The line indicates a 1:1 line.

### 3.7. Weighted absorbed dose rate to benthic invertebrates

Benthic invertebrates are exposed to sediment and surface water. At reference site 3, the predicted dose rates ranged from 0.01 to 10  $\mu\text{Gy h}^{-1}$  (Fig. 7). This variation is largely explained by the ERICA Tool (SCK CEN) assuming secular equilibrium of radionuclides with <sup>238</sup>U, the only measured value in water at that site. Other modellers only considered <sup>238</sup>U activity concentration in water. At reference site 4, the predicted dose rates ranged from 0.1 to 30  $\mu\text{Gy h}^{-1}$ . No water radionuclide activity

concentration data were provided to modellers so the sediment concentrations with a partition coefficient were used in the calculation of predicted dose, which explains the higher dose rate predictions compared to reference site 3. At exposed sites, the predicted dose rates ranged from 0.1 to 5000  $\mu\text{Gy h}^{-1}$  (Fig. 7). Variability was generally within two orders of magnitude within a site. Hokus predicted low dose rates because of only considering measured radionuclides; decay daughters in secular equilibrium with <sup>238</sup>U were not included in the dose rate calculation. Aside from this prediction, dose rate predictions were



**Fig. 4.** Predicted activity concentrations in Fingernail Clam at reference and exposure sites. Results from (●) RESRAD-BIOTA (ANL), (○) RESRAD-BIOTA (ANSTO), (■) K-BIOTA (KAERI), (□) ERICA (UK EA), (▲) R&D 128, (△) ERICA (SCK CEN), (▼) Hokus, (▽) ERICA (ANSTO), (◆) RESRAD-BIOTA (U of S), (◇) ERICA (UK CEH), and (●) ERICA (DAS). Dash line in <sup>226</sup>Ra panel is the highest activity levels measured in caged oligochaetes by Wiramanaden et al. (2015). Dash lines in the <sup>238</sup>U and <sup>210</sup>Pb panels are the highest activity levels measured in Beaverlodge tailings lakes by Swanson (1982).

generally within  $1 \mu\text{Gy h}^{-1}$  and  $1000 \mu\text{Gy h}^{-1}$  because modellers applied their respective default model CR values or CRs from the IAEA handbook of transfer parameters (IAEA 2014). The ERICA Tool (SCK CEN) also included <sup>234</sup>U and <sup>234</sup>Th in dose calculations; dose rates calculated did not differ much from other dose rate calculations.

### 3.8. Recommended dose assessment approach

The results obtained in modelling comparisons demonstrated that participants obtained variable predictions because the models had different default parameters and because modellers were making different decisions than other fellow modellers. The variability in results could be explained by the use of different partition coefficients and concentration ratios; different approaches to modelling when data were

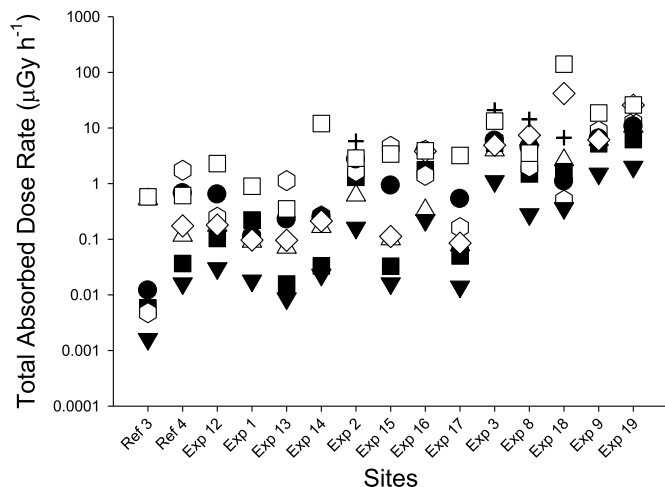


Fig. 5. Predicted total absorbed dose rates to pelagic fish at exposed and reference sites. Results from (●) RESRAD-BIOTA (ANL), (Δ) ERICA (SCK CEN), (■) K-Biota (KAERI), (□) ERICA (UK EA), (▼) HOKUS (○), ERICA (JSI), (◇) ERICA (UK CEH), (+) EDEN.

