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RADIUM-226 IS ACCUMULATED IN CALCIUM GRANULES IN THE TISSUES OF THE FRESHWATER MUSSEL, *VELESUNIO ANGASI*: SUPPORT FOR A METABOLIC ANALOGUE HYPOTHESIS?

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Abstract—1. The distribution of ^{226}Ra, Ca, Ba and Mg in the tissues of the freshwater mussel, *Velesunio angasi*, was studied.

2. The concentration of ^{226}Ra is significantly ($P < 0.05$) and positively correlated with the concentrations of the alkaline earths, Ca, Mg and Ba, in dissected tissues of *V. angasi*.

3. Alpha track autoradiography, electron microprobe and X-ray analysis, and histochemical studies have shown that ^{226}Ra, Ca, Mg and Ba are co-located predominantly in granular deposits that are dispersed throughout the body of *V. angasi*.

4. The value of such a co-locational study in supporting hypotheses of analogous metabolic behaviour between the non-essential ^{226}Ra and Ca has to be assessed in relation to the functions attributed to calcium granules.

5. Granules may simultaneously accumulate ^{226}Ra and act as a store of exchangeable Ca.

INTRODUCTION

The freshwater mussel, *Velesunio angasi*, in the Magela Creek system of Alligator Rivers Uranium Province, Northern Territory, Australia, accumulates high concentrations of the non-essential, alpha-emitting radionuclide ^{226}Ra in its tissues (Davy and Conway, 1974).

The mechanism of uptake and accumulation of this non-essential element by organisms is generally interpreted as being a case of mistaken identity, i.e. the chemically similar ^{226}Ra is treated metabolically as an analogue of the essential alkaline earth metal, Ca (Whicker and Schultz, 1982). Such explanations can be supported by the results of competitive inhibition experiments in which increased concentrations of the essential element inhibit uptake of its analogue and by studies demonstrating the co-location of the essential element and the hypothesized analogue. Investigations of ^{226}Ra and Sr metabolism in animals and humans, where types of competitive inhibition studies have been conducted, have indicated that the accumulation of ^{226}Ra can be reduced by increased Ca concentrations (Marey *et al.*, 1967; Muth and Glöbel, 1983) and Sr accumulation is reduced by increased Ca and Mg concentrations (Wasserman *et al.*, 1957; Volf, 1959; Brungs, 1965; Marey *et al.*, 1967; Ophel and Judd, 1967; Roushdy *et al.*, 1979). Moreover, ^{226}Ra distribution studies have indicated that it is predominantly located in Ca-rich tissues (Van der Borght, 1963; Lucas *et al.*, 1979; Stover *et al.*, 1957; Van Dilla *et al.*, 1958; Norris *et al.*, 1955; Evans, 1974).

We have investigated the distribution of the essential elements Ca and Mg, and their potential analogue ^{226}Ra, in the tissues of *V. angasi* to elucidate the mechanism of ^{226}Ra accumulation in *V. angasi*.

MATERIALS AND METHODS

Sampling site

Samples of *V. angasi* were taken from Georgetown and Corndorl Billabongs, which lie in the Magela Creek system and are close to the ore body at the Ranger Uranium minesite.

Distribution of ^{226}Ra *and other elements in the dissected organ and tissue clusters of* V. angasi

To compare the general distribution of ^{226}Ra with the other alkaline earths Ca, Mg and also Ba, three mussels from Georgetown Billabong were individually dissected into outer gill flaps, inner gill flaps, palps, visceral mass, kidney and heart, mantle, foot and adductor muscle. These tissues were then oven-dried for 10 hr at 70°C to constant dry weight. Tissues were then dissolved in concentrated HNO_3 and the solution was diluted with distilled H_2O to 250 ml for ^{226}Ra analysis by the Lucas emanation method (Lucas, 1957); Ca, Mg and Ba concentrations were analysed by atomic absorption spectrophotometry (AAS). The concentrations of the non-alkaline earth elements Zn and Pb in the tissues from one mussel were also determined by AAS, for comparison with the distribution of alkaline earth metals.

The ^{226}Ra concentration was then compared with the concentration of each of the other elements, for individual mussels and for pooled data from the three mussels.

Alpha-track autoradiography

Alpha-track autoradiography was carried out using the solid state nuclear track detector, CR-39. Ellis and Jeffree (1982) showed that this detector was sensitive enough to locate small concentrations of ^{226}Ra and other alpha-emitting radionuclides in histological sections of *V. angasi*.

Transverse histological sections, nominally 5 μm thick, were cut from mussel flesh that had been preserved in 10% phosphate buffered formalin, dehydrated in ethanol and embedded in paraffin, and from frozen, fresh tissue, nominally 10 μm thick. Sections were mounted on glass histological slides which were then superimposed on strips of CR-39 (Pershore Mouldings Ltd, Worcs., U.K.) and bound

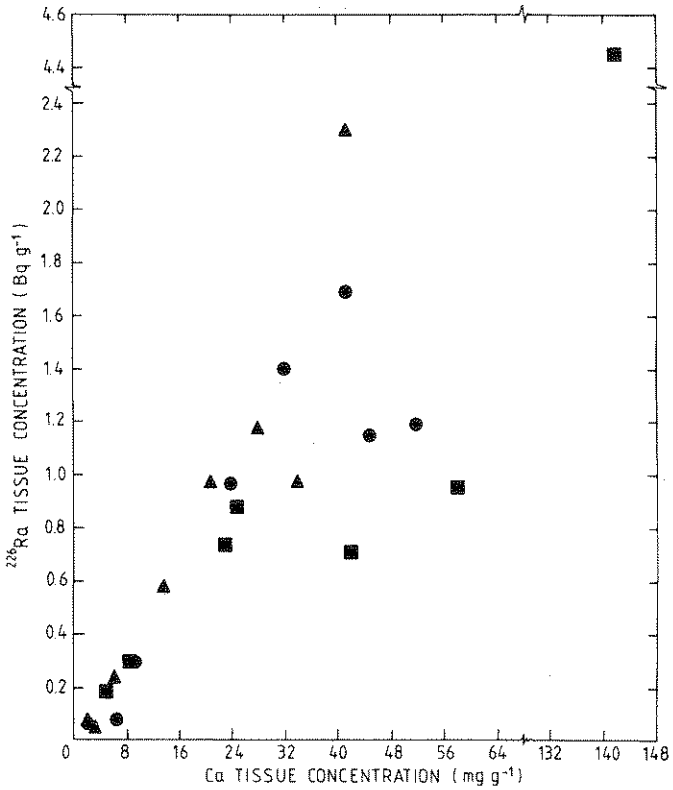

Fig. 1. ^{226}Ra concentration plotted against Ca concentration for the dissected tissues of *V. angasi* (●, mussel No. 1; ▲, mussel No. 2; ■, mussel No. 3). The concentrations for each dissected tissue are given in Table 1 and the correlation coefficients, *r*, are given in Table 2.

together with adhesive tape. Registration marks were scored in the CR-39 to enable accurate re-alignment with the glass slide.

