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Should we ignore U-235 series contribution to dose?

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Should we ignore U-235 series contribution to dose?

Highlights:

- Realistic ecological risk assessment infers a complete inventory of radionuclides
- U-235 family may not be minor when assessing total dose rates experienced by biota
- There is a need to investigate the real state of equilibrium decay of U chains
- There is a need to improve the capacity to measure all elements of the U decay chains

Should we ignore U-235 series contribution to dose?

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Abstract. Environmental Risk Assessment (ERA) methodology for radioactive substances is an important regulatory tool for assessing the safety of licensed nuclear facilities for wildlife, and the environment as a whole. ERAs are therefore expected to be both fit for purpose and conservative. When uranium isotopes are assessed, there are many radioactive decay products which could be considered. However, risk assessors usually assume ^{235}U and its daughters contribute negligibly to radiological dose. The validity of this assumption has not been tested: what might the ^{235}U family contribution be and how does the estimate depend on the assumptions applied? In this paper we address this question by considering aquatic wildlife in Canadian lakes exposed to historic uranium mining practices. A full theoretical approach was used, in parallel to a more realistic assessment based on measurements of several elements of the U decay chains. The ^{235}U family contribution varied between about 4% to 75% of the total dose rate depending on the assumptions of the equilibrium state of the decay chains. Hence, ignoring the ^{235}U series will not result in conservative dose assessments for wildlife. These arguments provide a strong case for more *in situ* measurements of the important members of the ^{235}U chain and for its consideration in dose assessments

1. Introduction

The mining and milling of uranium ore bodies result in releases of uranium and radioactive decay products to aquatic ecosystems. Although modern effluent controls are efficient, operational releases result in the accumulation of contaminants in near field sediments. Predicting ecological risks in these near field aquatic systems is complicated by the many radioactive daughters of the uranium decay series, and the partitioning of contaminants between water and sediments. Predictive ecological risk assessments are therefore conservative to compensate for data gaps and uncertainties to ensure the protection of the receiving aquatic environment.

It is our current understanding that ecological risks appear to be higher for chemical toxicity than radiological toxicity for natural uranium based on certain assumptions about attainment of secular

28 equilibrium and partitioning of daughters (Mathews et al., 2009). It therefore remains important to
29 refine radiological risk assessment methods to fully characterize the hazardous nature of uranium in a
30 fully integrated manner for all associated contaminants and pathways.

31 Both wildlife and human radioprotection systems share the concept of additive risk assuming that
32 effects of exposure to radioactivity are linked to the dose, or energy, received by organisms regardless
33 of the radionuclide. In theory such a concept relies upon a complete inventory of radionuclides to
34 which receptors are exposed so that total radiological risk is not underestimated.

35 Three radioisotopes of uranium are naturally found in the environment: ^{234}U , ^{235}U and ^{238}U . ^{238}U and
36 ^{234}U each represent 49% to the specific activity of natural uranium (Cossonnet et al., 2001) and are
37 generally considered in dose assessments. ^{238}U is the precursor of a radioactive decay chain, producing
38 a long series of radioactive daughters including isotopes such as ^{234}U , ^{230}Th , ^{226}Ra , ^{210}Pb , and ^{210}Po
39 (Fig. 1), that can contribute significantly to dose. As a result, ^{238}U and daughters radionuclides ^{230}Th ,
40 ^{226}Ra , ^{210}Po and ^{210}Pb are routinely monitored in the environment, for instance, downstream of
41 decommissioned and operating U mines and mills.

42 In contrast, Uranium-235 contributes only 2% to the specific activity of natural uranium (Cossonnet et
43 al., 2001), and is generally not explicitly considered in dose assessments, being either ignored or at
44 best estimated from ^{238}U data (the isotopic ratio $^{235}\text{U}/^{238}\text{U}$ is approximately 0.04). ^{235}U is also a
45 precursor of a radioactive decay chain, with seven radioactive daughters that may contribute
46 significantly to dose (^{231}Pa , ^{227}Th , ^{223}Ra , ^{219}Rn , ^{215}Po and ^{211}Bi (Table A1)). However, there are no
47 measured data for components of the ^{235}U decay series in environmental samples, because their
48 analysis methods are complex and costly (Sheppard and Herod, 2012). Instead, radio-ecologists can
49 only estimate the activity of daughter radionuclides in environmental media and in non-human biota
50 by assuming that radionuclide daughters are in a given equilibrium with the parent ^{235}U isotope (which
51 concentration is usually assumed and not measured).

52 This paper addresses if ignoring ^{235}U series radionuclides is justified using an example of a freshwater
53 environment at a historic uranium mining area in Canada. These data have also been used as part of a
54 scenario for an International Atomic Energy Agency modelling exercise (IAEA in-press).

55 **2. Overview of Canadian U mines scenario**

56 The scenario was based upon data collected in the vicinity of historic mining and milling sites in
57 northern Saskatchewan (Canada). Here we present an overview of the elements relevant to the present
58 study. Participants were asked to estimate the weighted dose rates received by benthic and pelagic
59 fish, and aquatic invertebrates. They were provided with radionuclide measurements (^{238}U , ^{230}Th ,
60 ^{226}Ra , ^{210}Po , ^{210}Pb) in water, sediments and organisms, the availability of data differed between
61 samples types and the sites included in the exercise. There were large differences between some model
62 results, from estimated activity concentrations to calculated dose rates. One major difference between
63 approaches was the way in which U isotopes and their decay products were taken into account. At one
64 extreme, assessments only considered the five measured radionuclides for which information was
65 provided, whereas others considered an exhaustive approach that included all U-238 series
66 radionuclides (IAEA in-press). Virtually all of the participants ignored the contribution of the U-235
67 series.

68 This paper focuses on the validity of this latter assumption. Firstly the ^{235}U family contribution to dose
69 rates experienced by aquatic organisms was assessed under the hypothesis of steady state equilibrium
70 between all components of the decay chains, as often assumed in the absence of any measurement
71 data. This result was then compared to a more realistic approach taking into account the available
72 information from one of the Canadian sites included in the scenario.

73 **3. Method**

74 *3.1 General principles*

75 Radionuclides having the same mode of action are assumed to have additive effects. To inform about
76 radiological risks, an environmental risk assessment for uranium should therefore consider all
77 daughter products associated with the element and which may contribute significantly to dose. Here

78 we will consider only those uranium decay products that exhibit a branching ratio higher than 0.9 (Fig.
79 1 and 2). Only some of the decay products are measurable *via* classical nuclear metrology methods:
80 the six first members of the chain are relatively easily quantified by spectrometry (γ or α), if their
81 activity is sufficiently high. For the others, it is only possible to make assumptions regarding the
82 equilibrium state of the decay chain to estimate their activities.

83 The basic equation to assess the total dose rate $DR(I,O)$ received by an organism O exposed to a
84 radionuclide I is the following (Beresford et al., 2007):

$$85 \quad DR(I,O) = DCC_{int}(I,O) \cdot CR(I,O) \cdot C_{media}(I) + DCC_{ext}(I,O) \cdot C_{media}(I)$$

86 where DCC_{int} and DCC_{ext} are the dose conversion coefficients relating organism activity and media
87 (C_{media}) activity concentrations to internal and external dose rates respectively ($\mu\text{Gy h}^{-1}/\text{Bq kg}^{-1}$). In the
88 case of aquatic systems C_{media} may be water or sediment activity concentrations for pelagic or benthic
89 organisms respectively; for organisms at the sediment-water interface $DCC_{ext}(I,O) \cdot C_{media}(I)$ is
90 estimated for both media types usually assuming 50% exposure to sediments and 50% to water.

91 There are two possibilities to take into account daughter products for more realism in an assessment.
92 The first approach consists of considering the decay chain of interest in an integrated manner through
93 the use of a ‘family DCC’ that includes all or some of the daughters, depending on their half-lives.
94 This assumes secular equilibrium between the parent radionuclide and the decay products both in the
95 external media and inside organisms. As an example of this first solution, dosimetric approach used to
96 derive DCC values in the ERICA Tool (Brown et al., 2008) includes daughter products with half-lives
97 up to 10 days (e.g. the DCCs for ^{234}Th include $^{234\text{m}}\text{Pa}$) (Ulanovsky et al., 2008). In contrast, the
98 RESRAD-BIOTA code (ISCORS, 2004) includes daughters with half-lives lower than a user-
99 selectable cut-off of 180 d or 100 years. These methods have one major limitation, they suppose that
100 daughter products and their parent are subject to the same transfer processes, i.e. the same transfer
101 parameters are in-effect applied to all the radionuclides included in the family DCC. This is a
102 simplifying assumption which has not been tested to our knowledge; moreover there is no clear
103 scientific justification rather it has been adopted for pragmatic reasons. Without evidence there is no

104 way to know if this approach is conservative. In addition, users have to take care to not calculate doses
 105 for daughter products already integrated in the DCCs, an easy conceptual error leading to an
 106 overestimation of the radiological risk (Vives i Batlle et al., 2007).

107 The work described here uses individual DCCs for each radionuclide of the U-decay series. The DCCs
 108 (Supplementary material, Table A1) were calculated using the EDEN software (Beaugelin-Seiller et
 109 al., 2006) assuming geometry details as provided in the Canadian U mine scenario (IAEA in-press) for
 110 two organisms living in contrasting habitats, a pelagic fish (pike, *Esox lucius*) and a benthic
 111 invertebrate, a *Pisidium* species mollusc (Table 1). A pike was assumed to spend 75% of its time in
 112 water (in the middle of a 2 m water column) and 25% at the sediment interface (on a 0.5 m sediment
 113 layer under the 2 m water column), whereas a mollusc was assumed to spend all its time at the water-
 114 sediment interface. A supplementary exposure scenario was also considered, in which the mollusc is
 115 located in the middle of the sediment layer. In addition to ^{238}U and daughters, including the ^{235}U series
 116 in an Ecological Risk Assessment (ERA) leads to consideration of two additional elements, Ac and Tl,
 117 and 11 additional radioisotopes (Fig. 2). DCCs were weighted according to the relative biological
 118 effectiveness of the different radiations as suggested by Pröhl et al. (2003): 10 for α , 3 for β and 1 for
 119 γ emissions.

