Forensic and Scientific Services

Groundwater influence on uptake of radium, polonium-210 and lead-210 in freshwater mussels (*Velesunio ambiguus*)

Peter Medley[a] and Drew Watson [a]

[a]Queensland Health, Radiation and Nuclear Sciences, Brisbane, Queensland, Australia.

peter.medley@health.qld.gov.au

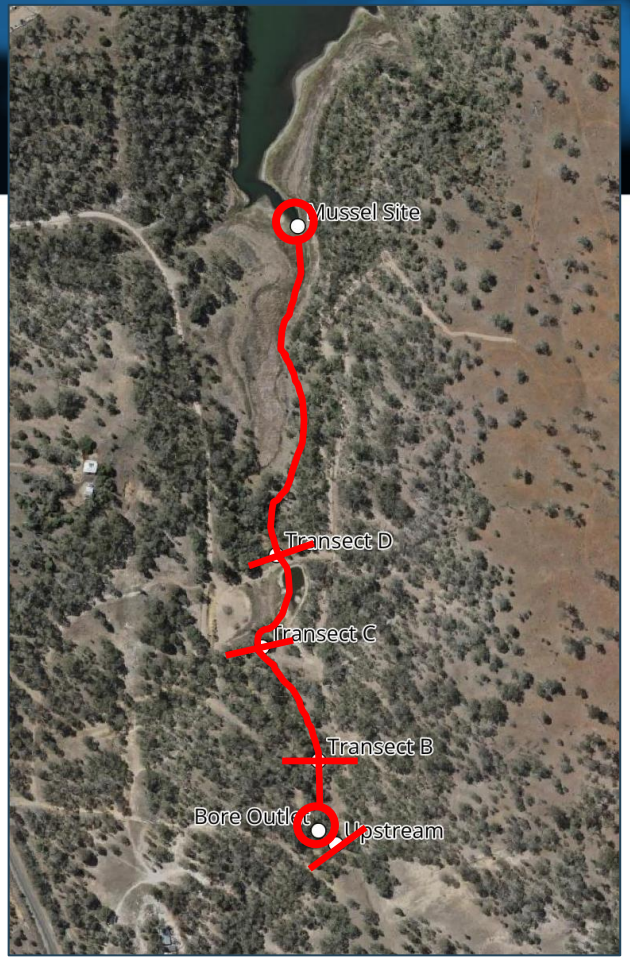
Research project assessing groundwater impacted site

- Southern Queensland
- Groundwater used to augment dam level during a period of drought
- Radium detected in unfiltered groundwater and in settling pond sediment
- Sampling of water, sediment and freshwater mussels

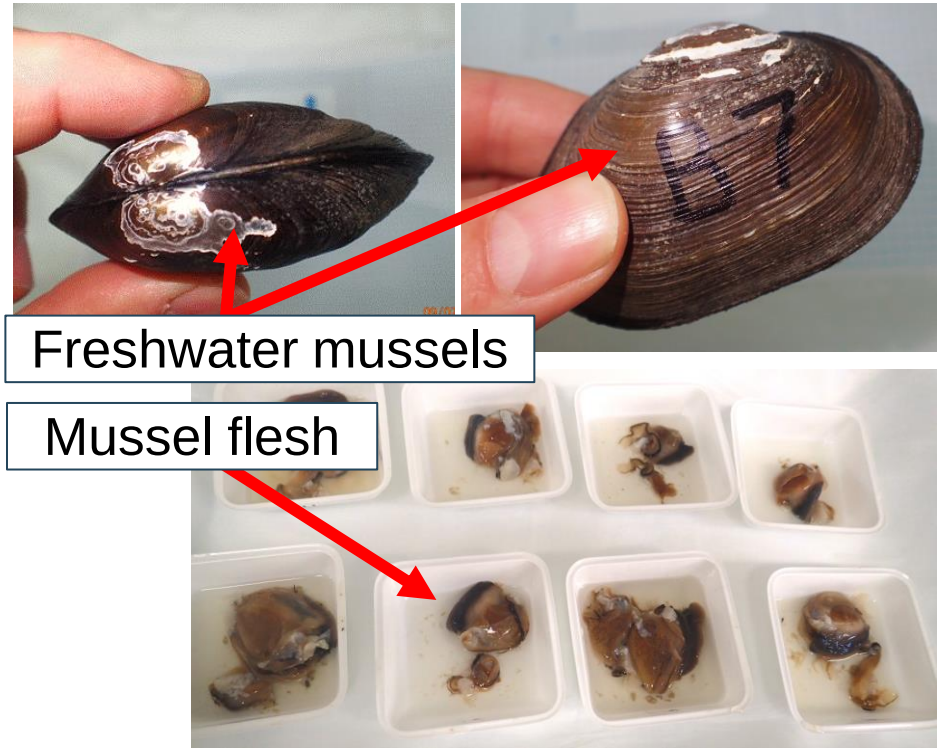
- Mapping radium in the creek bed sediment impacted by the groundwater discharge to be discussed on later presentation

