

\*Accumulation and retention of  $^{241}\text{Am}$  and  $^{237}\text{Pu}$  in the mussel Mytilus edulis

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#### Abstract

A series of laboratory experiments was conducted to explore the biokinetics of the transuranic elements Am and Pu in the common mussel, Mytilus edulis. Mussels were capable of accumulating these radionuclides (CF's  $\sim 10^2$ ) by ingestion and assimilation of labelled phytoplankton and by direct adsorption from the dissolved state. The tissue distribution of Am and Pu differed depending on the source term, with shell being the prime site of deposition ( $\sim 90\%$ ) of adsorbed radionuclide and visceral mass the main site ( $\sim 40\%$  to  $70\%$ ) for ingested radionuclide. The radionuclide content of shell was a direct function of ambient radionuclide concentration ( $P < .001$ ), supporting the use of mussels as quantitative indicators of water contamination. Am was generally 1.5 to 2 times more reactive for mussels than Pu, regardless of the source term. Assimilation of both radionuclides ranged from 1 to 15%. Retention half-times of these transuranics in mussels were on the order of  $10^2$  days in the slowest exchanging pools.

#### Résumé

Plusieurs expériences furent conduites en laboratoire afin d'étudier les biocinétiques d'éléments transuraniens Am and Pu dans la moule commune Mytilus edulis. Les moules furent capables d'accumuler ces radionucléides (CF's  $\sim 10^2$ ) par ingestion et assimilation de diatomées radioactives et par adsorption directe de la fraction dissoute de ces deux éléments. La distribution dans les tissus de l'Am et du Pu était différente et liée à leur mode d'accumulation; la coquille était le lieu principal de déposition ( $\sim 90\%$ ) des radionucléides adsorbés, et la masse viscérale ( $\sim 40\%$  à  $70\%$ ) celui des radionucléides ingérés. La quantité de radionucléides sur la coquille était directement proportionnelle à leur concentration ambiante ( $P < .001$ ); la coquille pouvant être utilisée comme indicateur quantitatif des contaminations de l'eau. L'Am était généralement 1.5 à 2 fois plus réactif pour les moules que le Pu, ceci sans tenir compte de leur origine. L'assimilation des deux radionucléides variait de 1 à 15%. Les demi-vies biologiques les plus longues des deux transuraniens dans les moules étaient de l'ordre de  $10^2$  jours.

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\*To be published in extenso elsewhere.

Table 1. Mytilus edulis. Half-lives ( $T_{b1/2}$ ) and radionuclide content (as % of the initial body burden) in compartments identified by mathematical treatment of the loss curves.

| Element           | Exposure |      | (A)                  |                     | (B)                  |                     | (C)                   |                     | $T_{b1/2}$<br>(hours) |
|-------------------|----------|------|----------------------|---------------------|----------------------|---------------------|-----------------------|---------------------|-----------------------|
|                   |          |      | $T_{b1/2}$<br>(days) | % of body<br>burden | $T_{b1/2}$<br>(days) | % of body<br>burden | $T_{b1/2}$<br>(hours) | % of body<br>burden |                       |
| Am                | water    | 5d   | 96                   | 63                  | 8.6                  | 14                  | 43                    | 23                  |                       |
| Am                | diatoms  | 5d   | 55                   | 37                  | 5.2                  | 20                  | 12                    | 43                  |                       |
| Am                | diatoms  | 1/2h | 58                   | 2.7                 | 4.5                  | 14                  | 29                    | 28                  | 2                     |
| Am ]              | diatoms  | 5d   | 108                  | 23                  | 10.2                 | 47                  | 25                    | 30                  |                       |
| Pu ]              |          |      | 73                   | 23                  | 14.5                 | 49                  | 17                    | 28                  |                       |
| Am ]              | water    | 5d   | 97                   | 61                  | 12.0                 | 12                  | 72                    | 27                  |                       |
| Pu ]              |          |      | 82                   | 45                  | 7.0                  | 55                  |                       |                     |                       |
| Am <sup>a</sup> ] | water    | 5d   | 113                  | 66                  | 7.4                  | 20                  | 43                    | 14                  |                       |
| Pu <sup>a</sup> ] |          |      | 85                   | 51                  | 9.5                  | 32                  | 31                    | 17                  |                       |

a : loss from dead shells

The International Mussel Watch Program has sought to use mussels as biological indicators of the presence of contaminants in marine waters. The many virtues of this approach as well as its disadvantages have been well-outlined (Goldberg *et al.*, 1978; Phillips, 1980). Among the contaminants of interest are the transuranic elements, most particularly plutonium and americium, and already hundreds of analyses of mussel tissue for these artificial radionuclides have been conducted (Goldberg *et al.*, 1983). However, interpretation of the mussel analyses from the field obtained to date has been hampered by the lack of source term data for the geographic regions of interest. Moreover, there is a paucity of experimental information regarding the rates and routes of transuranic incorporation into mussels and subsequent retention times of these contaminants in the various mussel tissues.