120 Table 1: Assumed organism characteristics

Species	Anatomical parameters (size in cm / mass in kg)				References
	Length	Height	Width	Mass	
Pike (<i>Esox lucius</i>)	50	15	10	1200	Golder Associates, 2006, 2008 Canada North Environmental Services, 2003, 2005
<i>Pisidium sp.</i>	2.5	1.5	1	1.6	Kilgour and Mackie, 1991 Funk and Reckendorfer, 2008

121

122 3.2. Theoretical approach

123 Assumptions of equilibrium within decay chains were made for both water and sediment. Table 2
 124 presents relative activity concentrations of the daughter products assuming 1 Bq L⁻¹ or Bq kg⁻¹ (dry
 125 mass, dm) ^{238}U in water or sediment respectively estimated under the hypothesis of radioactive decay

126 equilibrium. Outgassing of radon with a distribution coefficient of $0.4 \text{ m}^3 \text{ m}^{-3}$ (Sabroux, 1998) and a
 127 natural isotopic ratio between ^{235}U and ^{238}U of 0.047 (Cossonnet *et al.*, 2001) were assumed. The
 128 $^{235}\text{U}:$ ^{238}U ratio has some natural variability, depending on the matrix. Sheppard & Herod (2012) cited
 129 an average ratio of 0.028 and 0.035 respectively for water and soil from the literature. They also
 130 acquired new data for water, from which they estimated a ratio of 0.048. The exact value of this ratio
 131 is not critical to our study's objectives, as while the variability in environmental samples varies from
 132 less than 0.03 % (Cowan and Adler, 1976; Richter *et al.*, 1999; Bopp *et al.*, 2009) to 0.3% (Stirling *et*
 133 *al.*, 2007; Weyer *et al.*, 2008; Sun *et al.*, 2008; Del Papa *et al.*, 2010), the ^{235}U activity concentration is
 134 low relative to ^{238}U .

135 Table 2: Relative activity concentrations used for the theoretical approach

Radionuclide	Concentration (Bq L ⁻¹ or Bq kg ⁻¹)	Hypothesis
^{238}U	1	-
^{234}Th , $^{234\text{m}}\text{Pa}$, ^{234}U , ^{230}Th , ^{226}Ra	1	Radioactive equilibrium with ^{238}U
^{222}Rn	0.4	Loss by outgassing
^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po , ^{210}Pb , ^{210}Bi , ^{210}Po	0.4	Radioactive equilibrium with ^{222}Rn
^{235}U	0.047	Natural isotopic ratio $^{235}\text{U}/^{238}\text{U}$
^{231}Th , ^{231}Pa , ^{227}Ac , ^{227}Th , ^{223}Ra	0.047	Radioactive equilibrium with ^{235}U
^{219}Rn	0.019	Loss by outgassing
^{215}Po , ^{211}Pb , ^{211}Bi , ^{207}Tl	0.019	Radioactive equilibrium with ^{219}Rn

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137 When radioactive decay equilibrium was assumed in water, we assessed the sediment activity
 138 concentrations under the hypothesis of steady-state transfer as determined by the classical partition
 139 coefficient K_d (Table 3, Fig.3 upper graph). When decay equilibrium was assumed in sediment, water
 140 concentrations were estimated in an inverse way from sediment concentrations. In either case,
 141 organism activity concentrations were then obtained by applying concentration ratios to water activity
 142 concentrations (Table 3, Fig.3 lower graph).

143 Table 3: Values and origin of transfer parameters

Isotope	Kd		CR pike		CR <i>Pisidium</i>	
	Value	Origin*	Value	Origin	Value	Origin
²³⁸ U	2.87E+02	ERICA Tool 2014	7.24E+01	Copplestone et al., 2013	5.57E+02	Copplestone et al., 2013
²³⁴ Th	1.96E+07	ERICA Tool 2014	7.13E+02	Copplestone et al., 2013	1.04E+04	Mollusc Am**
^{234m} Pa	1.96E+07	ERICA Tool 2014	8.33E+02	Mollusc Am	1.04E+04	Mollusc Am
²³⁴ U	2.87E+02	ERICA Tool 2014	7.24E+01	Copplestone et al., 2013	5.57E+02	Copplestone et al., 2013
²³⁰ Th	1.96E+07	ERICA Tool 2014	7.13E+02	Copplestone et al., 2013	1.04E+04	Mollusc Am
²²⁶ Ra	1.40E+04	ERICA Tool 2014	1.81E+02	Copplestone et al., 2013	2.43E+04	Copplestone et al., 2013
²²² Rn	8.00E-01	Brown et al.,2004	8.00E-01	Brown et al., 2004	8.00E-01	Brown et al.,2004
²¹⁸ Po	1.78E+04	ERICA Tool 2014	2.03E+03	Copplestone et al., 2013	1.24E+05	Copplestone et al., 2013
²¹⁴ Pb	1.78E+04	ERICA Tool 2014	1.23E+03	Copplestone et al., 2013	5.79E+03	Copplestone et al., 2013
²¹⁴ Bi	1.20E+03	Wang et al., 2001 ; 2003	1.50E+01	Staven et al., 2003	1.00E+05	Staven et al.,2003
²¹⁴ Po	1.78E+04	ERICA Tool 2014	2.03E+03	Copplestone et al., 2013	1.24E+05	Copplestone et al., 2013
²¹⁰ Pb	1.78E+04	ERICA Tool 2014	1.23E+03	Copplestone et al., 2013	5.79E+03	Copplestone et al., 2013
²¹⁰ Bi	1.20E+03	Wang et al., 2001 ; 2003	1.50E+01	Staven et al., 2003	1.00E+05	Staven et al.,2003
²¹⁰ Po	1.78E+04	ERICA Tool 2014	2.03E+03	Copplestone et al., 2013	1.24E+05	Copplestone et al., 2013
²³⁵ U	2.87E+02	ERICA Tool 2014	7.24E+01	Copplestone et al., 2013	5.57E+02	Copplestone et al., 2013
²³¹ Th	1.96E+07	ERICA Tool 2014	7.13E+02	Copplestone et al., 2013	1.04E+04	Mollusc Am
²³¹ Pa	1.96E+07	ERICA Tool 2014	8.33E+02	Mollusc Am	1.04E+04	Mollusc Am
²²⁷ Ac	2.00E+06	IAEA 2001	2.50E+01	Staven et al.,2003	1.00E+03	Staven et al.,2003
²²⁷ Th	1.96E+07	ERICA Tool 2014	7.13E+02	Copplestone et al., 2013	1.04E+04	Mollusc Am
²²³ Ra	1.40E+04	ERICA Tool 2014	1.81E+02	Copplestone et al., 2013	2.43E+04	Copplestone et al., 2013
²¹⁹ Rn	8.00E-01	Brown et al.,2004	8.00E-01	Brown et al.,2004	8.00E-01	Brown et al.,2004
²¹⁵ Po	1.78E+04	ERICA Tool 2014	2.03E+03	Copplestone et al., 2013	1.24E+05	Copplestone et al., 2013
²¹¹ Pb	1.78E+04	ERICA Tool 2014	1.23E+03	Copplestone et al., 2013	5.79E+03	Copplestone et al., 2013
²¹¹ Bi	1.20E+03	Wang et al., 2001 ; 2003	1.50E+01	Staven et al.,2003	1.00E+05	Staven et al.,2003
²⁰⁷ Tl	2.00E-04	IAEA 2001	1.00E+02	IAEA 2014	5.00E+03	Staven et al.,2003

144 *ERICA Tool 2014: extracted from the databases according to the version 1.2 released in November 2014 (<http://www.ERICA-tool.eu/>)/ **Mollusc Am: extrapolation from Mollusc Am CR value

146 Up-to-date values were used, consulting the latest version of different databases (ERICA Tool -
147 V1.2.0- 2014 (<http://www.ERICA-tool.eu/>); Wildlife Transfer Database (Copplestone et al., 2013)). The
148 CR values for *Pisidium* are those for a bivalve mollusc when available, and for pike for a pelagic fish.
149 Due to lack of data, some older documents had to be consulted for some values, and finally, some
150 extrapolations were required as indicated in Table 3 (these extrapolations were in accordance with
151 those used in various assessment tools (e.g. Brown et al. 2013)).

152 3.3. Realistic approach

153 The approach presented above relies on assumptions about decay chain equilibrium, deriving all the
154 information for daughters from the ^{238}U activity concentrations in water or sediment. Comprehensive
155 data for all the decay products have yet to be obtained for environmental samples due to
156 methodological constraints, but some representative data are available for a few key isotopes. This was
157 the case for Keddy Bay of Beaverlodge Lake (one of the sites included in the Canadian U mine
158 scenario (IAEA, in-press)), from which we have selected data for analysis (Table 4). Data gaps were
159 filled following the same extrapolation rules as for the theoretical approach. These measurements were
160 used preferentially to model activity concentrations in media and organisms (and estimate dose rates).
161 *In situ* transfer parameters were derived when possible, using data obtained at, or close to, Keddy Bay
162 (IAEA, in-press). A Kd value for uranium isotopes was estimated for the site and we determined site
163 specific concentration ratios for uranium, radium and lead for pike (Table 5).