Tissue/CR-39 composites were stored for 12 weeks. After separation, the histological section was stained with eosin and haemotoxylin, and the CR-39 film etched in 6.25 mol l^{-1} NaOH solution for 6 hr at 75°C. The histological section and CR-39 film were then re-aligned microscopically.

Scanning electron microscopy and electron microprobe analyses

Earlier alpha-track autoradiographic studies had identified granular deposits as the major repositories of ^{226}Ra, with smaller but significant amounts in the non-granular tissues of the gills and also in the renal tissue (Ellis and Jeffree, 1982). To determine whether the other alkaline-earth metals were also concentrated in the granules, electron microprobe analyses were conducted on *in situ* granules and extracted granules, and also on gill support structures that were extracted with the granular deposits. These analyses were performed on a Jeol JSM/US scanning electron microscope (SEM) and Trakor Northern X-ray energy dispersion system at 25 kV acceleration, using a 1 μm diameter circular probe.

(i) *Extracted granules.* The tissues of 16 mussels, each trimmed of the ventral foot, were bulked and digested in 10% pancreatin in phosphate buffer at pH 7.4 and 37°C. Four days of digestion followed by repeated washings in distilled water and sucrose solutions, and repeated centrifugations were needed to separate the granules from most other tissue components. The granules were allowed to dry out at air temperature (Ch'ng-Tan, 1968). Some of the dry granules were mounted on carbon blocks, using double-sided adhesive, for scanning electron microscopy and associated electron microprobe and X-ray analyses.

(ii) In situ *granular deposits.* The whole tissue was removed from the shell of one mussel while being flooded with a fixative composed of 3% glutaraldehyde, 2% formaldehyde in 0.1 mol l^{-1} cacodylate buffer at pH 7.4. Small pieces (1 mm^3) of tissue from the visceral mass and mantle were fixed for 24 hr at room temperature, washed in cacodylate buffer at pH 7.3. and postfixed in 1% OsO_4 for 2 hr. The tissues were washed in distilled H_2O and dehydrated through a graded series of aqueous to absolute ethanol solutions, and then infiltrated with and embedded in Spurr's Resin.

Sections of 10 μm thickness were cut from each block using a microtome, mounted on carbon and prepared for microprobe analysis, as outlined in subsection (i) above.

Histochemical studies

To determine the general histological distribution of Ca throughout all tissues, full transverse sections of paraffin embedded tissue were stained with alizarin red S (Lillie and Fullmer, 1976).

RESULTS

Distribution of ^{226}Ra and other elements in dissected organ and tissue clusters

The graphs of ^{226}Ra tissue concentration against Ca (Fig. 1), Ba (Fig. 2) and Mg (Fig. 3) tissue concentrations show that ^{226}Ra is similarly distributed among the tissues to Ca, Ba and Mg. The concentration of ^{226}Ra among the tissues is strongly ($P < 0.01$) and positively correlated with that of Ca, Ba and Mg when mussels are considered individually and also when the data for the three mussels are pooled (Table 2).

The concentrations of ^{226}Ra, Ca, Ba vary by more

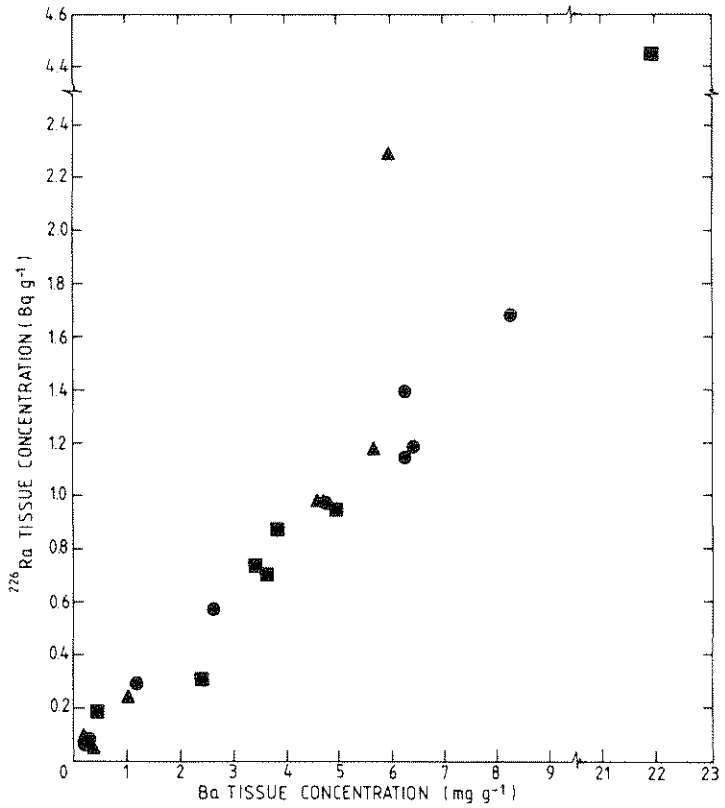

Fig. 2. ^{226}Ra concentration plotted against Ba concentration for the dissected tissues of *V. angasi* (●, mussel No. 1; ▲, mussel No. 2; ■, mussel No. 3). The concentrations for each dissected tissue are given in Table 1 and the correlation coefficients, *r*, are given in Table 2.