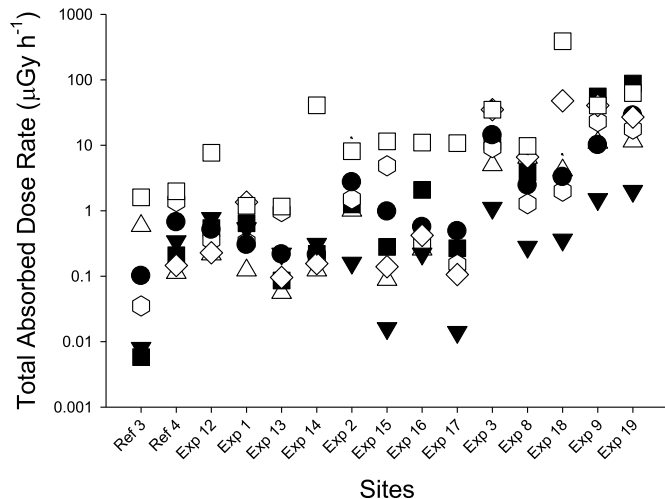


Fig. 6. Predicted total absorbed dose rates to benthic fish at exposed and reference sites. Results from (●) RESRAD-BIOTA (ANL), (Δ) ERICA (SCK CEN), (▼) HOKUS, (■) K-BIOTA (KAERI), (□) ERICA (UK EA), (◇) ERICA (UK CEH), (+) EDEN (○), ERICA (JSI).

not available for all media (e.g., only sediment data available but not water) and not considering all decay daughters of the radionuclide decay chain in dose rate calculations. Consequently, we recommended a standardized best-practice approach to be applied to different stages of mining and milling of radiological environmental impact assessment as the final exercise of the project, which would take into account the data available and the source of contamination at these project sites.

The proposed standardized best-practice approach was.

●Step 1: Identify radionuclides of concern.

- o At the project planning stage, include all radionuclides from the <sup>238</sup>U decay chain in a screening dose rate assessment [the user needs to understand the assumptions used with regards to daughter radionuclides in the estimation of individual model’s dose conversion coefficients (see Vives i [Battle et al. 2007; 2011](#))].
- o At the operating and closure stages, include all radionuclides from the <sup>238</sup>U decay chain and assumption of secular equilibrium should be verified based on monitoring information.

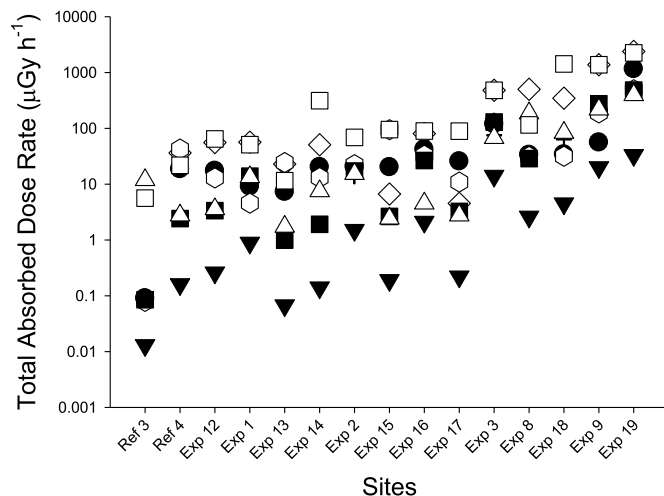


Fig. 7. Predicted total absorbed dose rates to Fingernail Clam at exposed and reference sites. Results from (●) RESRAD-BIOTA (ANL), (Δ) ERICA (SCK CEN), (▼) HOKUS, (■) K-BIOTA (KAERI), (□) ERICA (UK EA), (◇) ERICA (UK CEH), (+) EDEN (○), ERICA (JSI).

- o <sup>235</sup>U and decay daughters such as <sup>231</sup>Pa, <sup>227</sup>Ac, <sup>227</sup>Th, and <sup>223</sup>Ra may be important ([Beaugelin-Seiller et al., 2016](#)) and may be considered in the dose rate assessment at sites that have released radionuclides for decades

●Step 2: Select aquatic organisms.