164 Table 4: Available measurements at Keddy Bay

Radionuclide concentration		
Radionuclide	water (Bq L ⁻¹)	sediment (Bq kg ⁻¹ dm)
^{238}U	1.83x10 ⁰	1.18x10 ³
^{226}Ra	1.00x10 ⁻²	n.a.
^{210}Pb	n.a.	2.53x10 ²

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168 Table 5: *In situ* values of transfer parameters for the Keddy Bay case study

Isotope	Kd	CR pike
U	6.44×10^2	2.70×10^0
Ra	n.a	2.62×10^1
Pb	n.a	8.04×10^1

169

170 4. Results

171 4.1. Activity concentrations

172 In the medium (water or sediment) where decay chains are assumed to be at equilibrium, activity
 173 concentrations obtained applying the theoretical approach decrease gradually from ^{238}U to the ^{235}U
 174 chain. This logical continuity is not seen when converting water activity concentrations into sediment
 175 activity concentrations, or vice-versa, using Kd values. For instance, if we assume decay equilibrium
 176 in water then the highest values in sediment are predicted for Th (and Pa as its Kd is extrapolated from
 177 the value for Th) and Ac, due to their high Kd values (Fig. 3 upper graph). A similar phenomenon
 178 occurs when calculating organism activity concentrations, for which the highest values are obtained
 179 for Pa in fish, Po in fish and invertebrates, and Bi in invertebrates, due to the high associated CRs (Fig.
 180 3 lower graph). The ranking of radionuclides differs as if it is established from water activity
 181 concentration, from sediment activity concentration or from dose rates.

182 At Keddy Bay, the ^{226}Ra concentration measured in water is actually about one hundred times lower
 183 than expected assuming decay equilibrium in water based on the activity of ^{238}U . No data were
 184 available for ^{235}U and its daughters, and consequently we applied the theoretical approach described
 185 above to estimate activity concentrations of this radionuclide. From this we obtained a mixed
 186 (measurement plus extrapolation) concentration spectrum in water, considering decay equilibrium,
 187 different to the fully theoretical one (Fig.4 upper graph). ^{235}U family activity concentrations are in this
 188 case considerably higher than those of ^{226}Ra and its decay products. If decay equilibrium is assumed in
 189 sediment and the additional data from Tables 4 and 5 are used, the two approaches give estimates
 190 broadly in agreement (Fig.4 lower graph).

191 4.2. Total dose rates

192 Following the theoretical approach, Figure 5 presents the estimated contribution to total dose rate of
193 pike and *Pisidium* assuming isotopic equilibrium in water (upper graph) and comparing this with an
194 equilibrium assumption for sediment (lower graph). Assuming radioactive decay equilibrium in water,
195 Po isotopes are the major contributors to total dose for both organisms considered (70 and 80% of the
196 total dose rates for fish and *Pisidium* respectively). This is in part due to the high CR values for Po.
197 These isotopes also contribute significantly to the dose rate assessed for the mollusc when considering
198 isotopic equilibrium in sediment, but to a lesser extent, contributing about 50% of total dose. This is
199 not the case for fish, for which 80% of the total dose rate originates from its internal exposure to ^{222}Rn ,
200 ^{238}U and ^{234}U when sediment isotopic equilibrium is assumed. For these cases, the contribution of the
201 ^{235}U family to total dose rate for *Pisidium* varies from about 4 % (equilibrium in water) to 12%
202 (equilibrium in sediment), when the invertebrate is in the sediment or at its surface (Fig. 5). For fish,
203 the percentage increases to about 5% considering decay chains at equilibrium in water, but is lower
204 (~ 4%) considering decay chains at equilibrium in sediment.

205 Using the realistic approach, differences in activity concentrations measured or estimated at Keddy
206 Bay propagated through all calculations, from transfer to dosimetry. After the contribution of ^{230}Th (~
207 40%, Fig. 6 upper graph), the highest dose rates were for the mollusc for decay chains at equilibrium
208 in water due to exposure from ^{215}Po and ^{211}Bi , two members of the ^{235}U decay chain. These contributed
209 more than 25% of the total estimated absorbed dose rate. This resulted mainly from the internal
210 exposure. These three radionuclides have some of the highest CR and internal DCC values, combined
211 with comparatively higher activities of ^{235}U and daughter products in water with regard to the
212 theoretical case. For fish, the highest dose rate is associated with ^{230}Th (50%), a decay product of ^{238}U ,
213 followed by one of the ^{235}U daughter products, ^{227}Th (25%) (Fig.6). Overall, the whole ^{235}U family
214 contribution is similar for both organisms. The contribution is substantial as it approaches 40% of the
215 total dose rate.

216 Assuming decay equilibrium in sediment (Fig.6 lower graph) results in a dominance of ^{210}Po in the
217 total dose rate (~63%) for mollusc though less for fish (~38%). For fish, the main contributor is ^{222}Rn
218 (~45%). Its dominance in fish results from a high internal dose rate. Radon internal DCCs are among
219 the highest, along those of Po. In contrast to Po, Rn shows a low CR though also a low Kd, leading to
220 a high activity concentration in water.

221 Although ^{222}Rn is acknowledged to contribute potentially highly to doses for terrestrial organisms, *via*
222 inhalation pathways (Beresford et al., 2012), the importance of its respiration in terms of doses is
223 likely to be less for aquatic animals (Hosseini et al., 2010), exposure from dissolved ^{222}Rn to some
224 organs (e.g. gills and alimentary tract) requires further consideration. Such an argument justifies
225 assessing the impact of ^{222}Rn , despite the lack of robustness of the available CR value. In the absence
226 of measured data (Lucas et al., 1979), the CR value used here for all organisms was obtained from
227 Brown et al. (2004). Brown et al. simply assumed that radon in the water in any organism is in
228 equilibrium with radon in the surrounding water. This is a reasonable assumption for a noble gas
229 which is highly soluble/mobile in water-based “media”. However, this assumption may be far too
230 conservative for the deposition/retention of radon’s short-lived daughters, which are responsible for
231 much of the dose from radon in our simplified theoretical treatment. The retention of any radium
232 decaying *in vivo* in any tissue other than bone may be only a few percent (ICRP, 1993). Our treatment
233 of radon and its daughters in a transfer factor context is highly uncertain, however, despite the need for
234 data, relevant experimental and environmental information remain sparse. Therefore, due to the
235 paucity of data, we acknowledge that it is difficult to interpret the relative importance of the radon
236 contribution to fish exposure, though here we have made an assessment based upon the limited
237 information available.

238 The estimated contribution of the ^{235}U family may be as high as 40% of the total dose rates
239 experienced by aquatic organisms exposed to uranium at Keddy Bay (Fig. 6). This percentage
240 decreases to about 3 to 6% for both organisms when considering decay equilibrium in sediment rather
241 than in water. Lifestyle of organisms significantly impacts the result. Increasing the time spent in the
242 water column by pike to 100% decreases the contribution of the ^{235}U family for the fish to about 17%,

243 assuming decay equilibrium in water. This effect is not seen when decay equilibrium is considered in
244 sediment.

245 The greater contribution of ^{235}U series radionuclides compared to the theoretical approach, at least for
246 decay equilibrium considered in water, is the consequence of the lower concentrations of ^{226}Ra in
247 water based upon measurements rather than assumed equilibrium.

248 5. Discussion

249 5.1. Estimating dose rates using the theoretical as oppose to a more realistic approach

250 The realistic scenario from Keddy Bay identified three dominant radionuclides, ^{230}Th , ^{215}Po and ^{211}Bi ,
251 in the estimates of both mollusc concentrations and total dose rate. Together, they contribute about
252 70% of the total dose. Considering equilibrium of their respective decay chains in water and in the
253 absence of any other information, activity concentrations in water of these isotopes were extrapolated
254 directly from the activity of ^{238}U or, for the two members of the ^{235}U decay chain, from the natural
255 isotopic ratio $^{235}\text{U}/^{238}\text{U}$, taking into account radon outgassing. This last assumption led to relatively
256 low concentrations of these radionuclides in water that are counterbalanced by their high default
257 transfer parameters. Moreover, the three radioisotopes have DCC values amongst the highest for
258 internal exposure of the mollusc of all the radionuclides of the two U-isotope decay chains. The total
259 *Pisidium* dose rate estimated for ^{230}Th , ^{215}Po and ^{211}Bi is then dominated by the internal contribution.
260 Calculation was done assuming transfer at equilibrium, applying element CRs without distinction
261 between isotopes. This approach does not account for half-lives that may be very short (e.g. less than a
262 second for ^{215}Po , about 2 m for ^{211}Bi). Assessing activity concentration of such isotopes in organisms
263 *via* the equilibrium approach may therefore overestimate activity concentration and hence dose rates.

264 The large disequilibrium between ^{238}U and ^{226}Ra activity concentrations measured in water increases
265 the contribution of the ^{235}U family to the dose rates received by both organisms. Compared to the
266 assumption of steady state throughout the ^{238}U decay chain applied in the theoretical approach
267 (implying equal concentrations of the two radionuclides), the break in equilibrium at ^{226}Ra decreased
268 its concentration (and all subsequent daughters) by two orders of magnitude compared to ^{238}U .

269 Whereas ^{235}U family concentrations were reasonably derived from the ^{238}U measurement, applying the
270 natural isotopic ratio $^{235}\text{U}/^{238}\text{U}$ to the entire ^{235}U chain (which includes ^{223}Ra - ^{219}Rn) may not be
271 realistic. This assumption is a potential weak point in the theoretical calculations. This issue needs to
272 be addressed by measuring at least some of the more important members of the ^{235}U chain in
273 sediments, where concentrations are likely high enough to obtain meaningful results. Finally, taking
274 into account the radon degassing for the four last members of the ^{235}U family only decreased their
275 concentrations by about a factor two.