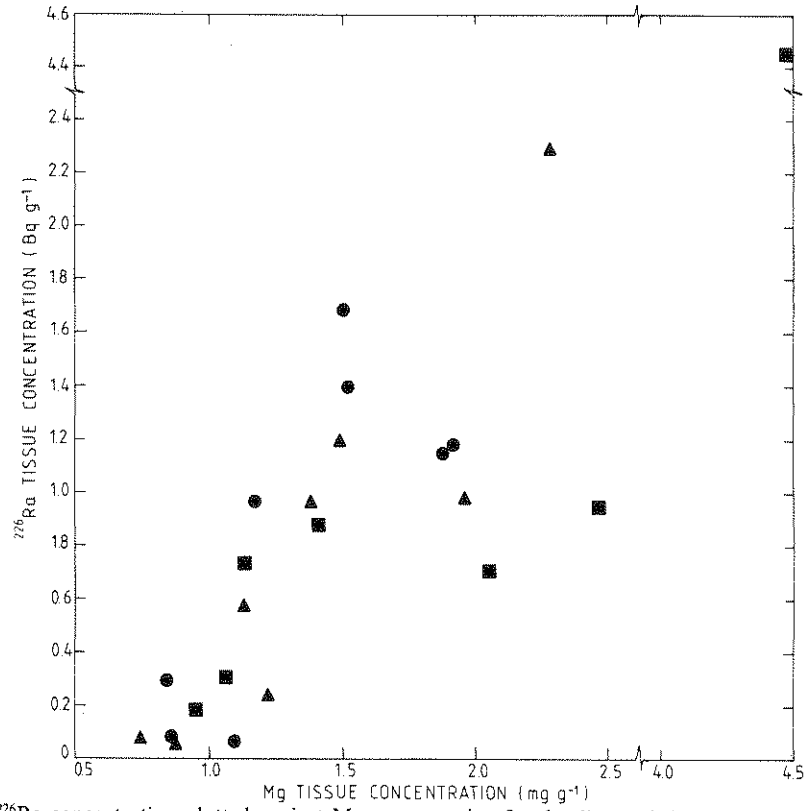

Fig. 3. ^{226}Ra concentration plotted against Mg concentration for the dissected tissues of *V. angasi* (●, mussel No. 1; ▲, mussel No. 2; ■, mussel No. 3). The concentrations for each dissected tissue are given in Table 1 and the correlation coefficients, *r*, are given in Table 2.

Table 1. The concentrations of ^{226}Ra, Ca, Mg, Ba, Pb and Zn for dissected organ and tissue clusters of *V. angasi*

Organ/Tissue Cluster	Tissue concentration (^{226}Ra	Ba	Ca	(Bq g^{-1} and mg g^{-1}) Mg	Pb	Zn
			MUSSEL NO. 1			
Outer gills	1.18	6.35	52.04	1.92	0.072	1.49
Inner gills	1.14	6.18	45.40	1.88	0.05	1.12
Palps	1.39	6.20	32.34	1.52	0.203	1.14
Visceral mass	1.68	8.19	41.45	1.50	0.052	0.38
Kidney and heart	0.29	1.20	9.37	0.84	0.084	0.314
Mantle	0.96	4.71	24.33	1.17	0.024	0.375
Foot	0.06	0.17	1.99	1.09	0.017	0.204
Adductor muscle	0.08	0.32	6.46	0.86	0.065	0.281
			MUSSEL NO. 2			
Outer gills	2.28	5.94	41.56	2.19		
Inner gills	0.96	4.71	34.12	1.96		
Palps	0.96	4.59	21.10	1.38		
Visceral mass	1.17	5.61	28.26	1.49		
Kidney and heart	0.07	0.18	2.21	0.74		
Mantle	0.57	2.57	13.82	1.13		
Foot	0.23	1.02	6.01	1.22		
Adductor muscle	0.05	0.29	3.06	0.87		
			MUSSEL NO. 3			
Outer gills	0.70	3.64	42.05	2.05		
Inner gills	0.94	4.93	57.85	2.47		
Palps	0.73	3.39	23.16	1.13		
Visceral mass	4.45	21.90	140.19	4.93		
Kidney and heart	0.18	0.47	5.21	0.95		
Mantle	0.87	3.80	24.93	1.41		
Foot	0.30	1.24	8.33	1.06		
Adductor muscle	0.09	-	-	-		

*Mussel No.	Shell length (mm)	Shell breadth (mm)
1	52.7	20.0
2	55.7	19.4
3	52.8	18.6

than an order of magnitude between the separate tissues. For Mg, however, concentrations in separate tissues vary by only a factor of 2 to 5 (Table 1).

The similarity in distribution between ^{226}Ra and Ca and Mg is to be expected, if ^{226}Ra is treated metabolically as an analogue of Ca and/or Mg. By the same deduction, these results indicate that the non-essential element Ba could also be regarded as a metabolic analogue of Ca and/or Mg. The tissue concentration of the non-alkaline earths Pb and Zn are not significantly ($P > 0.05$) correlated with the ^{226}Ra tissue concentration, thus indicating that the pattern of correlations for the alkaline earth metals does not necessarily exist for other elements.

Alpha track autoradiography

Figures 4 and 5 show the high concentrations of alpha tracks associated with the granular deposits and lower concentrations associated with other tissues in paraffin-embedded, histological sections of *V. angasi*. Figure 6 shows the lower but significant ($P < 0.05$) density of alpha tracks associated with an area of gill tissue (Ellis and Jeffree, 1982). Similar results were obtained from sections of frozen tissue. The alpha tracks produced by the radioactive decay of ^{226}Ra and its daughters (and other alpha-emitting radionuclides) occur over other tissues adjacent to granular deposits (Figs 4 and 5). However, this effect is related to their proximity to granular deposits, probably as a result of ^{222}Rn migration or alpha particles travelling at angles less than 90° from the plane of the histological section to the alpha track recorder, CR-39 (Ellis and Jeffree, 1982).

Scanning electron microscopy and electron microprobe analysis

(a) *Extracted granules.* The scanning electron micrograph of the extracted granules (Fig. 7a) shows that they are roughly spherical in shape having a diameter of about 1 μm. Electron microprobe analyses were performed on several single granules selected from those shown in Fig. 7(a). The spectrum (Fig. 7b (i)) indicates an abundance of Ca and also detected the other alkaline-earth metals, Mg and Ba. High