Study site

- Sampling of water, sediment and freshwater mussels:
  - From groundwater outlet (Water)
  - Along intermittent creek line of groundwater flow (Water)
  - At flow point into the dam (Water, sediment and freshwater mussels)
  - At unimpacted ‘background’ site in the dam (Water, sediment and freshwater mussels)
- Assess impacts - Radiological dose to people
  - *Velesunio ambiguus*



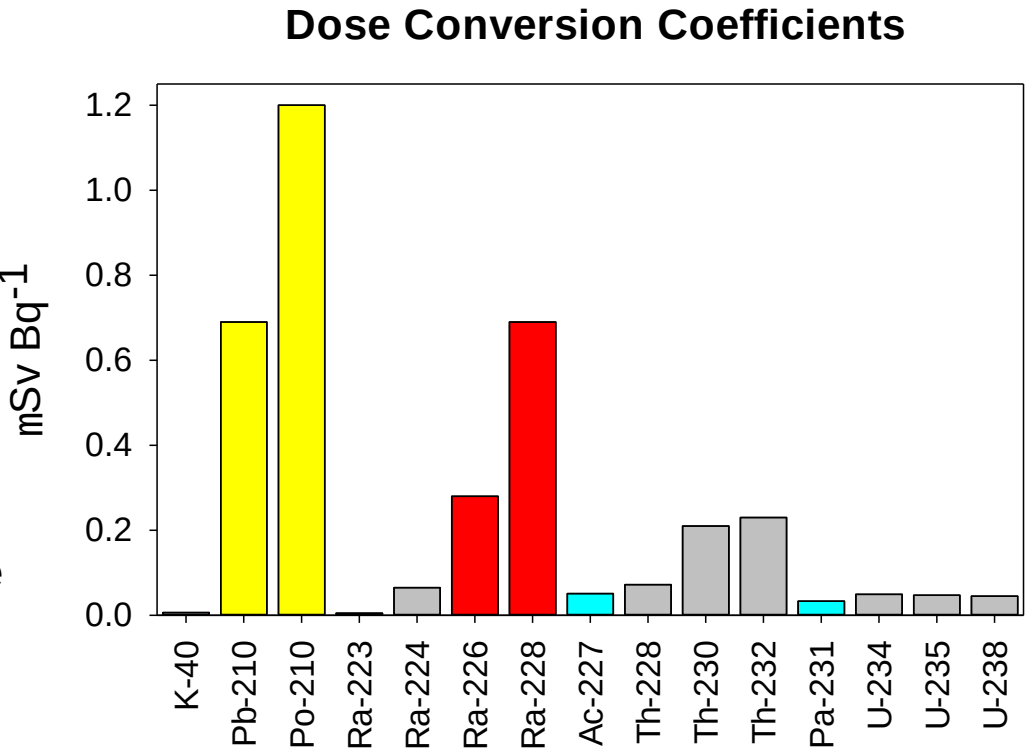
Sample preparation and analysis



- Mussels purged flesh removed and dried for analysis:
  - ^{210}Pb by LSC, ^{210}Po and ^{226}Ra by alpha spec., ^{228}Ra by HPGe gamma spec., ICP for metals
- Water samples → quite clean water, acidified in bottle (unfiltered)
- Sediment samples dried and measured by HPGe gamma spec.