276 The approach described above was based on the use of individual DCCs for each of the radioisotope
277 of the decay chains. It could be argued that this will limit the number of underlying assumptions
278 regarding decay equilibrium. It has to be noted that to conduct the calculation, other numerous
279 assumptions (e.g. transfer parameters, transfer of decay products etc.) are required that may influence
280 the final result. The extent to which the use of individual DCCs may change the weight of the ^{235}U
281 family contribution to dose rates was tested relative to the use of the alternative approach of family
282 DCCs (or integrated DCCs) by applying the ERICA Tool (Brown et al., 2008). The tool lumps
283 together parents and daughters with half-lives ≤ 10 days (Ulanovski et al., 2008). The DCC of ^{226}Ra
284 includes DCCs related to ^{222}Rn , ^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po and ^{218}At (Fig.1). The same assumption
285 applies to ^{210}Pb (daughter included: ^{210}Bi), ^{235}U (daughter included: ^{231}Th) and ^{223}Ra (daughters
286 included: ^{219}Rn , ^{215}Po , ^{211}Pb , ^{211}Bi , ^{207}Tl) (Fig.1 and 2). Uncertainty was considered *via* the production,
287 in parallel, of four sets of predictions, issued from various combinations of transfer parameters values
288 and media concentrations (Table 6). The data set 2b in Table 6 corresponds to the ‘individual DCC’
289 approach discussed above. Aside from the difference in daughter radionuclides considered, EDEN and
290 the ERICA Tool have been shown to generally give comparable results (Vives i Batlle et al., 2011).

291

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294

295 Table 6: The different sets of hypotheses defined to run the ERICA Tool (V1.0)

Data set	Transfer parameters	Estimation of missing concentrations	
		In water	In sediment
1	ERICA Kd values with $CR_{wo-water}$ being taken from IAEA (2014) except Pa and Ac (Table 3)	from sediment applying the Kd's	$^{234}Th, ^{234}U, ^{230}Th = ^{238}U$ $^{210}Po = ^{210}Pb$ $^{235}U = 0.047 \times ^{238}U$ $^{231}Pa, ^{227}Ac, ^{227}Th, ^{223}Ra = ^{235}U$ ^{226}Ra from water applying the Kd
2a	Same as 1	$^{234}Th, ^{234}U, ^{230}Th = ^{238}U$ $^{210}Po, ^{210}Pb = ^{226}Ra$ $^{235}U = 0.047 \times ^{238}U$ $^{231}Pa, ^{227}Ac, ^{227}Th, ^{223}Ra = ^{235}U$	Same as 1
2b	Same as 1	Same as 2a except $^{210}Po, ^{210}Pb = 0.4 \times ^{226}Ra$ (Rn degassing)	From water applying the Kd's
3*	Site specific except Pa and Ac (Table 3)	Same as 1	Same as 1
4a	Same as 3	Same as 2a	Same as 2a
4b	Same as 3	Same as 2b	Same as 2b

296 * Canadian Mining exercise IV (IAEA, in-press)

297 All sets of calculation hypotheses used with the ERICA Tool result in the ^{238}U family dominating the
 298 total dose rates experienced for fish, varying from 91 to 100% (Fig.7B). In contrast, for the mollusc,
 299 the organism closely linked to sediment, the ^{235}U contribution increases from 6% to 75% respectively
 300 from the first to fourth dataset (Fig.7A). As discussed before, the most significant factor contributing
 301 to these differences is the derivation of the water and sediment inputs. The more realistic scenarios,
 302 where measured media data and site specific transfer parameters were input when available (i.e. data
 303 sets 2a, 4a and 4b), resulted in the highest estimated contributions from the ^{235}U -series. Absorbed
 304 internal dose rates from ^{223}Ra dominated (the DCC for ^{223}Ra includes contributions to dose from ^{219}Rn ,
 305 $^{215}Po, ^{211}Pb, ^{211}Bi$ and ^{207}Tl). These Po and Bi isotopes were consistently identified as major
 306 contributors to the internal and total exposure of *Pisidium* applying the individual DCC approach.

307 Dose rate obtained with the ERICA Tool for a given radionuclide is logically sensitive to the transfer
308 parameter value. Site specific values result in higher U-isotope dose rates for fish (by a factor of ~30)
309 but lower Th-isotope dose rates (by a factor of ~15). This effect is smoothed when summing dose rates
310 assessed for each radionuclide to obtain total dose rates. For instance, estimated total dose rates using
311 data set 2a (literature CR and Kd values) and data set 4a (site specific CR and Kd values) are within a
312 factor of two to 10 of each other for fish and mollusc respectively.

313 We demonstrated that realistic scenarios may lead to a contribution of the ^{235}U family to dose rates
314 which, far from being negligible, may become the dominant source of exposure. This is definitively
315 illustrated by the most realistic assessment conducted for Keddy Bay (data set 3; IAEA, in-press), for
316 which the ^{235}U family produced more than 70% of the total dose rate for the mollusc. Ignoring this
317 decay chain may result in underestimations of the radiological risk for the environment. This is
318 particularly true for wildlife closely linked with sediment, especially when decay equilibrium is
319 reached there. However, we should also acknowledge that ^{223}Ra , the main contributor to dose of the
320 ^{235}U -series radionuclides obtained with the ERICA Tool, has a relatively short physical half-life (~11
321 days). Hence equilibrium will not be achieved between tissues and environmental concentrations, i.e.
322 internal dose rates may not be as high as estimated here. Therefore, the present study should be seen as
323 an exercise to assess what could be the consequence of not taking into account ^{235}U and its decay
324 products when assessing biota exposure to radiation. Even if ecological risks appear to be higher for
325 chemical toxicity than radiological toxicity, at least for natural uranium (Mathews et al., 2009), there
326 is a need for a complete characterization of the hazardous nature of uranium. Fully integrating all
327 associated contaminants and pathways is the only way to provide a robust demonstration of the level
328 of associated radiological risk to fauna and flora.

329 5.2. Decay equilibrium in water as opposed to sediments

330 If decay equilibrium is considered in water of the Keddy Bay scenario, activity concentrations of the U
331 chain members in the mollusc (Fig. 8, upper graph) vary generally from 10^1 (lead isotopes, ^{235}U ,
332 ^{227}Ac ...) to 10^3 - 10^4 (Th isotopes, ^{215}Po , ^{211}Bi , etc.) Bq kg^{-1} fresh mass (fm). Radon is an exception,

333 exhibiting especially low values (10^{-3} to 10^{-2} Bq kg⁻¹ fm) due to a low assumed CR. A somewhat
334 similar pattern is observed for pike, which presents lower activity concentrations for all radionuclides.
335 Conversion into dose rate preserves partly the relative isotope distribution (Fig. 8, lower graphs),
336 which explains the contribution of the ²³⁵U family to total dose rate close to 40%.

337 Assuming decay equilibrium in sediment changes drastically both the activity concentrations and dose
338 rate distributions. This hypothesis increases the importance of chain members beyond radon. Po, Pb
339 and Bi isotopes are estimated to have high activity concentrations in *Pisidium* and pike, up to four
340 orders of magnitude higher than those of the chain parents. Measured data were too scarce to support
341 the validation of one assumption vs the other (i.e. decay equilibrium in water rather than in sediment,
342 or vice versa).

343 Considering decay equilibrium in water, the theoretical assumption of equilibrium throughout the two
344 decay chains led to a contribution of the ²³⁵U family to total dose rates of 4% for both organisms.
345 Compared to this result, this estimated contribution is increased in our case study (from 16 to 40%
346 depending on occupancy factors for pike) due to the large disequilibrium between ²³⁸U and ²²⁶Ra, the
347 concentration of the latter being two orders of magnitude lower than expected when considering decay
348 equilibrium. Consequently, all its daughter products activity concentrations are also estimated to be
349 two orders of magnitude lower, increasing the relative part of total dose rates due to the ²³⁵U family.
350 Predicted ²³⁵U concentrations in water are about one order of magnitude higher than those of ²²⁶Ra.
351 These concentrations exceeded those of the members at the end of the ²³⁸U chain, explaining the
352 difference observed between the theoretical calculation and the case study results.

353 **6. Conclusions**

354 We obtained from both the theoretical (assumption of isotope equilibria) and more realistic (inclusion
355 of available site data) approaches significant contributions of the ²³⁵U family, up to 75% of the
356 estimated total dose rate experienced by an organism. These results contradict the common opinion
357 that doses rates from the ²³⁵U series radionuclides may be neglected compared to those from the ²³⁸U
358 series radionuclides. While many aspects of the present work are uncertain and use simplistic

359 assumptions there is a weight of evidence that ^{235}U -series radionuclides have the potential to make
360 important contributions to dose rates.

361 Given the current state of knowledge, we were not able to improve on our assessment (presented here)
362 of the ^{235}U family contribution to dose rate assessment for non-human biota. This exercise
363 nevertheless shows the need for determining the actual state of decay equilibrium of these chains, at
364 least for some characteristic situations. To understand the contribution of the ^{235}U family further, it is
365 essential to ensure a high quality of validated measurement methods. In addition to assessments of
366 contaminated sites this conclusion has implications for current background exposure rates estimated
367 for wildlife due to natural series radionuclides (e.g. Hosseini et al., 2010; Beresford et al., 2008) as
368 these do not take the ^{235}U series into account.

369 The final conclusion of this work concerns the best way to limit estimation bias identified when
370 dealing individually or globally with decay chain members during dose rate assessment. The most
371 realistic result should be obtained with a combination of the two studied approaches, applying family
372 internal DCCs to realistic parent nuclide concentrations in organisms and individual external DCCs to
373 media activity concentrations of individual daughter products.

374

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380

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487

Figure 1: ^{238}U decay chain (italic text: half-life; normal text: branching ratio (BR); grey lines: secondary decay chain with first daughter BR less than 0.9; solid grey boxes: stable element; Nucleonica GmbH, 2015)

Figure 2: ^{235}U decay chain (italics: half-life, normal: branching ratio -BR; grey lines: secondary decay chain with first daughter BR less than 0.9; grey box: stable element; Nucleonica GmbH, 2015)

Figure 3: Theoretical activity concentrations per isotope, based on K_d and CR values, in water vs. sediment (upper graph) and water vs. organisms (lower graph), considering a unit activity concentration of ^{238}U in water where decay equilibrium is achieved in all daughters and considering the ^{235}U decay chain.