- o At the planning stage, identify species requiring dose rate assessments. Not all species need to be represented individually, but should be covered by using a set of broad organism groups. The ICRP recommends a series of reference (i.e. representative species) animal and plants to include in dose assessments ([ICRP, 2008](#)). Selected species should consider stakeholder input and should include multiple trophic levels.
- o During operations and site closure, species of concern may change as a result of observed effects to specific species investigated under an environmental effects monitoring program for a given site.

●Step 3: Calculate or measure activity concentrations in water.

- o At the project planning stage (i.e., new facility), activity concentrations of radionuclides in water prior to construction and operation should be measured to estimate a conservative (i.e. 95th percentile value ([Kilgour et al., 1998; Brown et al., 2016](#))) baseline.
- o Then, predict conservative radionuclide activity concentrations in water by adding conservative estimates of facility discharges to the conservative baseline water activity concentrations.
- o During operation and site closure, radionuclide activity concentrations should be measured in water and 95th percentile and mean values used in more realistic dose rate assessments. Measurements can also be used to verify predictions at the planning stage.

●Step 4: Calculate or measure radionuclide activity concentrations in sediments.

- o At the project planning stage, activity concentrations of radionuclides in sediment prior to construction and operation should be measured to estimate a conservative baseline.
- o Then, predict conservative activity concentrations of radionuclides in sediments by adding conservative estimates of facility discharges to the conservative baseline sediment activity concentrations.

- o During operations and site closure, radionuclide activity concentrations should be measured in sediments and 95th percentile and mean values used in more realistic dose rate assessments. Measurements can also be used to verify predictions at the planning stage.

**•Step 5: Calculate or measure radionuclide activity concentrations in the whole organism body.**

- o At the project planning stage, baseline fish tissue and whole organism body of benthic invertebrate activity concentrations should be measured to estimate a conservative baseline, when possible.
- o Then, modellers predict conservative activity concentrations by multiplying the selected conservative CR with the conservative water activity concentration estimated at step 3 and added to baseline activity concentrations when possible.
- o When selecting conservative CR values, modellers should consider the following:
  - consult the compilations such as [IAEA \(2014\)](#) or preferably its underlying database which is periodically updated with new data ([Coppstone et al., 2013](#); [Brown et al., 2016](#); <http://www.wildlifetransferdatabase.org/>)
  - select CRs for organisms as similar to the species assessed as possible, and;
  - select CRs from other similar operating sites, and;
  - Select CRs by considering water quality downstream of the proposed uranium mining or milling site.
- o During operations and site closure, radionuclide activity concentrations should be measured in benthic invertebrate whole organism body and fish tissue. Fish whole organism body activity concentrations can be calculated from activity concentration in bones and flesh using Equation (3) for each fish species (or relationships between tissue and whole organism activity concentrations such as presented by [Yankovich et al., 2010a](#)). 95th percentile and mean values should be used in more realistic dose rate assessments. Measurements can also be used to verify predictions at the planning stage.

**•Step 6: Calculate total absorbed dose rates.** Use an appropriate model (e.g., [Brown et al. \(2016\)](#), [Beaugelin-Seiller et al. \(2006\)](#), [USDOE \(2019\)](#) which are free of charge, freely available, which have gone through various IAEA evaluations to calculate total absorbed dose rates. At the project planning phase, follow the proposed steps:

- o Step 6a: Calculate internal dose rates using equation 4 and the conservative radionuclide activity concentrations calculated at step 5 along with the appropriate dose conversion factor. If appropriate, use a default geometry or calculate your own dose conversion coefficients by specifying a new organism geometry and exposure scenario. The different dosimetry methods used by the available models add little to the overall uncertainty in estimated dose ([Vives i Batlle et al., 2007, 2011](#)).
- o Step 6b: Then, calculate external dose rates with equation 5 by using conservative water (Step 3) and sediment activity concentrations (Step 4) and by selecting an appropriate occupancy factor.
- o Step 6c: Select occupancy factor. Pelagic and benthic fish species will have different occupancy factors. These should be based on site-specific data or based on literature review and then used to calculate external dose rates from sediment and water. Similarly, appropriate occupancy for any other organisms will need selecting.
- o Step 6d: Calculate total absorbed dose rates.
- o Step 6e: Select a radiological weighting factor. The [ICRP \(2021\)](#) propose an RBE weighted absorbed dose of 1 for all low-LET radiations and 10 for alpha particles. Use of a single value of 1 for all low-LET radiations is consistent with the approach taken to protection of humans.
- o Step 6f: During operations and site closure, use measured water, sediment and whole organisms body activity concentrations to

calculate internal, external and total absorbed dose rates. These updated dose rate predictions can also be used to verify predictions at the planning stage.