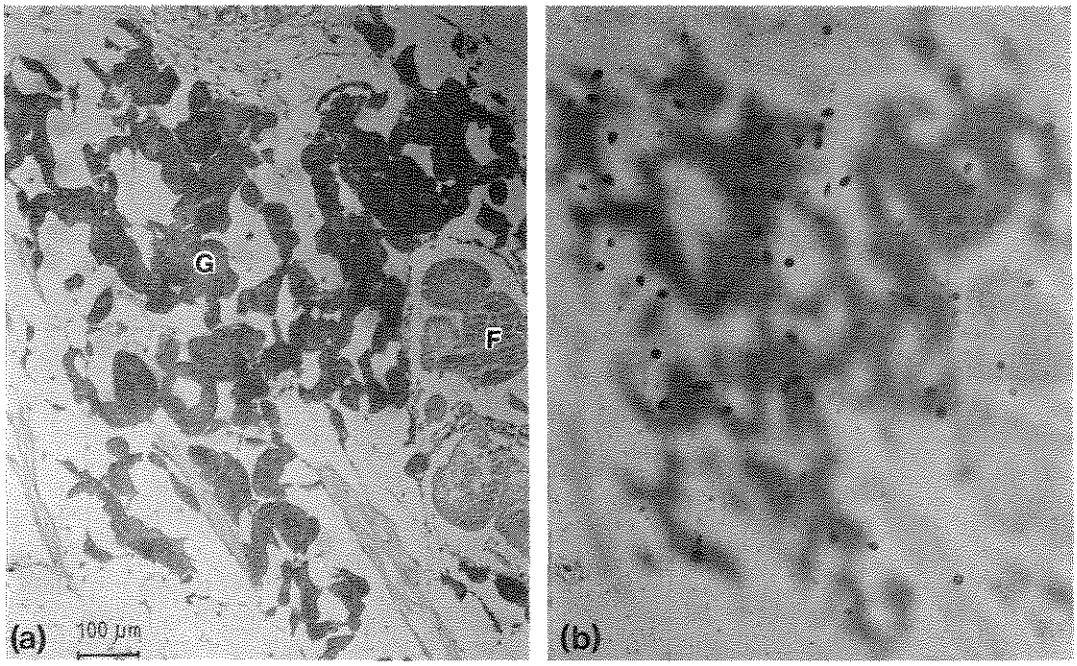

Fig. 4. Histological section showing (a) granular deposits (G) and ♀ gonad (F) and (b) alpha tracks (in focus) in CR-39 that overlays the histological section.

peaks are also shown for P and Fe and the presence of Al and Mn in the granules is also indicated. Silicon, S and Cl present in this spectrum are artefacts caused by the use of tape to adhere the granules to the carbon block (Fig. 7b (ii)).

Similar spectra were obtained from components of the gill support structures.

(b) In situ *granular deposits*. Figure 8(a) is a scanning electron micrograph of a region near the mantle edge containing large aggregations of granules. From this general region, microprobe analyses of many individual granules all gave a spectrum similar to that shown in Fig. 8(c) (i). The alkaline earths Ca and Ba, but not Mg, are present. Phosphorus, Mn and Fe are also present in the granules. Silicon and Cl present in this spectrum are artefacts caused by the use of

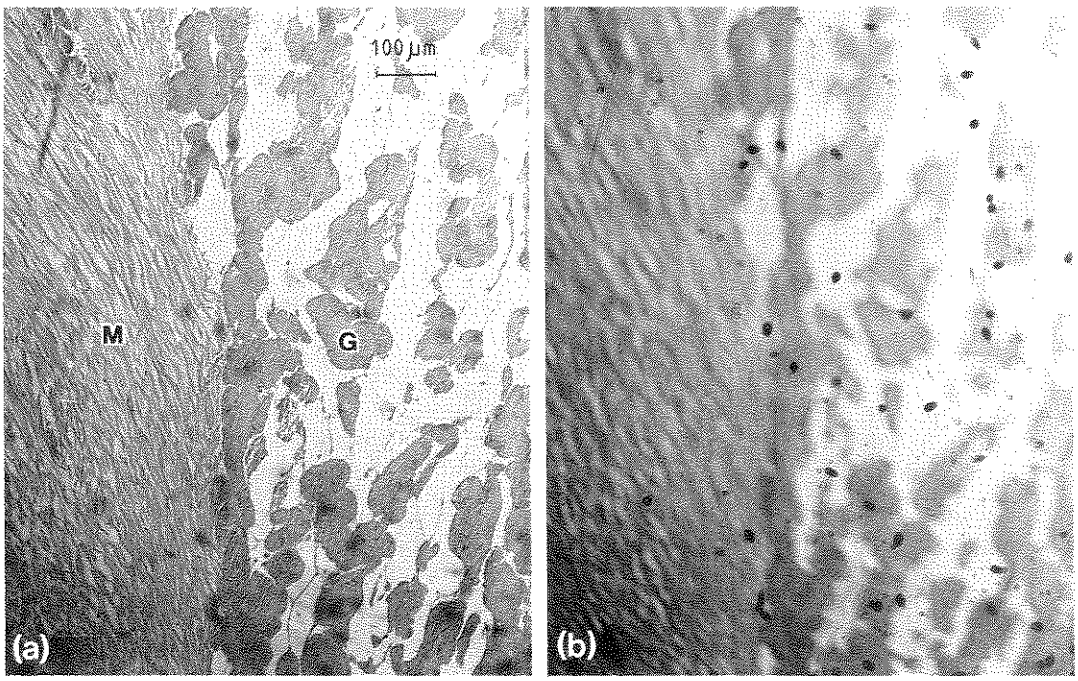

Fig. 5. Histological section showing (a) granular deposits (G) adjacent to muscle tissue (M) and (b) alpha tracks (in focus) in CR-39 that overlays the histological section (from Ellis and Jeffree, 1982).

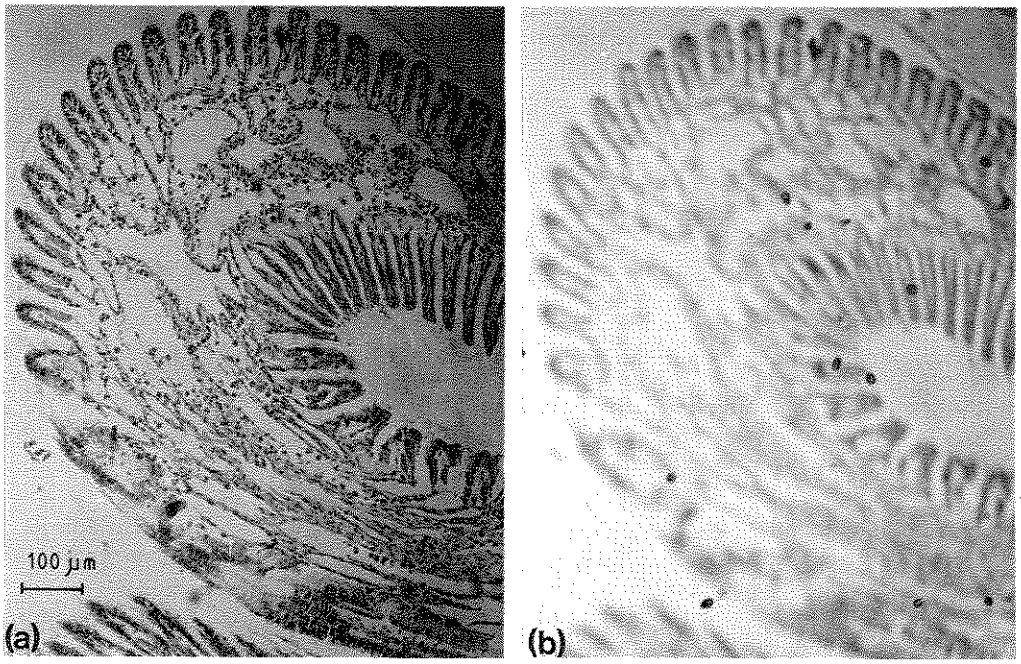

Fig. 6. Histological section showing (a) gill and (b) a lower density of alpha tracks (in focus) in CR-39 that overlays the histological section.