Figure 4: Distributions of media concentrations of radionuclides at Keddy Bay (decay chains at equilibrium in water - upper graph- or in sediment - lower graph; black bar: data extrapolated from ^{238}U concentration, grey bar: measurements completed by extrapolations)

Figure 5: Contribution (%) to total dose rates per member of the uranium decay chains for benthic invertebrate (*Pisidium* at the water/sediment interface or in sediment) and fish (pike in water), considering equilibrium either in water (upper graph) or in sediment (lower graph), from an initial theoretical unit concentration of ^{238}U (only main contributors are identified on the graphs).

Figure 6: Contribution (%) to total dose rates per member of the uranium decay chains for benthic invertebrate (*Pisidium* at the water/sediment interface) and fish (pike in water), considering equilibrium either in water (upper graph) or in sediment (lower graph), at Keddy Bay (only main contributors are identified on the graphs).

Figure 7: ^{235}U family contribution (light grey) vs. other contribution (dark grey) to the total dose rate to organisms (A: mollusc; B: fish) as estimated with the ERICA tool for the combinations of transfer parameters and media concentrations in Table 6

Figure 8: Distribution per radionuclide of activity concentrations (upper graphs) and dose rates (lower graphs) for *Pisidium* (on the left) and pike (on the right) at Keddy Bay (decay equilibrium in water -black bar- or in sediment - white bar)