Assessing radiological dose to people and to the environment

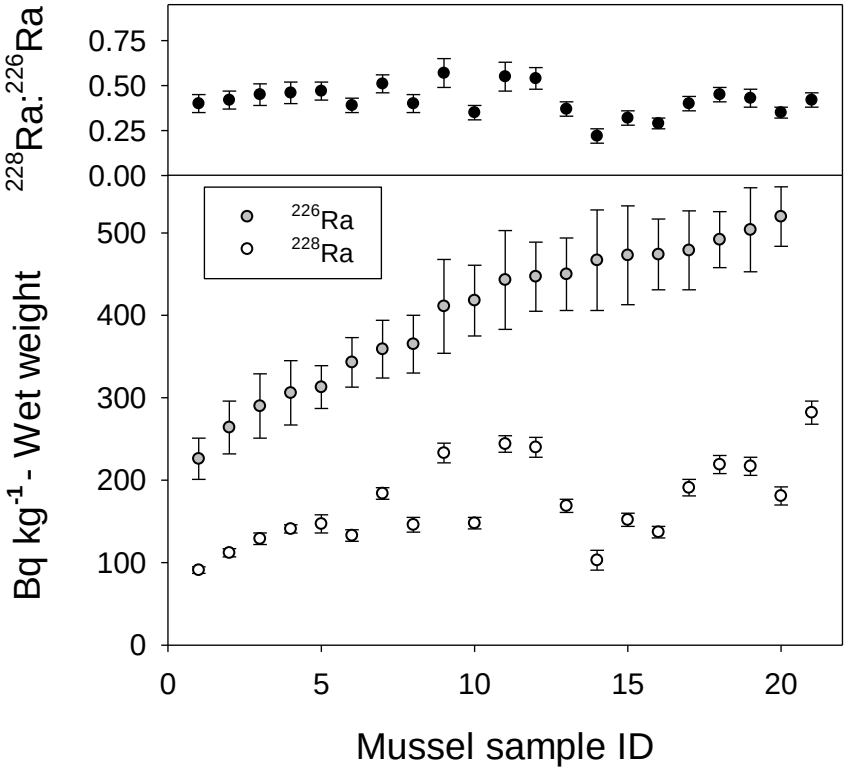
- Ingestion of bush foods can lead to radiological dose to members of the public
  - Dose conversion coefficients suggest relative importance is:
    $^{210}Po > {}^{210}Pb$ & $^{228}Ra > {}^{226}Ra$
- Freshwater mussels may strongly bioaccumulate Ra
  - Ra behaves as an analogue to Ca
  - Ca is an essential element and uptake is regulated
  - Ra is not removed after uptake



^{228}Ra and ^{226}Ra activity concentrations in mussels

- Impacted site in graph → Much higher than background site:
  - ^{226}Ra: 4.6–5.4 Bq kg^{-1}
  - ^{228}Ra: <6.1 Bq kg^{-1}

  } ^{228}Ra:^{226}Ra <1
  - No correlation with age (^{226}Ra R^2 = 0.12)
  - Moderate correlation of ^{228}Ra and ^{226}Ra R^2 = 0.48, slightly better if ^{228}Ra decay is accounted for
  - ^{228}Ra:^{226}Ra ratios 0.42±0.18 (σ=2)



Radiological dose from ingestion and to the mussels

- ^{210}Po ~10 Bq kg^{-1}
- ^{210}Pb <2 Bq kg^{-1}
  - No difference in background and impacted site
- Dose estimate:
  - ^{226}Ra = ^{228}Ra
  - Combined >95%
- Actual consumption by the general public is low

Radionuclide	Dose 10 y (Child; mSv $year^{-1}$)	Dose Adult (mSv $year^{-1}$)
^{210}Pb	0.003 ± 0.003	0.001 ± 0.001
^{210}Po	0.02 ± 0.05	0.01 ± 0.02
^{226}Ra	0.3 ± 0.2	0.12 ± 0.06
^{228}Ra	0.3 ± 0.2	0.12 ± 0.07
Total	0.7 ± 0.4 (1.1)	0.25 ± 0.15 (0.4)

Note: Annual dose from assumed consumption of 1 kg per year fresh weight.