embedding medium (Fig. 8c (ii)) and Os results from postfixing of the tissue in OsO_4. An X-ray map (Fig. 8b) showing the relative abundance of Ca over a broader area indicates a distribution identical to that of the granular deposits in Fig. 8(a). Similar distributions were obtained for Ba, P, Mn and Fe. The absence of Mg in the spectra for *in situ* granular deposits suggests that Mg was leached during preparation of the section to a level below the limit of detection of the microprobe technique. Similar spectra were obtained for granular deposits in the visceral mass.

Histochemical studies

Figure 9 shows the distribution of Ca in a transverse section of the tissues of *V. angasi*, indicating that it is predominantly located in the granular deposits. They are dispersed throughout the visceral mass and are particularly abundant in the labial palps. The mantle only contains large deposits of granules within its edge region (Fig. 8). The muscular foot has few granular deposits. Calcium is also located in the gill support material.

DISCUSSION

Co-location of ^{226}Ra and the other alkaline earth metals

This study was aimed at determining whether ^{226}Ra was similarly distributed among the tissues of *V. angasi* as a potential metabolic analogue to the elements Ca and Mg. The results demonstrated that firstly, at the most general level, i.e. among the dissected components of the mussel, ^{226}Ra, Ca, Mg and Ba are distributed similarly throughout the body. The dissected components of the body have considerably varied concentrations of ^{226}Ra and other alkaline earth metals; this variation is mainly explained by the heterogeneous distribution of the granular deposits. Examination of histological sections showed high concentrations of granules in tissues containing large amounts of ^{226}Ra, such as the visceral mass and palps, but very low concentrations in tissues that contained low levels of ^{226}Ra such as the foot. In contrast to other molluscs which accumulate high concentrations of metals in their excretory organs (Coombs and George, 1978; George *et al.*, 1980; Guary *et al.*, 1981), the kidney/heart of *V. angasi* contained lower concentrations of ^{226}Ra and the other alkaline earth metals than other dissected components such as the visceral mass and palps.

Confirmation of the co-location of ^{226}Ra with the other alkaline earths in the regions of granular deposits was obtained from the histochemical studies and from the combination of alpha-track autoradiography with X-ray elemental analysis.

The co-location of ^{226}Ra and Ca in the tissues of *V. angasi* is consistent with the findings of other studies which showed that ^{226}Ra is deposited in areas of high Ca deposition. In the freshwater gastropod, *Lymnaea stagnalis*, the highest concentration factor for ^{226}Ra is in the newly formed border of the shell (Van der Borght, 1963) and in the bony fish, *Lepomis macrochirus*, the ^{226}Ra is associated with the jaws, vertebrae and scales (Lucas *et al.*, 1979). Similarly ^{226}Ra, like Ca, is deposited and retained almost exclusively in the skeletal tissues of the dog, *Canis familiaris* (Stover *et al.*, 1957; Van Dilla *et al.*, 1958) and humans (Norris *et al.*, 1955; Evans, 1974). The co-location in the granules of the non-essential element Ba with ^{226}Ra, Ca and Mg suggests a similar metabolic pathway for Ba to that of ^{226}Ra, i.e. it is an analogue of Ca and/or Mg.

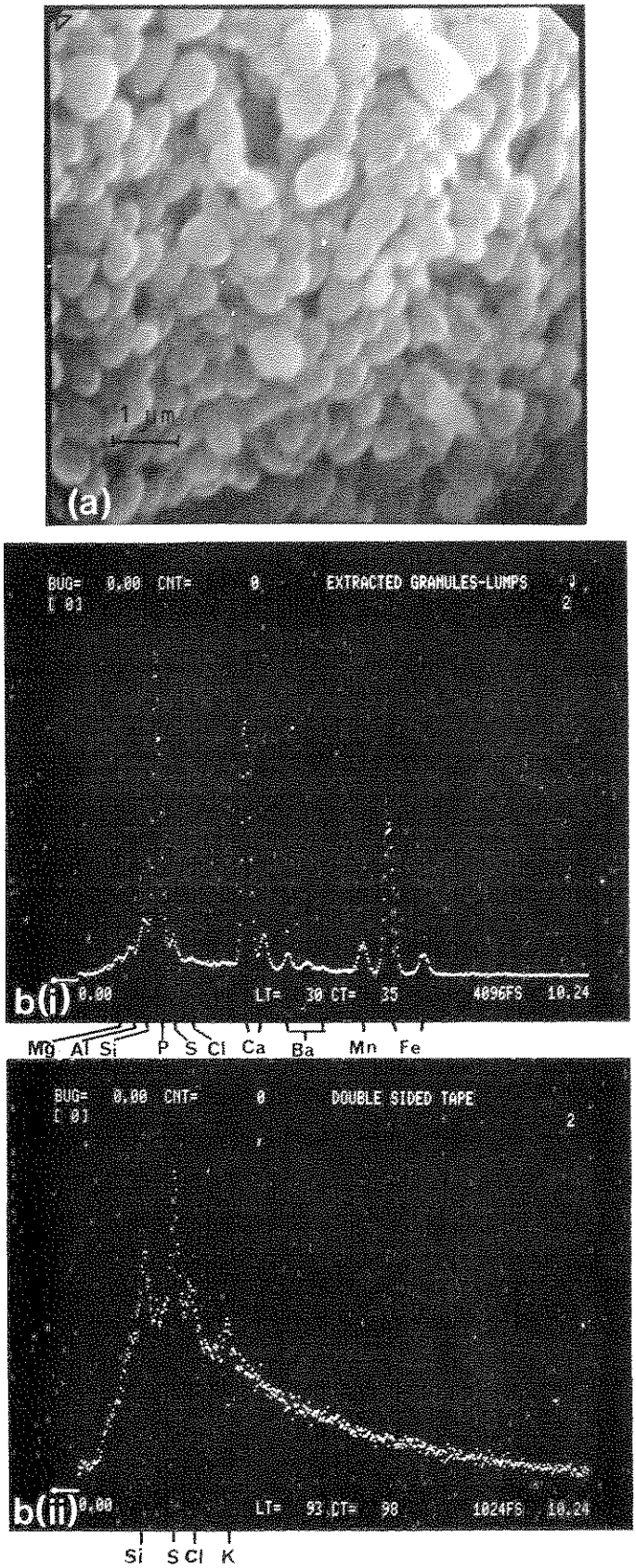

Fig. 7. (a) A scanning electron micrograph of extracted granules and (b) a spectrum of elements in (i) extracted granules and adhesive tape and (ii) in adhesive tape.