Figure 1

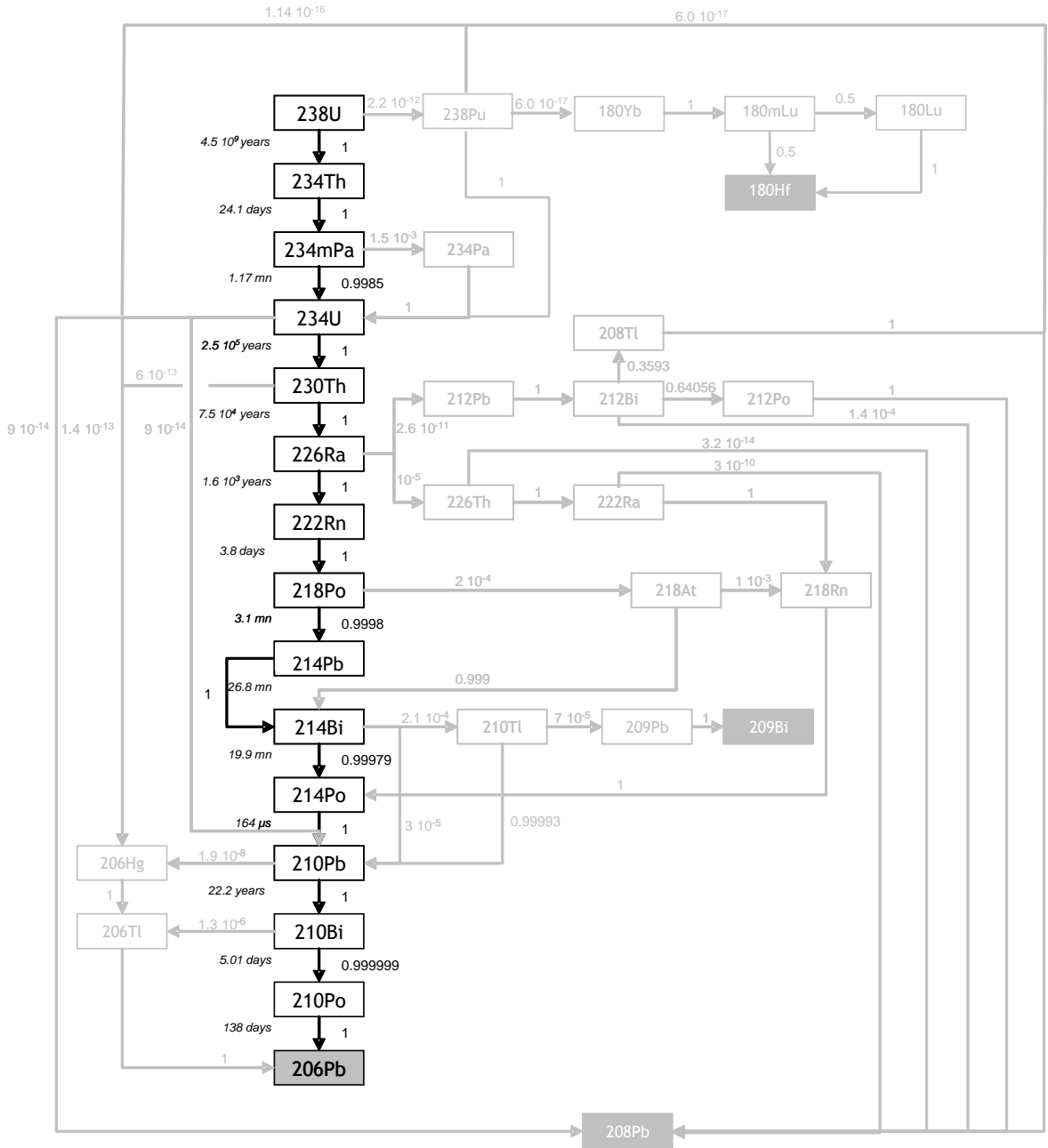


Figure 2

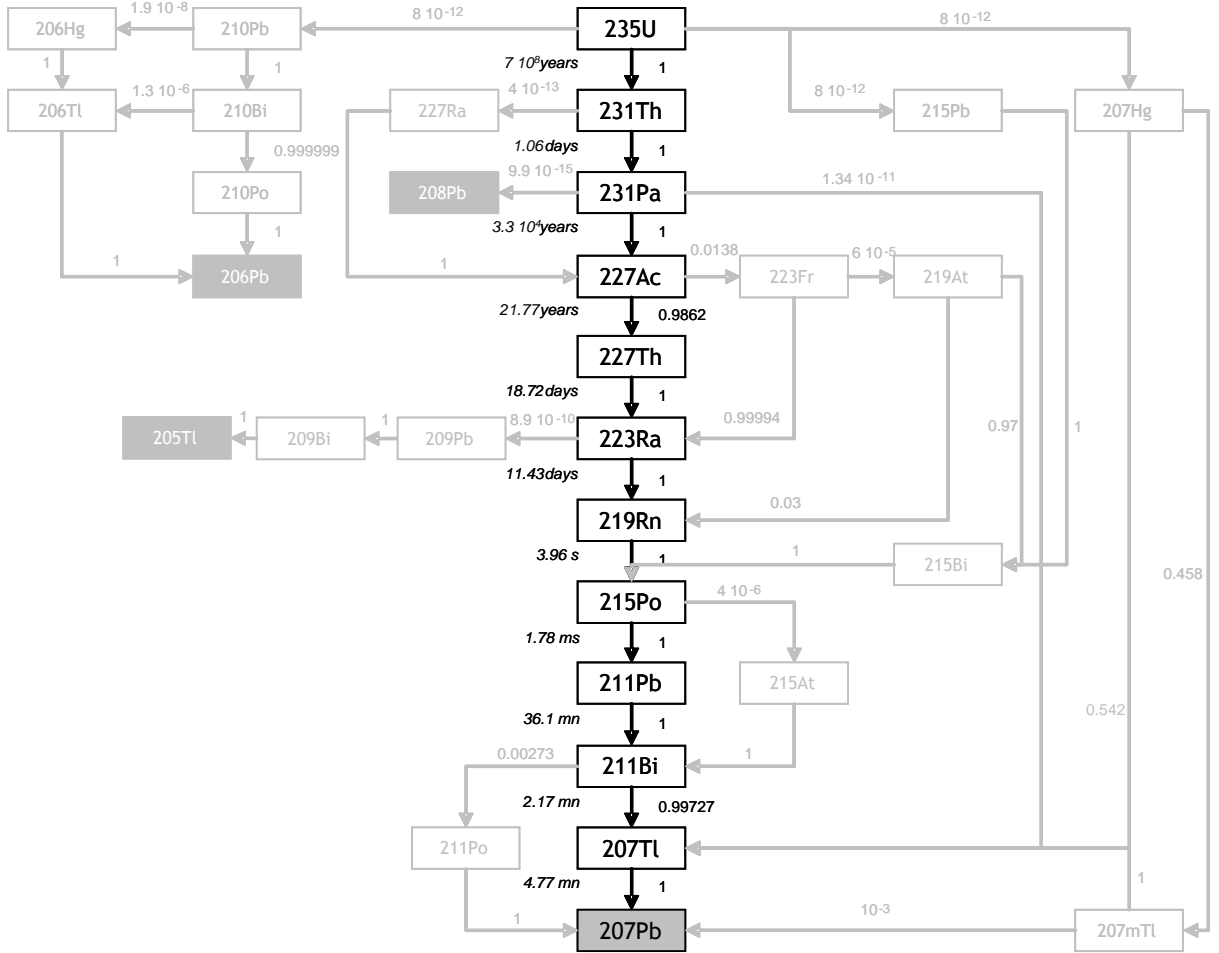


Figure 3

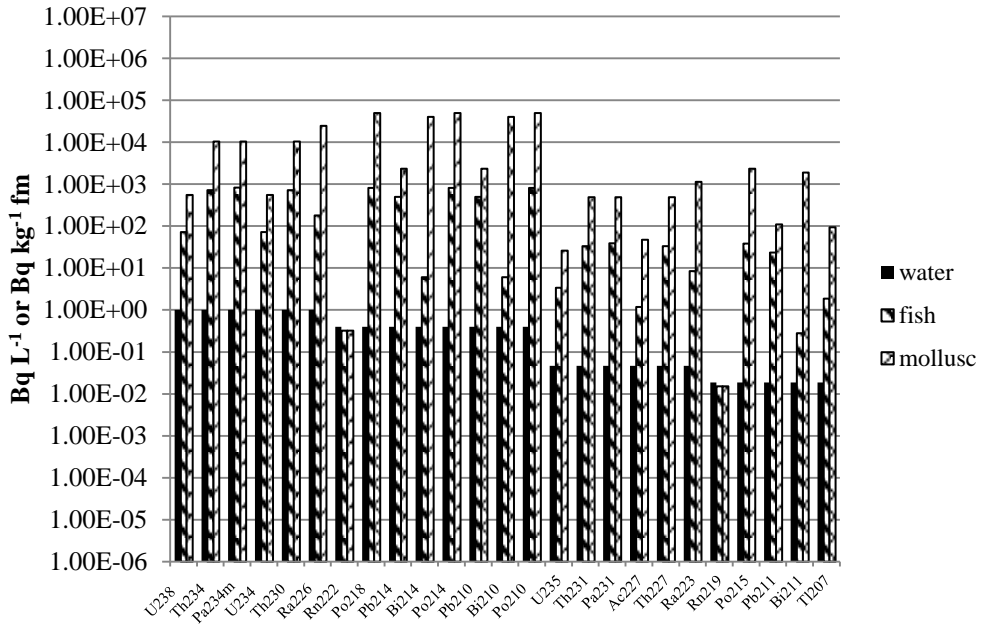
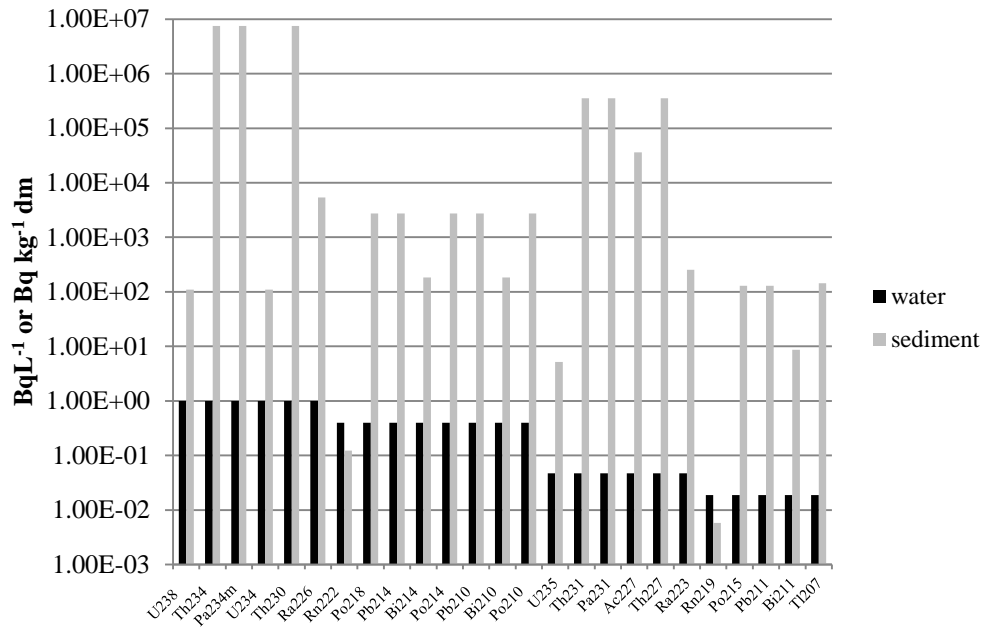
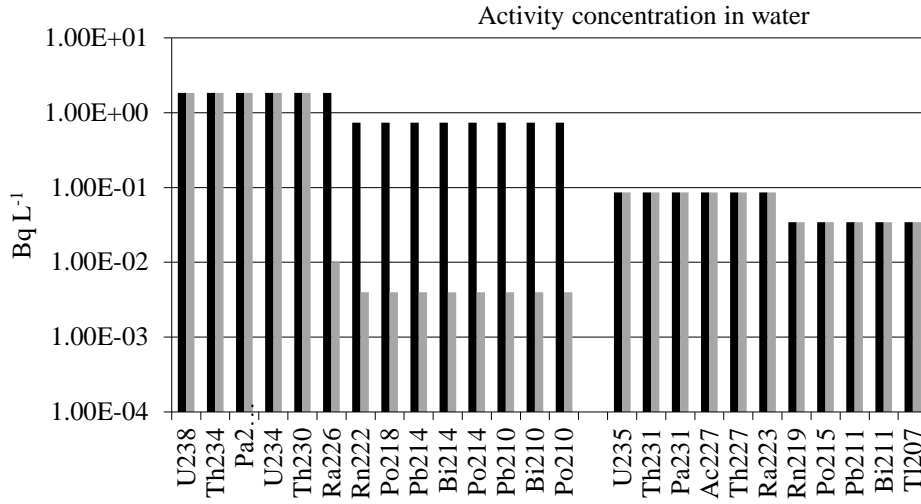


Figure 4

Decay chains at equilibrium in water



Decay chains at equilibrium in sediment

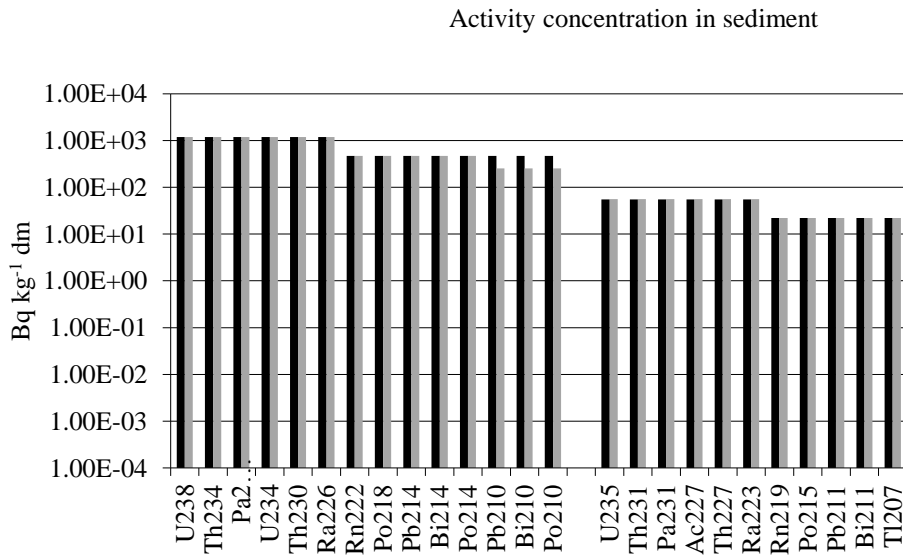
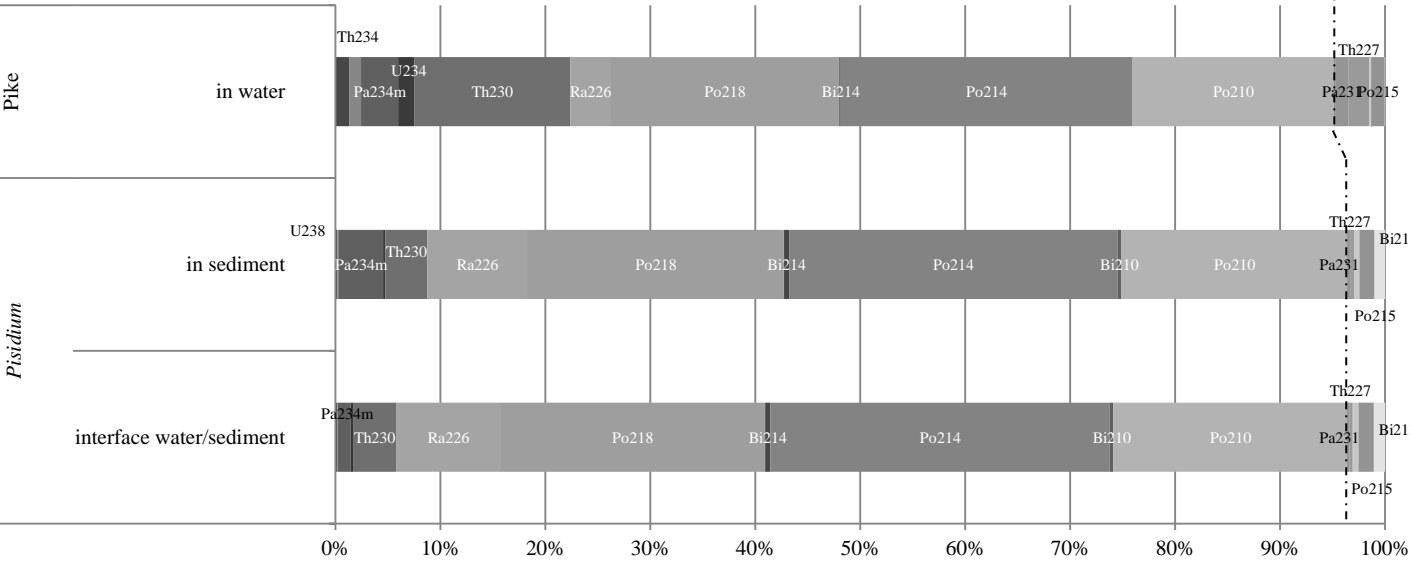