Two modellers using the ERICA Tool (SCK CEN) and ERICA Tool (UK EA) and one modeller using K-BIOTA implemented this recommended approach starting at Step 2. We tested the approach using two references (Reference sites 2 & 4) and six exposed sites (i.e. sites 8, 9, 15, 16, 18, 19). Results of dose to pelagic fish, benthic fish, and benthic invertebrates are depicted in [Fig. 8](#).

Dose rates to pelagic fish at the two reference sites were as expected and predicted to be well below the generic screening benchmark of  $10 \mu\text{Gy h}^{-1}$  ([Garnier-Laplace et al., 2010](#)), as used in the ERICA Tool. Variation in absorbed dose rate predictions was lower when modellers used the best approach at the reference and most exposed sites ([Table 7](#)). For instance, standard deviation in predicted dose rates to pelagic fish by K-BIOTA, the ERICA Tool (SCK CEN) and ERICA Tool (UK EA) was lower at reference site 4, exposure 9, 18 and 19 when using the standardized best-practice than when not using it.

However, appreciable variations in predictions were noted at exposed sites 8 and 16. The variation was even higher than when not using the best-practice approach. At exposed site 8, since there were no measured whole organism body concentrations for  $^{210}\text{Po}$  and  $^{230}\text{Th}$ , K-BIOTA used CRs instead of measured values at other sites, which yielded higher dose rates compared to others who used measured concentration in fish which were caught in exposed site 19. Using measured radionuclide activity concentrations in a given fish species from another site to calculate dose rates to the same species at a given sites is recommended over the use of literature CRs unless the water quality parameters that modify radionuclide bioavailability (i.e. Calcium, dissolved organic carbon, pH, phosphorus, etc...) at both sites are strikingly different. In the case of different water quality, choosing a CRs for a similar species in comparable water quality would be justified. This again confirms that the choice of CRs is important in dose predictions. Finally, at exposed site 16, K-BIOTA again used CRs to predict activity concentrations in fish, while the ERICA Tool (EA UK) and the ERICA Tool (SCK CEN) used the same measured activity concentrations in White sucker ([Table 4](#)). The dose rate was higher in the ERICA Tool (EA UK) in comparison to the ERICA Tool (SCK CEN) because of higher dose contribution from  $^{210}\text{Po}$  and  $^{238}\text{U}$ . This difference is explained by one modeller calculating external dose from  $^{210}\text{Po}$  in water using secular equilibrium assumptions while the other modeller used the mean  $^{210}\text{Po}$  concentrations at other exposed sites. Using mean  $^{210}\text{Po}$  concentrations at other exposed sites is recommended over assuming secular equilibrium as milling of uranium targets the removal of  $^{238}\text{U}$  from the ore, likely leaving the remaining daughters in the tailings and effluent stream.

Dose rates to benthic fish species were, as expected, predicted to be higher than to pelagic fish, considering their additional exposure to sediments from radionuclide accumulation over time ([Fig. 8](#); middle panel). The predicted dose rates to benthic fish at the two reference areas were well below the screening benchmark of  $10 \mu\text{Gy h}^{-1}$ . The best practice approach reduced the variability in predictions of absorbed dose rates to benthic fish ([Table 7](#)). For instance, the standard deviation of the predicted dose rates to benthic fish by K-BIOTA and the ERICA Tool (SCK CEN) and ERICA Tool (UK EA) at all sites was lower when using the best-practice approach than when modellers did not use the best-practice approach.

The predicted dose rates to benthic invertebrates (i.e. Fingernail Clams) were higher than for benthic and pelagic fish ([Fig. 8](#); lower panel). This is because CRs for benthic invertebrates are generally higher than for fish ([IAEA 2014](#)). The predicted dose rates to Fingernail Clams at the two reference areas were even higher than the screening benchmark of  $10 \mu\text{Gy h}^{-1}$ . The ERICA Tool (EA UK) tended to predict lower dose rates due to the use of CRs from [IAEA \(2014\)](#), whereas model default CR values were used by the ERICA Tool (SCK CEN) and K-BIOTA.

