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URANIUM & DOSIMETRY IN AQUATIC ORGANISMS: WHICH ISOTOPES & PROGENY TO CONSIDER ?

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CONTEXT

Activity in water (Bq/L)
Long term and low [U]
Short term and high [U]

^{238}U & progeny
^{234}Th ^{238}U ^{234}U
^{235}U & progeny
$^{234}Pa^{m}$
^{235}U ^{231}Th

? ?

i) Measured by ICP-MS and gamma spectrometry

ii) Calculated from transfer factor(1) and Nuclides 2000 software

Activity of each element accumulated in organism (Bq/kg): U isotopes & progeny

⬇ Dose Conv. Coef. (DCC) calculations (EDEN(2))

Total Dose Rate (µGy/h) = Internal + external

Comparison between i) and ii)
Discussion about the use and refinement of tf value

⬇

Predicted-No-Effect-Dose Rate (PNEDR) (µGy/h)

Assessment of U radiological ecotoxicity.

EXPERIMENTS

Breeded in synthetic water to monitor chemical speciation of uranium (conductivity, pH, T°C et O_2% regulation).

Experimental design

Source isotopic composition (mass %)

^{238}U	99.65
^{235}U	0.33
^{236}U	0.010
^{234}U	0.0019

RNs Transfer factor for whole body (Bq/kg per Bq/L) for crustaceans(1)

U	500
Th	100
Pa	30

➔ **LONG TERM-LOW [U]** : 30d / 11µg/L <--> **0.18 Bq Utot. /L**

➔ **SHORT TERM-HIGH [U]** : 1d and 4d / 540µg/L <--> **9.25 Bq Utot. /L**

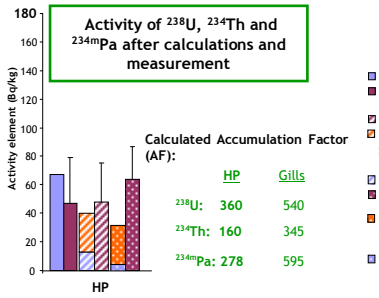
- Hepatopancreas (HP) and gills were dissected out at each sampling time and then were mineralized. Other organs were analyzed but γ-spectrometry results were <LD.
- γ-spectrometry analyses were performed on Ge radiation detector at the Modane Underground Laboratory, which offers a low background environment to measure low levels of ^{234}Th, ^{234m}Pa, ^{231}Th.
- ^{238}U and ^{235}U were measured by ICP-QMS (Agilent 7500 Cx)
- EDEN calculations of Dose Conversion Coefficient(2) (DCC in (Gy/d per Bq/kg)) for all the radionuclides and their progeny, to HP from a/HP, b/water, c/carapace and d/internal medium (see details in "Parameters for dose calculation to HP").
- Dose rate calculations for each exposure condition

EXPERIMENTAL RESULTS

LONG TERM - LOW [U]: « CHRONICAL SITUATION »

➡ 11µg/L U in water <--> **0.133 Bq /L ^{238}U + 0.133 Bq /L ^{234}Th + 0.133 Bq /L ^{234m}Pa** (measured by ICP-MS and γ-spectrometry)

Activity of ^{238}U, ^{234}Th and ^{234m}Pa after calculations and measurement



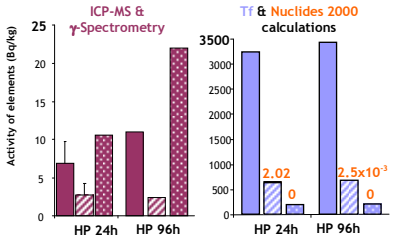
Calculated Accumulation Factor (AF):

	HP	Gills
^{238}U:	360	540
^{234}Th:	160	345
^{234m}Pa:	278	595

SHORT TERM-HIGH [U]: « INCIDENTAL SITUATION »

➡ 500µg/L U in water <--> **6.7 Bq /L ^{238}U + 6.7 Bq /L ^{234}Th + 6.7 Bq /L ^{234m}Pa** (measured by ICP-MS and γ-spectrometry)

Activity of ^{238}U, ^{234}Th and ^{234m}Pa after calculations and measurement



Calculated Accumulation Factor (AF):

	HP	Gills
^{238}U:	1.4	118
^{234}Th:	0.2	82
^{234m}Pa:	2.53	100

➔ Activity of progeny coming from parent accumulated (orange bars), calculated *via* Nuclides 2000(3) at 24, 48h or 30d, represents 22-70% of the total activity of the descendant considered.

➔ Except for " Gills-Chronical situation" (Data not shown, under-estimation of 6%), the assessment of ^{238}U activity in the organs *via* activity calculated with tf is hugely over estimated (40 - >1000%).