Figure 5

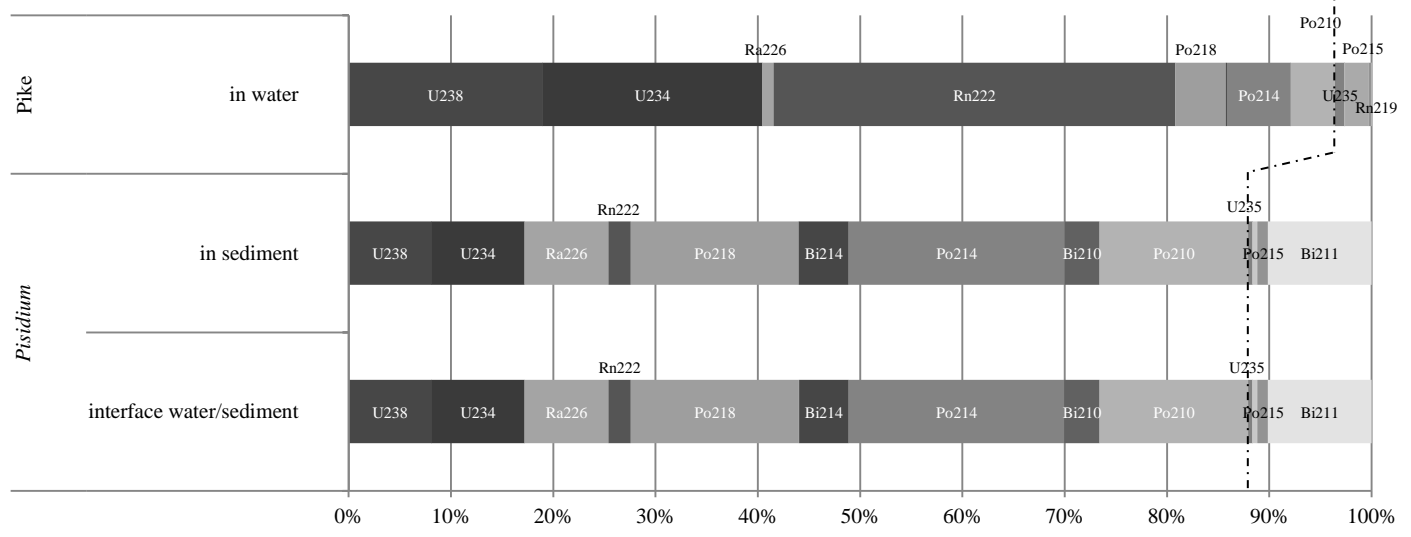
Decay chains at equilibrium in water

Contribution of the U-235 decay chain



Decay chains at equilibrium in sediment

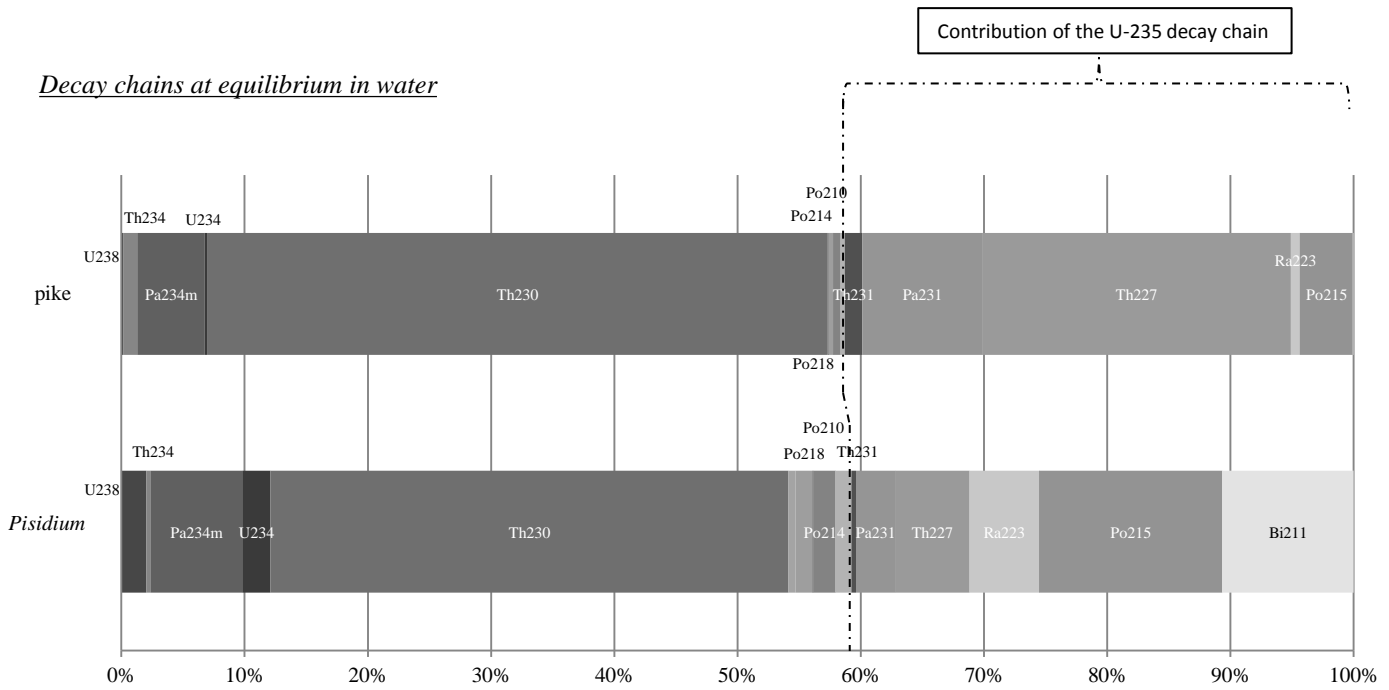
Contribution of the U-235 decay chain



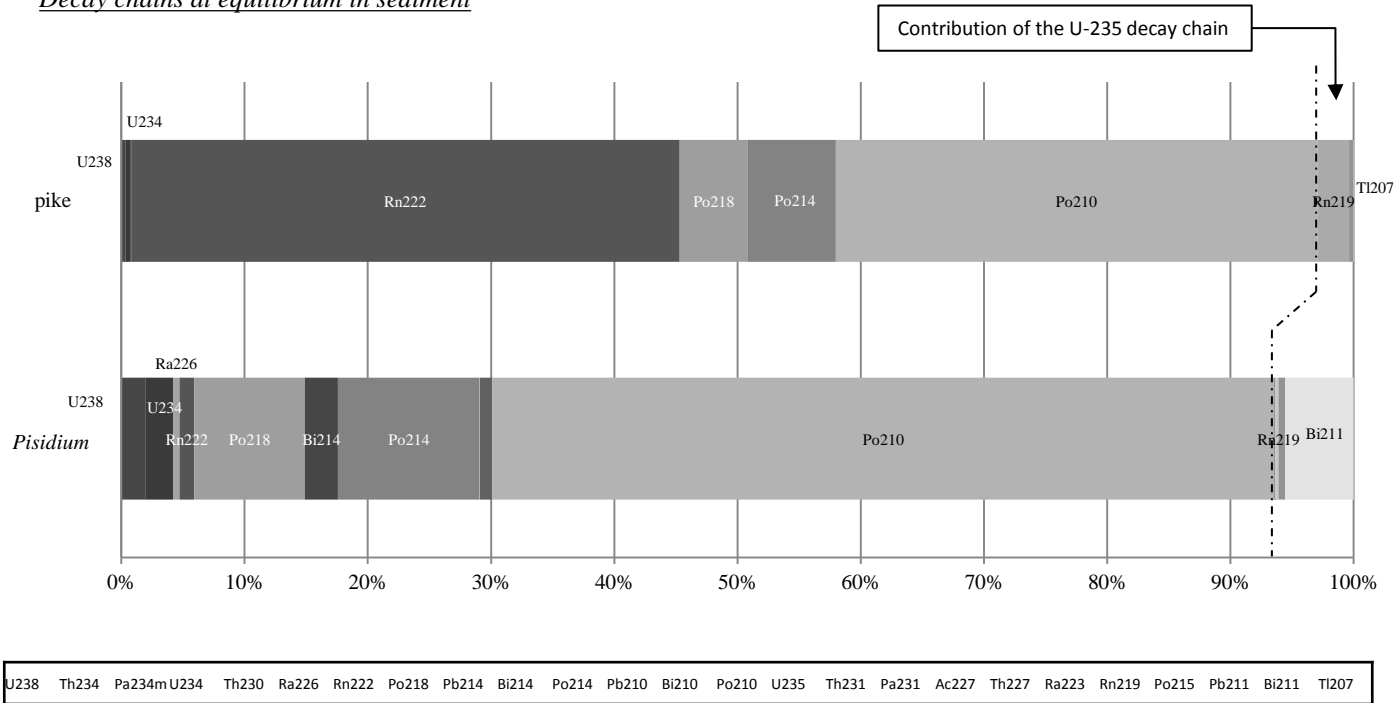
U238 Th234 Pa234m U234 Th230 Ra226 Rn222 Po218 Pb214 Bi214 Po214 Pb210 Bi210 Po210 U235 Th231 Pa231 Ac227 Th227 Ra223 Rn219 Po215 Pb211 Bi211 Tl207

Figure 6

Decay chains at equilibrium in water



Decay chains at equilibrium in sediment



U238 Th234 Pa234m U234 Th230 Ra226 Rn222 Po218 Pb214 Bi214 Po214 Pb210 Bi210 Po210 U235 Th231 Pa231 Ac227 Th227 Ra223 Rn219 Po215 Pb211 Bi211 Tl207