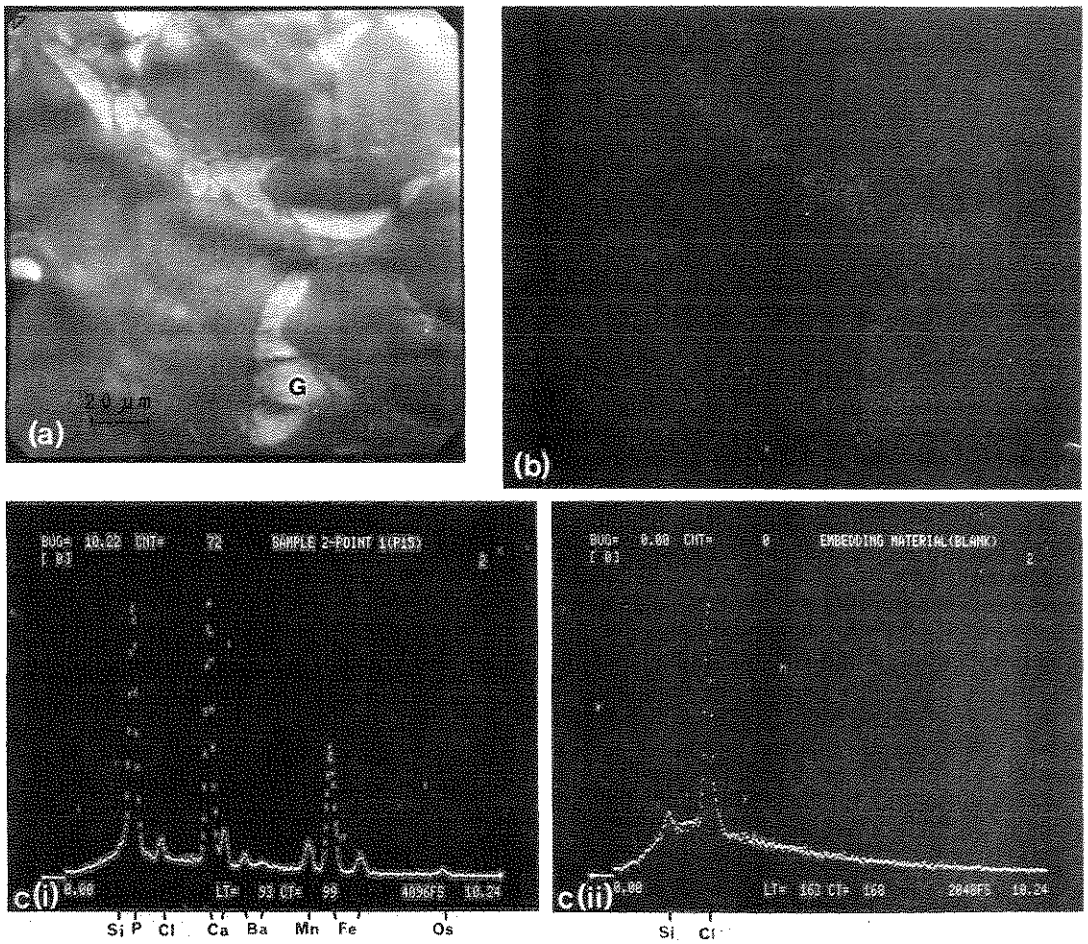

Fig. 8. (a) A scanning electron micrograph of a region near the mantle edge, containing large deposits of granules (G), (b) an X-ray map of Ca (Kα scan) in this region, (c) a spectrum of elements in (i) the granular deposits and embedding medium and (ii) in the embedding medium.

Granules as sites of pollutant accumulation

As well as ^{226}Ra, the granular deposits in *V. angasi* are the accumulation sites for the radionuclides, ^{234}U, ^{238}U, ^{230}Th, ^{210}Po and ^{218}Po (Jeffree, unpublished) as well as Ca, Mg, Ba, Fe, Mn and Al. Granules are also known to be the accumulation sites for other metals in other organisms. The radionuclides ^{237}Pu and ^{241}Am are associated with the Ca granules in the crab hepatopancreas (Guary and Negrel, 1981) and Ca, Pb, Zn, Ca, Mn, Fe, Hg and Au have been found in granules or vesicles in organisms among many animal phyla (for a summary, see Coombs and George, 1978).

Although the co-location of ^{226}Ra with Ca and Mg in the granules of *V. angasi* is consistent with the notion that ^{226}Ra is treated as a metabolic analogue of these two essential elements, the presence of so many other elements in granular deposits requires an explanation that is consistent with that proposed for the presence of ^{226}Ra.

Two hypotheses are offered:

(i) The first is simply that the non-alkaline earth metals are accumulated in the granules, via different metabolic pathways to that of the alkaline earths.

(ii) The second is that many or all these elements do follow a similar metabolic pathway to Ca or Mg. It is more likely they follow the pathway of the dominant Ca because *V. angasi* has a high demand for Ca to enable it to build a large shell relative to its tissue mass. Coombs and George (1978) have also suggested that the presence of a variety of metals in vesicles or granules together with Ca points to a common metabolic pathway.

From the second proposition it follows that *V. angasi* must have a rather undiscriminating mechanism for Ca uptake; possibly the mechanism for the accumulation of large amounts of Ca from its low Ca environment has sacrificed some powers of discrimination against other non-essential elements.