Radiological dose from ingestion and to the mussels

- ^{210}Po ~10 Bq kg^{-1}
- ^{210}Pb <2 Bq kg^{-1}
  - No difference in background and impacted site
- Dose estimate:
  - ^{226}Ra = ^{228}Ra
  - Combined >95%
- Actual consumption by the general public is low → Potentially high doses from unexpected sources or from traditional diet.

Radionuclide	Dose 10 y (Child; mSv year^{-1})	Dose Adult (mSv year^{-1})
^{210}Pb	0.003 ± 0.003	0.001 ± 0.001
^{210}Po	0.02 ± 0.05	0.01 ± 0.02
^{226}Ra	0.3 ± 0.2	0.12 ± 0.06
^{228}Ra	0.3 ± 0.2	0.12 ± 0.07
Total	**0.7 ± 0.4 (1.1)**	0.25 ± 0.15 (0.4)

Note: Annual dose from assumed consumption of 1 kg per year fresh weight.

^{228}Ra:^{226}Ra activity ratios in mussels, sediment and water

- Impacted site
  - ^{226}Ra in waters 0.3–0.7 Bq L^{-1}
  - ^{228}Ra in waters 0.6–1.5 Bq L^{-1}
  - Decreasing with distance from bore
- Background
  - ^{226}Ra <0.005 Bq L^{-1}, ^{228}Ra <0.05 Bq L^{-1}
- Sediments → Elevated at impacted sites (see Drew's talk tomorrow)
- ^{226}Ra:^{228}Ra ratios **very** different in mussels compared both water and sediment → Unknown why

Sampling sites (impacted)	^{228}Ra:^{226}Ra
Sediment Mussel site	3.6 ± 0.3
Sediments (transects)	1.7 ± 0.5
Sediment (upstream)	1.8 ± 0.1
Waters (average all sites)	1.9 ± 0.2
Mussels	0.42 ± 0.18