Figure 7

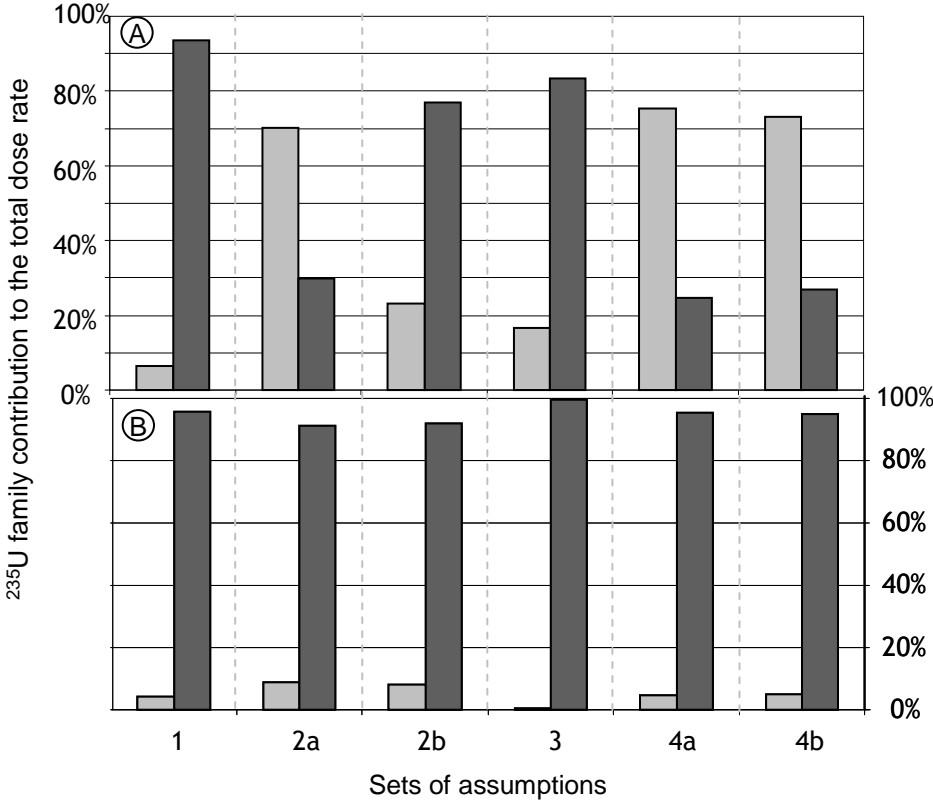
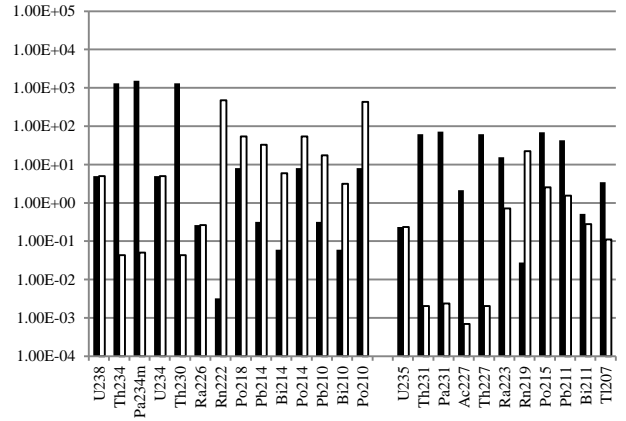
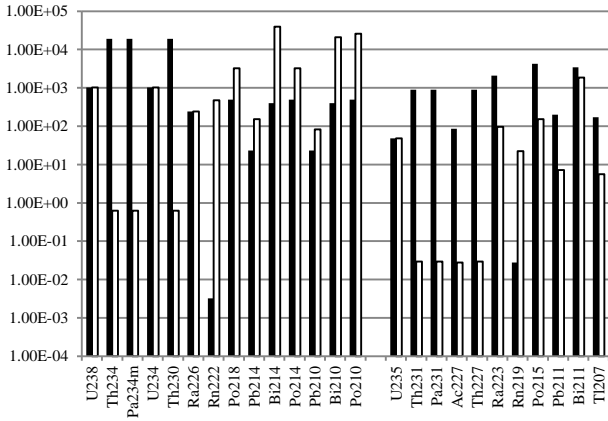
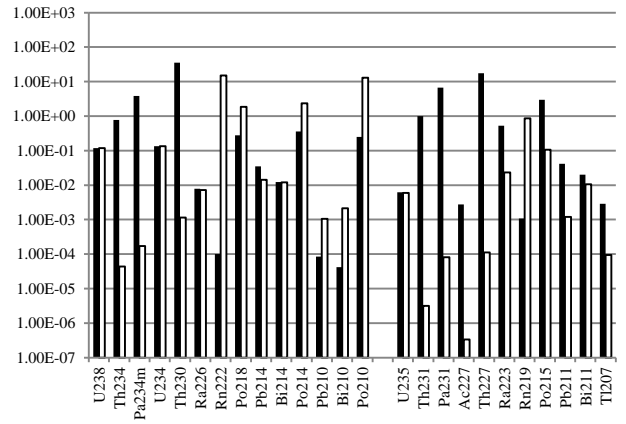
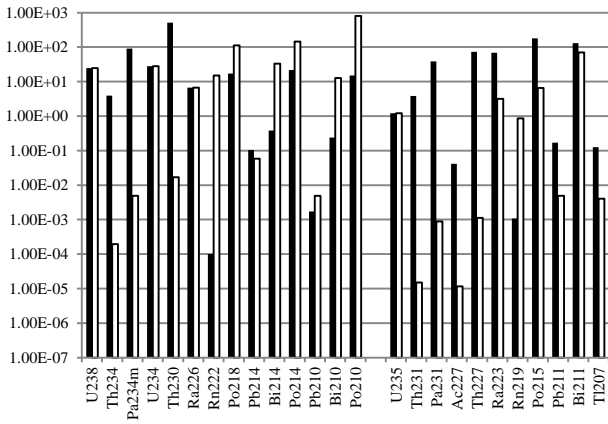


Figure 8

Activity concentration (Bq kg⁻¹ fm)



Total dose rate (μGy h⁻¹)



Pisidium

Pike

Should we ignore U-235 series contribution to dose?

Supplementary material

Table A1: weighted DCCs (1 to γ DCC, 3 to β DCC and 10 to α DCC) calculated with EDEN 2.2 (organism: $\mu\text{Gy h}^{-1}$ per $\text{Bq kg}^{-1} \text{wm}$; water: $\mu\text{Gy h}^{-1}$ per Bq L^{-1} ; sediment: $\mu\text{Gy h}^{-1}$ per $\text{Bq kg}^{-1} \text{wm}$)

Organism	<i>Pisidium</i>				Pike			
	internal		external		internal		external	
Location	In water		On sediment		In water		On sediment	
Source	organism	water	water	sediment*	organism	water	water	sediment
U238	2.41E-02	4.25E-05	4.29E-05	2.33E-08	2.41E-02	4.17E-06	4.10E-06	3.03E-09
Th234	8.00E-05	5.17E-06	2.67E-06	6.46E-07	8.13E-05	3.46E-06	2.50E-06	3.55E-07
Pa234m	1.08E-03	3.65E-04	3.35E-04	1.87E-05	1.38E-03	4.02E-05	3.75E-05	9.08E-07
U234	2.74E-02	6.04E-05	6.00E-05	3.91E-08	2.74E-02	5.83E-06	5.79E-06	5.25E-09
Th230	2.68E-02	5.71E-05	5.71E-05	5.83E-08	2.69E-02	5.63E-06	5.54E-06	1.82E-08
Ra226	2.75E-02	6.42E-05	6.21E-05	7.38E-07	2.75E-02	8.67E-06	7.83E-06	4.96E-07
Rn222	3.15E-02	8.75E-05	8.58E-05	9.38E-08	3.16E-02	8.42E-06	8.42E-06	3.46E-08
Po218	3.45E-02	1.10E-04	1.10E-04	7.58E-08	3.45E-02	1.06E-05	1.05E-05	1.42E-09
Pb214	3.65E-04	1.36E-04	7.79E-05	2.80E-05	4.08E-04	9.96E-05	5.75E-05	2.04E-05
Bi214	8.46E-04	9.08E-04	5.67E-04	1.93E-04	1.18E-03	6.33E-04	3.49E-04	1.07E-04
Po214	4.42E-02	2.24E-04	2.18E-04	2.37E-07	4.42E-02	2.10E-05	2.12E-05	8.75E-09
Pb210	6.00E-05	1.23E-06	6.46E-07	9.96E-08	6.00E-05	8.33E-07	6.08E-07	3.78E-08
Bi210	6.00E-04	7.54E-05	7.25E-05	1.67E-06	6.67E-04	7.21E-06	7.04E-06	1.76E-08
Po210	3.05E-02	8.00E-05	7.88E-05	4.58E-08	3.05E-02	7.58E-06	7.63E-06	1.13E-09
U235	2.53E-02	1.29E-04	8.88E-05	1.72E-05	2.54E-02	7.50E-05	5.54E-05	1.20E-05
Th231	1.58E-04	9.71E-06	5.67E-06	9.75E-07	1.60E-04	5.92E-06	4.38E-06	5.29E-07
Pa231	2.87E-02	8.29E-05	7.50E-05	3.45E-06	2.88E-02	1.90E-05	1.40E-05	2.50E-06
Ac227	4.09E-04	1.03E-06	9.71E-07	1.65E-08	4.10E-04	1.74E-07	1.52E-07	9.83E-09
Th227	3.40E-02	1.57E-04	1.31E-04	1.14E-05	3.41E-02	5.33E-05	3.75E-05	8.17E-06
Ra223	3.28E-02	1.64E-04	1.28E-04	1.35E-05	3.28E-02	6.33E-05	4.46E-05	9.25E-06
Rn219	3.88E-02	1.81E-04	1.65E-04	6.46E-06	3.89E-02	3.64E-05	2.70E-05	4.75E-06
Po215	4.25E-02	2.00E-04	1.96E-04	2.18E-07	4.25E-02	1.89E-05	1.90E-05	2.15E-08
Pb211	6.71E-04	1.33E-04	1.16E-04	1.07E-05	7.67E-04	3.86E-05	2.39E-05	5.38E-06
Bi211	3.76E-02	1.65E-04	1.52E-04	5.58E-06	3.78E-02	3.20E-05	2.36E-05	4.10E-06
Tl207	7.33E-04	1.25E-04	1.20E-04	3.80E-06	8.42E-04	1.29E-05	1.23E-05	1.80E-07

*multiplied by 2 for exposure in sediment (2Π exposure instead of Π exposure)