Radium-226 accumulation in calcium phosphate granules and their function in V. angasi

Ch'ng-Tan (1968) concluded from her studies on the composition of granules in *V. ambiguus* that they were mainly insoluble phosphates of Ca, Fe and Mg. Similarly, granules or spherites from the mantle of the freshwater mussel, *Amblema plicata perplicata*, contain significant amounts of Ca and P (Davis *et al.*, 1982) as well as Si, Fe, Zn, Mn and Al (Petit *et al.*, 1980). Calcium phosphate granules are also common

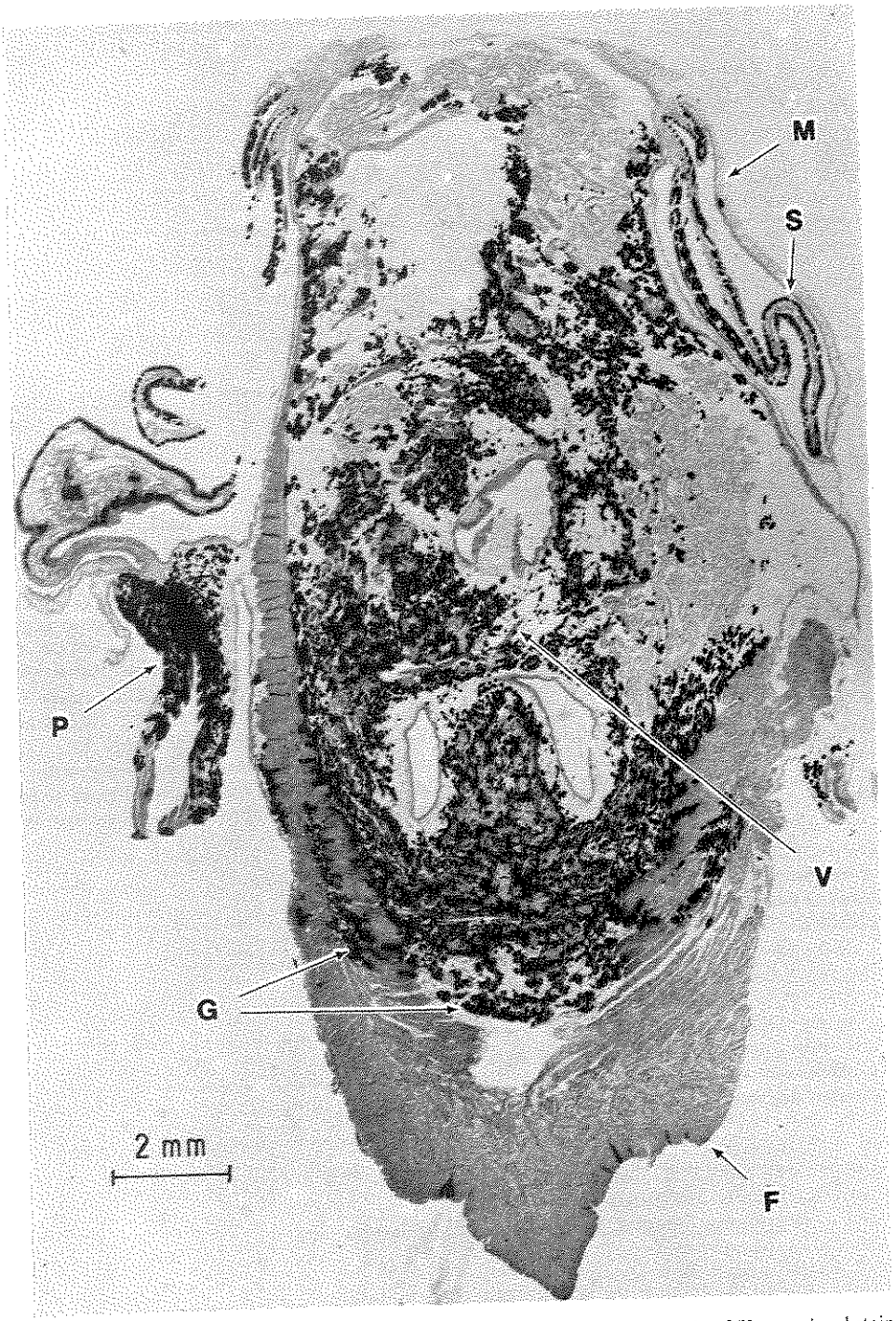

Fig. 9. A transverse histological section taken from the anterior third of the body of *V. angasi* and stained for Ca (dark areas) with alizarin red S. The Ca is located predominantly in the granular deposits (G) that are dispersed throughout the visceral mass (V) and are particularly abundant in the labial palps (P). The mantle (M) only contains large deposits of granules within its edge region, shown in Fig. 8. The muscular foot (F) has very few granular deposits. Ca is also located in the gill support material (S).

in other various animal phyla (Simkiss, 1976; Coombs and George, 1978).

The high peaks for Ca and P obtained from the electron microprobe analyses of *V. angasi* granules is consistent with their being composed of calcium phosphate, with ^{226}Ra also occurring in the granules as a phosphate. Similarly, Ca and ^{226}Ra are deposited in the bone of vertebrate mainly as phosphates; the bone not only has a structural role but, depending on the taxonomic group, may also function as a reservoir for Ca which can be remobilized (Dacke, 1979). Many functions have been attributed to calcium phosphate granules in invertebrates (for a summary, see Simkiss, 1976) and these need to be considered

Table 2. The correlations between the concentrations of ^{226}Ra and other elements in the dissected organ and tissue clusters of *V. angasi*

Mussel No.	Ca	Ba	Mg	Pb	Zn	N
1	0.88**	0.99***	0.76*	0.33	0.57	8
2	0.93***	0.90**	0.89**			8
3	0.96***	0.999***	0.95***			7
Pooled data	0.93***	0.97***	0.90***			23

***$P \leq 0.001$; **$P \leq 0.01$; *$P \leq 0.05$.

when evaluating the co-location of ^{226}Ra, Ca and Mg as evidence that is supportive of the metabolic analogue hypothesis.

The granules have been discussed as the end product of a mechanism for the immobilization and detoxication of metals, being excreted or transferred to other tissues (Coombs and George, 1978). These conclusions have, in general, been based on studies of animals that accumulate high concentrations of granules in organs having excretory functions (Coombs and George, 1978; George *et al.*, 1980; Guary *et al.*, 1981; Guary and Negrel, 1981).

It has also been proposed that granules supply Ca for shell repair and for the elevation of Ca blood concentration in gastropods (Abolins-Krogis, 1968; Watabe *et al.*, 1976; Simkiss, 1976). Studies by Istin and Girard (1970a,b) on extracellular granules in the mantle of several freshwater bivalves demonstrated that Ca from the granules can be remobilized into solution. They concluded that the granules provided a source of exchangeable Ca to maintain saturated levels of Ca in the haemolymph and extrapallial fluid. Moreover the body fluids of the freshwater bivalve, *Anodonta cygnea*, are saturated with respect to aragonite (Potts, 1954), thus facilitating shell deposition. Studies by Petit *et al.* (1980) on the calcification process in the freshwater mussel, *Amblema plicata perplicata* also indicated that calcium granules supply calcium for shell construction.