To this end, we have conducted a series of laboratory experiments using radiotracer methodology to investigate transuranic accumulation and retention processes in *Mytilus edulis* (Bjerregaard *et al.*, (in press)). We have concluded that:

- (1) Mussels can accumulate  $^{241}\text{Am}$  and  $^{237}\text{Pu}$  from both contaminated food and from the dissolved state. The relative importance of each is a function of the partitioning of the radionuclides in the water column, which in turn directly reflects the particulate load in the water. In most natural waters, the dissolved state tends to dominate. In the Mediterranean, generally  $\leq 10\%$  of the Pu and Am are associated with particles  $> 0.45 \mu\text{m}$ .
- (2)  $^{241}\text{Am}$  and  $^{237}\text{Pu}$  dissolved in filtered seawater adsorbs principally to mussel shells. Radionuclide concentrations in shell, whole mussels, and "soft parts" reflect ambient dissolved concentrations, with shell giving the most regular response. Pu:  $y = .989x + 0.603$ ,  $r^2 = .999$  ( $P < .001$ ); Am:  $y = .982x + 0.720$ ,  $r^2 = .982$  ( $P < .001$ ); where  $y = \log$  molar concentration of radionuclide in shell and  $x = \log$  molar concentration of radionuclide in solution. This finding supports the use of mussels as quantitative bio-indicators of transuranic concentrations in seawater and further suggests that shells may be the most appropriate material for analyses of metals.
- (3) Concentration factors of  $^{241}\text{Am}$  and  $^{237}\text{Pu}$  in mussels generally ranged from 10 to  $10^2$ , regardless of the source term. Am was found to be about 1.5 to 2 times more reactive than Pu for mussels, consistent with the comparatively high Am/Pu ratios observed in field collected mussels.
- (4) When mussels fed on labelled diatoms, nearly all the ingested Am or Pu was initially associated with the visceral mass and then released via fecal pellets. Assimilation of ingested radionuclide ranged from 1 to 15%, depending upon experimental conditions. In particular, assimilation efficiency decreased when the diatom cell density increased from  $10^4 \text{ ml}^{-1}$  to  $5 \times 10^4 \text{ ml}^{-1}$ . The retention half-times of Am and Pu in fecal pellets were 2 - 3 weeks, increasing inversely with temperature ( $5^\circ\text{C}$  to  $22^\circ\text{C}$ ) and salinity (19‰ to 38‰).
- (5) After exposure to Am and Pu through food or water, mussels were placed into unlabelled flowing seawater and allowed to depurate. A multi-compartmental distribution was evident from the loss curves. In the

slowest exchanging fraction (A), retention half-times were of the order of  $10^2$  days, with Am showing somewhat longer half-times than Pu, regardless of the uptake mechanism (Table 1).

#### REFERENCES

- BJERREGAARD, P., S. TOPCUOGLU, N.S. FISHER and S.W. FOWLER. Biokinetics of americium and plutonium in the mussel Mytilus edulis L. Mar. Ecol. Prog. Ser. (in press).
- GOLDBERG, E.D. V.T. BOWEN, J.W. FARRINGTON, G.R. HARVEY, J.H. MARTIN, P.L. PARKER, R.W. RISEBROUGH, W. ROBERTSON, E. SCHNEIDER and E. GAMBLE. The mussel watch. Environ. Conserv. 5: 101-125 (1978).
- GOLDBERG, E.D., M. KOIDE, V. HODGE, A.R. FLEGAL and J.H. MARTIN. U.S. mussel watch: 1977-1978 results on trace metals and radionuclides. Est. Coastal Shelf Sci. 16: 69 -93 (1983).
- PHILLIPS, D.J.H. Quantitative Aquatic Biological Indicators. Appl. Sci. Publ., London, 488 pp. (1980).

#### Discussion

A. BOLOGA: Have you any particular reason to carry out uptake experiments on mussels with  $^{241}\text{Am}$  and  $^{237}\text{Pu}$ ?

N. FISHER: These are radiotracers of two important contaminants ( $^{241}\text{Am}$  and  $^{239}\text{Pu}$ ) about which we have relatively little comparative information. Knowledge of the biokinetics of these transuranics in these species is particularly important in the context of the proposed "International Mussel Watch Programme".