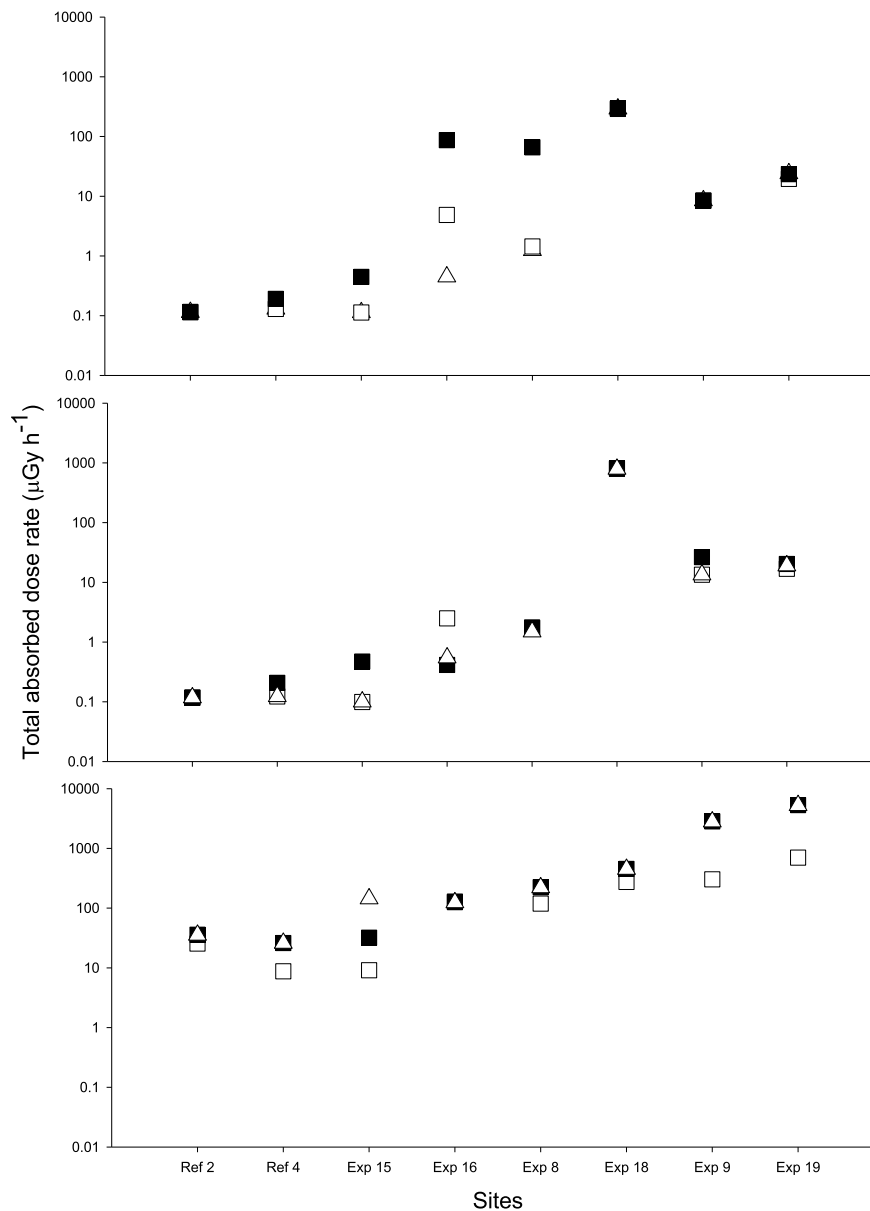


Fig. 8. Predicted total absorbed dose rates to pelagic fish (upper panel), benthic fish (middle panel) and Fingernail Clam (bottom panel) at reference and exposed sites by (Δ) ERICA (SCK CEN), (□) ERICA (UK EA) and (■) K-BIOTA.