If the granular deposits of *V. angasi* function as sources of Ca that can be remobilized for shell construction, then the co-location of ^{226}Ra and Ba with Ca in the granules would be consistent with the proposal that ^{226}Ra and Ba are stored as metabolic analogues of Ca. If granules function only as accumulation sites and as the end product of a mechanism that immobilizes unrequired elements such as ^{226}Ra and other alpha-emitting radionuclides, Ba, Al, Mn and Fe, then the co-location of ^{226}Ra and Ca and Mg in the granules cannot be regarded as supportive evidence for the metabolic analogue hypothesis. However, the low concentrations of ^{226}Ra and the other alkaline earth metals in the kidney/heart discounts the idea that *V. angasi* uses the granules both to immobilize and excrete unrequired elements.

In effect the granular deposits may, as a result of two possible mechanisms that are not necessarily mutually exclusive, simultaneously provide Ca for shell building and immobilize ^{226}Ra; this is suggested by the following pattern of accumulation of ^{226}Ra and Ca.

Both Ca and ^{226}Ra tissue concentrations increase with size and age in *V. angasi* (Jeffree, 1983; Jeffree and Davy, 1983). Furthermore, ^{226}Ra multiplies in concentration at a greater rate than Ca (Table 1, Jeffree, 1983). This relationship is exemplified in Fig. 10 where the ratio of ^{226}Ra:Ca tissue concentration increases significantly ($P < 0.00001$) with age for mussels from Corndorl Billabong.

Because there is greater apparent discrimination against ^{226}Ra than Ca across the mantle, compared to the tissue of *V. angasi*, as indicated by the lower Ra:Ca ratio in shell (Davy and Conway, 1974), ^{226}Ra may be selectively retained in the body fluids and returned to the granular deposits. Radium-226 may also be retained more selectively than Ca in the granular deposits as they exchange with the body fluids owing to the lower solubility of radium phosphate. Figure 11 shows the values for the critical

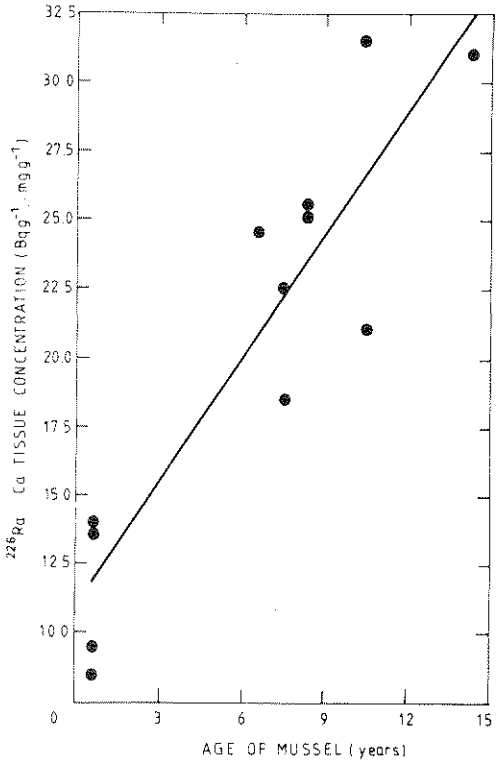

Fig. 10. The linear regression of the ratio of ^{226}Ra tissue concentration to Ca tissue concentration (Bq g^{-1} ^{226}Ra: mg g^{-1} Ca) versus mussel age (years). ($P < 0.00001$, $R^2 = 0.84$).

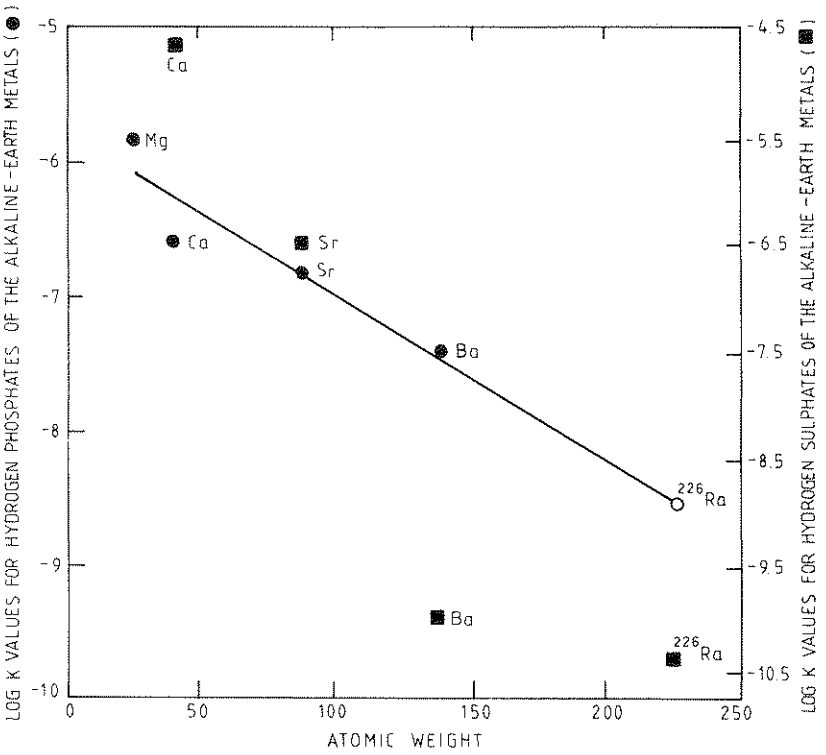

Fig. 11. The stability constants (log K) for the hydrogen phosphates (●) and hydrogen sulphates (■) of the alkaline earth metals as a function of their atomic weight. The log K values were taken from Smith and Martell (1976). The linear regression of log K for the hydrogen phosphate ($\hat{Y}$) versus the atomic weight of the alkaline earth metal (X) gave the equation, $\hat{Y} = -5.80 - 0.012X$, that was used to predict a log K value for the hydrogen phosphate of ^{226}Ra (○).

stability constants, log K (Smith and Martell, 1976) for the hydrogen phosphates of ^{226}Ra, Ba, Ca and Mg. Log K is a measure of solubility and these values are plotted as a function of the atomic weight of the alkaline earth metal. Values of log K for the hydrogen sulphates of the alkaline earth metals, from Mg through to Ra, have also been plotted against atomic weight for comparison with the hydrogen phosphates.

Both sets of data indicate that solubility decreases with increasing atomic weight of the alkaline earth metal. Depending on the actual log K value for the particular Ca and ^{226}Ra phosphates or phosphate complexes that occur in the granules, ^{226}Ra may be retained in the granules, whereas the more soluble Ca may be remobilized into the body fluids.

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