➔ Except for " Chronical situation" (under-estimation of 40%), the assessment of ^{234}Th activity in the organs *via* the sum of activity calculated with tf and activity calculated with Nuclides 2000 is over-estimated, above all for HP (up to +90%)

➔ Except for HP "Incidental situation" (over-estimation of 95%), the under-estimation of ^{234m}Pa activity in the organs via the sum of activity calculated with tf and activity calculated with Nuclides 2000 is quite high (120%) and **would required a better estimate of the tf of protactinium.**

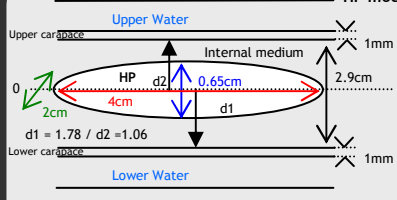
➔ By using Accumulation Factor (calculated after experiment at 24h and 96h with Th(natural)) instead of tf, the assessment of ^{234}Th activity in the organs via calculation is slightly better: over-estimation of roughly 60%. **Tf being calculated at the equilibrium, the latter result is not surprising**

➔ **However, these experiments enable the calculation of ^{234}Th and ^{234m}Pa Accumulation Factor (AF) with 2 exposure conditions, in both HP and Gills. These AF should be combined to determine "new tf", which is representative of whole body and more operational.**

PARAMETERS FOR DOSE CALCULATION TO HP

➡ Different potential contributors (water, carapace, HP,...) for dose rate calculation to HP

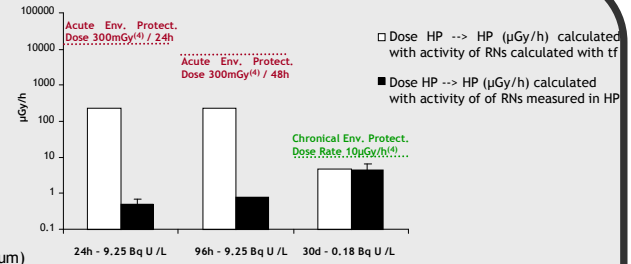
HP modelling for EDEN calculations of:



- DCC HP
- DCC Internal medium
- DCC Carapace
- DCC Water

--> HP (µGy/h)

- Use of radiation weighted DCC (α: 10; β: 3; γ: 1)



➡ DCC (HP --> HP) are much higher than DCC of other contributors (Water, carapace, Internal Medium)

➔ **For all the exposures, uranium isotopes are the major contributors to the dose rate (95-99%).**

➔ In all cases, **dose rate to HP is hugely due to the accumulation of RNs in HP itself.** According to the theoretical contribution of ^{234m}Pa to the total dose rate, the refinement of its AF in different organs is necessary.

➔ Experimental dose rate levels increase with the exposure duration from 0.5 up to 4.7 µGy/h but always remains < environment protection threshold of 10.

➔ It's important to notice that in case of "incidental situation", tf calculated dose rates are hugely over-estimated but << acute environment protection.

CONCLUSION

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➔ In these experiments, target organs of ^{234}Th and ^{234m}Pa were found to be the same as for ^{238}U (others organs<<LD), *i.e.* HP and Gills. However biokinetics were not studied. The low detection limit of Modane facilities enables the calculation of ^{234}Th and ^{234m}Pa Accumulation Factor (AF) with 2 exposure conditions, in HP and Gills.

➔ Assessment of different contributions of RNs to the global radiotoxicity needs to take DCC into account and not only compares activities of RNs.

➔ Transfer factors (tf), calculated at the equilibrium, for the whole body are not convenient to use for all exposure conditions and notably for exposure with alpha-emitters for which the maximum dose rate is due to the accumulation of RNs in the organ considered. Accumulation factors (AF) calculated for each organ are more representative. To study the effect/dose rate relationship AF should even be calculated for different organites such as mitochondria or nucleus.

➔ Calculations validated for low level of exposure are not so easily extrapolated to high level of exposure in case of, for instance, incidental pollution: the determination of Accumulation Factors in the maximum of exposure conditions and for the maximum of organs is necessary

REFERENCES: (1)Environment Agency Habitats regulations, **(2003)** p. 58; R&D128 in **ERICA DB**; (2) Beaugelin-Seiller, K., Jasserand, F., Garnier-Laplace, J., Gariel, J.C. Modeling radiological dose in non-human species: Principles, computerization, and application *Health Physics* 90 (5) (**2006**), pp. 485-493; (3)J. Magill, Nuclides 2000: An Electronic Chart of the Nuclides, European Commission (**1999**), J.R.C ITU, Karlsruhe, Germany; (4) Garnier-Laplace J., and Gilbin R. (Eds) ERICA **(2006)** *EC, 6th Framework* Deliverable D5. Contract no. FI6R-CT-2003-508847