For Fingernail Clams, the best practice approach did reduce the variability in predicted absorbed dose rates only at four sites out of the seven selected sites (Table 7). The main reason is the lack of whole organism body concentration of radionuclides in Fingernail Clams and other species of benthic invertebrates because of their small size. Measurements of radionuclide activity concentrations in whole organism body of benthic invertebrate would reduce the uncertainty in dose predictions.

Overall, our recommended best-practice approach to predict dose rates reduced variations in dose rate predictions to pelagic and benthic fish but this was not as convincing for benthic invertebrates. The variability could be reduced even further if sufficient baseline information on background radionuclide activity concentrations in water, sediment and freshwater organisms (fish and benthic invertebrates) was collected. Adequate baseline information would then allow for conservative incremental dose predictions at the project proposal stage by selection of site-specific CRs using measured radionuclide activity concentrations in organisms during operations and decommissioning of other similar

projects. Accumulation of CR data during operation and closure of uranium mining projects in databases such as the IAEA (2014) (Coplestone et al., 2013; Brown et al., 2016; <http://www.wildlifetransferdatabase.org/>) will likely reduce uncertainties in radionuclide accumulation in freshwater organisms at the planning stage of other similar projects.

### 3.8.1. Remaining uncertainties

The results of this comparison study demonstrated that using different modelling approaches can lead to considerable variation in predicted dose rates to freshwater organisms if measured activity concentrations in organisms are not available. This variability can be reduced simply as a result of the life-cycle of a project (i.e. planning stage vs operations where environmental monitoring data can be collected to verify predictions made at the planning stage) and by following the approach outlined above which uses predictions more closely resembling measured activity concentrations (where available),

**Table 7**

Comparison of variability (i.e. mean  $\pm$  standard deviation) in absorbed dose rate predictions when using or not using the standardized best-practice approach. Red color indicates when the variability in absorbed dose rate predictions by ERICA (SCK CEN), ERICA (UK EA) and K-BIOTA modellers was reduced with the best-practice approach compared to not using the best-practice approach.

Modelling approach	R4	E15	E16	E8	E19	E9	E18
Pelagic fish							
Not using best practice	0.25 $\pm$ 0.30	1.17 $\pm$ 1.93	1.98 $\pm$ 1.80	3.70 $\pm$ 2.31	14.33 $\pm$ 10.41	8.43 $\pm$ 9.38	71 $\pm$ 97
Using best-practice	0.15 $\pm$ 0.03	0.22 $\pm$ 0.20	30.78 $\pm$ 48.74	22.88 $\pm$ 37.34	22.67 $\pm$ 2.31	8.50 $\pm$ 0.00	292 $\pm$ 4
Benthic fish							
Not using best practice	0.77 $\pm$ 1.06	3.96 $\pm$ 6.53	4.45 $\pm$ 5.74	6.70 $\pm$ 2.91	53.83 $\pm$ 38.58	35.67 $\pm$ 22.47	196 $\pm$ 271
Using best-practice	0.15 $\pm$ 0.05	0.22 $\pm$ 0.20	1.15 $\pm$ 1.17	1.67 $\pm$ 0.15	18.67 $\pm$ 1.53	17.67 $\pm$ 8.08	797 $\pm$ 11
Fingernail clams							
Not using best practice	9.1 $\pm$ 11.2	33.3 $\pm$ 53.4	40.2 $\pm$ 43.8	110 $\pm$ 79	1039 $\pm$ 1040	624 $\pm$ 655	751 $\pm$ 945
Using best-practice	20.3 $\pm$ 9.8	61.7 $\pm$ 72.2	126.3 $\pm$ 2.5	188 $\pm$ 60	3750 $\pm$ 2641	1974 $\pm$ 1447	392 $\pm$ 101

or CRs derived for similar sites. However, uncertainties remain, the most important of which are outlined below.