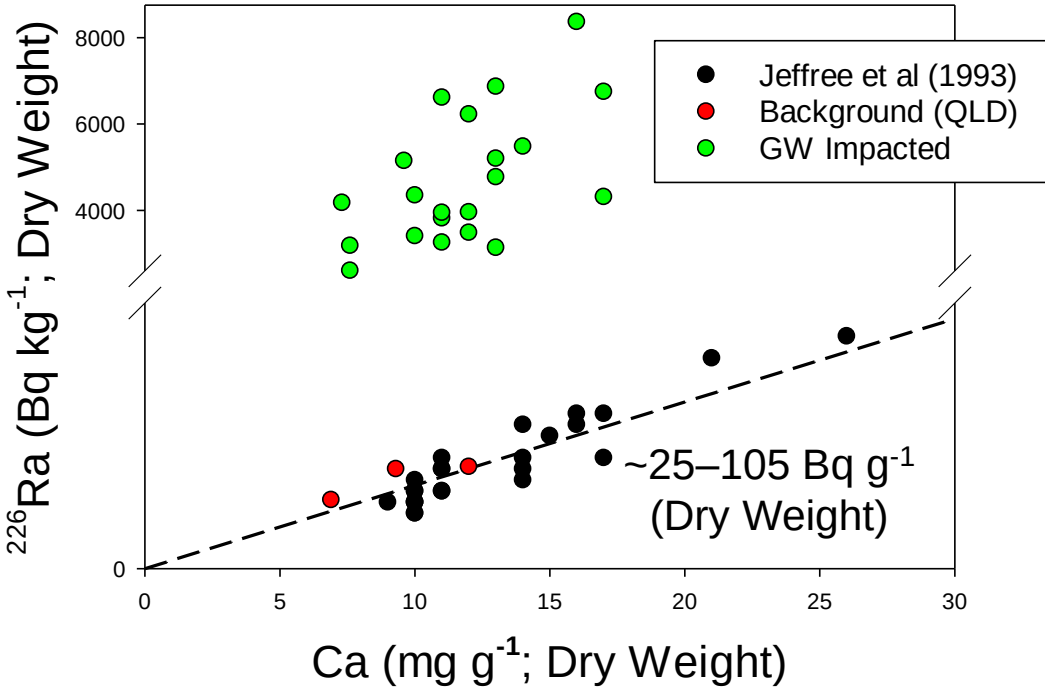
Velesunio ambiguus: Biomonitoring tool

- Jeffree et al (1993) suggested *Velesunio ambiguus* as a useful biomonitoring tool
  - Ca tissue concentration a better predictor or ^{226}Ra concentration than size or age.
  - Removes variability in Ra tissue concentrations as issue in biomonitoring
- Our data from the background site in good agreement with Jeffree et al (1993)
- Impacted site shows very clear contrast


Comparative Accumulation of Alkaline-earth Metals by Two Freshwater Mussel Species from the Nepean River, Australia: Consistencies and a Resolved Paradox

Ross A. Jeffree, Scott J. Markich and Paul L. Brown

Aust. J. Mar. Freshwater Res., 1993, **44**, 609–34

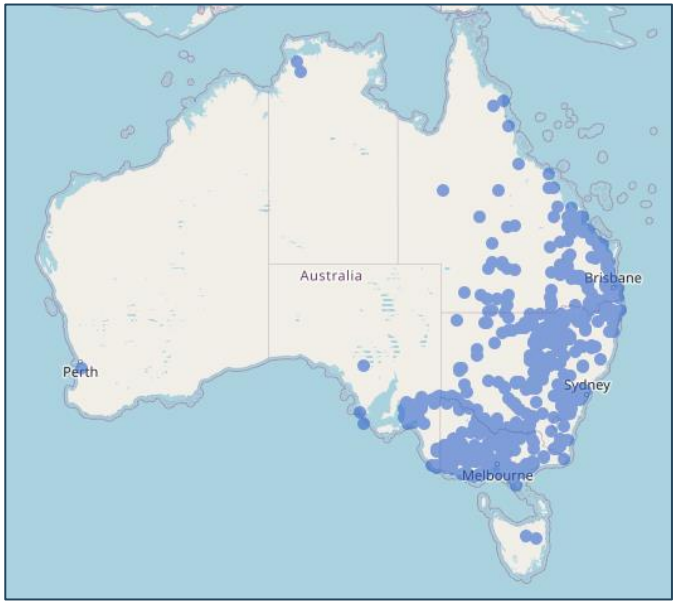


Mining wastewater discharge regulations: What about Radium?

- International example: Metal and Diamond Mining Effluent Regulations in Canada
  - ^{226}Ra a "*Prescribed Deleterious Substance*"
- Australian regulations
  - Mentions uranium
  - No specific mention of radium → Typically only considered for uranium mining
- Should Australia have regulations/limits for ^{226}Ra (and ^{228}Ra) discharge in mining, mineral processing or other industrial discharge waters?

Retrospective assessment: Need for ^{226}Ra discharge limits

- Measure mussels
  - Upstream/downstream of discharge points
  - Mining, other industrial activities
- *Velesunio ambiguus*
  - Distribution is very broad
  - Suitable for any current operations
    - Can be retrospective (to the limit of mussel age)
- Relatively low-cost, non-prescriptive way to determine need for ^{226}Ra discharge limits, monitoring or regulation



Distribution of *Velesunio ambiguus*

(Atlas of Living Australia (n=1065))

Conclusions

- ^{226}Ra and ^{228}Ra: High uptake into Freshwater mussel *Velesunio ambiguous*
  - Low uptake of ^{210}Pb and ^{210}Po
- Ingestion doses high at impacted site → >1 mSv year^{-1} for a child (1 kg)
  - >95% of dose from ^{226}Ra and ^{228}Ra (approx. equal contribution)
- ^{228}Ra:^{226}Ra ratios at impacted site different in mussels compared to sediment/water
  - Suggests potential different environmental origin and behaviour of each isotope
- Freshwater mussel *Velesunio ambiguus* represents a useful biomonitoring tool
  - For industrial and development activities
  - For assessment of need for ^{226}Ra discharge limits for mining and mineral processing operations, and other industrial activities