At the project planning stage, although baseline radionuclide activity concentrations in water and sediment can be collected, there remains considerable uncertainty in predicting radionuclide distribution in the receiving environment. Overall,  $K_d$  values varied over six orders of magnitude for  $^{210}\text{Po}$  and  $^{230}\text{Th}$ , three orders of magnitude for  $^{226}\text{Ra}$  and  $^{238}\text{U}$  and two orders of magnitude for  $^{210}\text{Pb}$  (Fig. 1). The wide variation in partition coefficient values between models proportionally resulted in wide variations in predicted water and sediment concentrations, and consequently organism activity concentrations. At the planning stage of a uranium mine and mill, partition coefficients predicted during operations need to be carefully chosen based on site-specific affinity of each radionuclide to the sediment phase (Lofts and Tipping 1999; Monte et al., 2009; Lofts et al., 2015), which is affected by pH, ionic strength, clay minerals and organic carbon content (Sheppard and Thibault 1990; Thibault et al., 1990; US Environmental Protection Agency, 1999; 2004; Environmental Modelling for Radiation Safety, 2012; Kumar et al., 2019). Modellers also need to consider that during early operations, radionuclides activity concentrations are at background levels in the sediment, but the radionuclide is present in water, the partition coefficient may initially be low and then increase over time as sediments become increasingly contaminated. This effect can be observed in our dataset, which shows low partition coefficients at sites not exposed to uranium mining activities and higher partition coefficients at sites exposed to mining activities (Table 2). The predicted increase in partition coefficients during operations will further depend on the time required to reach equilibrium of radionuclide activity concentrations between water and sediments. The time required to reach equilibrium will in turn be affected by the controls implemented to limit releases of radionuclides from mining effluent, tailings and waste rock management facilities. During operations, predicted partition coefficients can be verified over time by measuring radionuclide activity concentrations in water and sediments.

In our opinion, it is best practice to measure radionuclides in tissue and organs for large organisms and whole organism body for small organisms such as benthic invertebrates if possible. This is feasible during baseline, operations and closure of uranium mining and milling projects. However, we recognize that this is not possible for initial REIAs where future activity concentrations are predicted. To estimate activity concentrations in freshwater organisms, appropriate CRs are needed. The

IAEA handbook of transfer parameters (IAEA 2014) and its underlying database (Coppstone et al., 2013) are helpful sources. As the CRs database only indicates if CRs are from lakes, rivers or marine environments, modellers should obtain water quality information from the original scientific paper. An appropriate CR value would be one with similar water chemistry. Water quality parameters such as pH, calcium, magnesium, alkalinity and dissolved organic carbon can affect the accumulation of radionuclides by biota (Jeffree and Simpson 1984; Sheppard et al., 2005; Goulet et al., 2011). It has been shown that  $^{226}\text{Ra}$  uptake depends on calcium, pH and dissolved organic matter in water (Jeffree and Simpson 1984; Jeffree 1991; IAEA 2013). For  $^{238}\text{U}$ , several authors have also shown that uranium uptake depends on the pH, dissolved organic carbon and alkalinity levels in surface water (Markich, 2002; Fortin et al., 2007; Goulet et al., 2011; van Dam et al., 2012; Goulet et al., 2015; Lofts et al., 2015). Adding to this complexity is exposure of radionuclides from dietary uptake.

In addition, uncertainties remain with the estimation of whole organism body activity concentrations from tissue measurements. Assuming whole organism activity concentration of fish by only measuring radionuclides in flesh and bones and assuming they contribute 85% and 15% of total dose to fish is a simplification. The assumption that skeleton accounted for 15% of total fish body mass is based on only six channel catfish that were raised in the laboratory (Cameron 1975). Yankovich (2009) suggested that skeleton can account for 2–9% of total body mass. As the radionuclides considered in this exercise accumulate more in bones than in muscle, assuming the skeleton accounts for 15% of total fish body mass appears conservative, based on the limited information available. Additional uncertainty arises by assuming that the remaining fish mass (i.e., 85%) is muscle tissue. In fact, review of the scientific literature indicates that the proportion of muscle to total body mass is lower and ranges from 47 to 77% (Cameron 1975; Hogstrand et al., 2003; Yankovich, 2009) because other organs contribute to whole organisms body mass. For instance, Hogstrand et al. (2003) reported that whole blood, intestine, skin, gill, liver and kidney accounted for 13% of total body mass in Rainbow Trout. Yankovich (2009; Table 1) reported that the proportion of organs other than bone and muscle could account for as much as 30% of total body mass; any impact of this depends upon the distribution of radionuclides among the soft tissues. Hence, we recommend measuring radionuclide activity concentrations in organs of fish and whole organism body of smaller organisms under different exposure when possible.

Organs could have different accumulation or dose rates of some radionuclides (Stark et al., 2017). Anatomically detailed numerical phantoms have been developed for some animals including Rainbow Trout to better predict radiation doses to organs (Ruedig et al., 2014, 2015; Martinez et al., 2016). However, for regulatory assessment, such models are unnecessarily complex and the commonly used simplified geometries (usually an assumption of a homogenous ellipsoid) appear to give conservative assessments compared to voxel models (Ruedig et al., 2014, 2015). Voxel phantoms likely have a role in trying to interpret effects studies including those at impacted sites (Beaugelin-Seiller et al., 2020).

Assumptions related to  $^{238}\text{U}$  decay chains also introduce uncertainty in dose rate assessments. First, the milling of uranium targets the removal of  $^{238}\text{U}$  from the ore, likely leaving the remaining daughters in the tailings and effluent stream. As a result, using secular equilibrium assumption may not be conservative and so monitoring of  $^{238}\text{U}$  and decay daughters activity concentrations in water, sediment, fish tissue and whole organism body of small organisms is recommended during operations and site closure. Second, at sites that have operated for a long period of time, the activity concentrations of  $^{238}\text{U}$  and decay daughters change with depth in response to historical releases. In those sediments, benthic invertebrates can be exposed within the top 15 cm sediment layer (US Environmental Protection Agency, 2015) but this varies between species as, for example, *Chaoborus flavicans* larvae remain in the top 2 cm (Gosselin and Hare 2005). Hence, vertical heterogeneity of radionuclide activity concentrations in sediment also needs to be considered when assessing exposure to organisms exposed to sediments (Beaugelin-Seiller 2014). Finally, there is also uncertainty regarding the assumption of secular equilibrium when calculating dose conversion factors. The models used in this study had different assumptions with regard to the daughters included within the parent dose conversion coefficient. For instance the ERICA Tool version used assumed daughter with half-lives shorter than 10 d are included in the parent dose conversion coefficient, whilst RESRAD-BIOTA users could select a 100 year or 180 d cut-off depending upon the assessment level, the default within R&D128 is to include  $^{234}\text{U}$  within the  $^{238}\text{U}$  dose conversion coefficient (see discussion in Vives i Batlle et al., 2007; 2011). These assumptions of secular equilibria in effect mean that the activity concentrations of parent and daughters included in the dose conversion coefficient are assumed to be the same in both environmental media and organisms. Such an assumption will not always be valid when assessing uranium industry sites, the ICRP (2017) have published an approach whereby daughters could be considered separately in an assessment, though this requires input data or potential appropriate CR values to be available for the daughters.

#### 4. Conclusions

The objective of this international comparison exercise was to provide a systematic and structured methodology to reduce uncertainty in dose assessments to non-human biota for the entire life-cycle of uranium facilities discharging into aquatic systems. The results showed that considerable variations in activity concentration predictions were derived from the choice of all or a subset of radionuclides, from difference in how decay chains are included, and from the choice of partition coefficients and concentration ratios. The exercise indicated that use of the default concentration ratio values available in some models, which are often designed to be conservatively protective for screening assessment scenarios to be applied at the project planning stage, can lead to poor prediction of the transfer of some radionuclides from water to freshwater organisms. Poor predictions, if overly conservative, can raise unnecessary public concerns and regulatory/management activities whilst under-prediction may underestimate the risk to biota. Hence, to aim for balanced regulatory/management decisions, this body of work proposes a more systematic and structured approach to characterizing sources of uncertainty in the assessment of risk.

As such, we presented a recommended best-practice approach to predict radionuclide activity concentrations and dose rates to freshwater organisms exposed to radionuclides from uranium mining/milling activities. This prioritizes measurements of whole organism body activity concentrations to estimate internal dose rates, and measurements of radionuclide activity concentrations in water and sediment to calculate external dose rates. As a result, we recommend the measurement of radionuclide activity concentrations in whole organism body of appropriate species at operating uranium mines and mills sites along with water quality parameters. Integration of these concentration ratios with associated water quality in databases would help in the selection of realistic concentrations ratios at the project planning stage, reduce uncertainties in dose assessments and allow for balanced regulatory/management decisions regarding the necessary controls to be implemented for the protection of the environment.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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