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Distribution and Retention in Bone of ^{226}Ra and Comparison with the ICRP 20 Model*

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ABSTRACT

Analyses are presented of the ratios of ^{226}Ra to calcium in over 650 samples of compact and cancellous bone from 66 female and 26 male subjects who had died from less than one to 60 years after first exposure to radium. The $^{226}Ra/Ca$ ratios were normalized to the terminal ^{226}Ra skeletal content. The $^{226}Ra/Ca$ ratios for vertebrae were essentially identical to those for other cancellous bone for a given subject.

Comparisons of the data with predictions of the ICRP model of alkaline earth metabolism show that for female cancellous bone the normalized $^{226}Ra/Ca$ ratios tended to be greater than predicted, while those for female cortical bone (femoral and tibial shaft) tended to be less. The data for males were fitted better by the model. A modification of the model to reduce the amount of radium deposited in soft tissue fitted the data better in some respects. A straight line linear least squares fit to the data appeared to fit as well as, or

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DISCLAIMER

better than, the models. A radiation effect was suggested in that the normalized $^{226}Ra/Ca$ ratio for vertebrae relative to the ratio expected increased with skeletal absorbed dose for vertebra. However, no such effect was apparent for compact bone or for the cancellous bone as a whole.

INTRODUCTION

Since the formation of the Center for Human Radiobiology (CHR) in 1969, extensive measurements have been made on samples of tissues, excreta and diet from subjects exposed to radium (Ho70,Ho71,Ho73a, Ho73b,Ho76,Ho77,Ru76). Reports dealing with radium in bone have been confined largely to studies of the distribution in an individual skeleton or bone (Ho71,Ho72,Ho73a) or to the relationship of the content of the whole skeleton to that in a single bone sample (Ho70,Ho73a).

A preliminary analysis of multiple bone samples was reported by Rundo et al. (Ru78). We have extended that analysis and examine here the results for 654 samples of bone to determine the extent to which the data provide support for the ICRP model for alkaline earth metabolism (ICRP73) and to seek evidence for an effect of radiation on the metabolism of radium in man.

MATERIALS AND METHODS

Samples for these studies were obtained from exhumed and willed bodies, from surgical specimens, and from autopsies. Exhumation was the primary source. In mid-1972, the sampling of bone was standardized; when sufficient bone was available, the set of samples consisted of seven pieces of femur, one piece of rib (mid-section), one-half a vertebra, and one-half the iliac crest. Figure 1 shows the standard sampling from remains exhumed in 1972 after 12 years burial. The samples taken for chemical analysis are identified by the adjacent letters RC and by the arrows. Other labels are codes for the anatomical locations of the samples or give the case number and date. The pieces of bone with the adjacent letters MA were used for microradiography and autoradiography; the remaining material was stored for possible future use.

The samples were either ashed overnight at 600°C and dissolved in nitric acid, or wet-ashed with nitric and perchloric acids. Once in solution, the samples were analyzed for ^{226}Ra by the emanation method of Lucas (Lu77,Ho79) and for calcium by atomic absorption spectrophotometry (Ho79).

TREATMENT OF DATA

In our analysis of the data we attempt to allow for all variables inherent in the sampled materials and the subjects from whom they came. The most obvious confounding factor is the total body content of ^{226}Ra, which ranges from less than 7 nCi (260 Bq) to 25 µCi (925 kBq); other factors are sex, age of the subject at first exposure, duration of exposure, retention time, duration of burial (which might relate to differential leaching by ground water of radium versus calcium), radiation dose-rate (including the effect of the 5.75-yr ^{228}Ra content) and biological variability between individuals. Taking the last point first, there is considerable variability in the gross distribution of radium measured in vivo (Ru76); in 18 of 40 subjects investigated the highest activity was located in the lowest third of the (erect) body, and in 12 of the subjects the highest activity was located in the uppermost third of the body. These differences are of particular importance because the femur is heavily represented in the standardized sampling and they must be borne in mind when a comparison is made of results for femur from many subjects. Variations of a factor of about two on either side of the means of the measured quantities are, therefore, to be expected.

The effects of variations in terminal radium content among the subjects were eliminated by normalizing the radium concentration (pCi/g calcium) in the samples to the total radium content of the body in μCi. Use of this $^{226}Ra/Ca$ ratio (rather than radium concentration in dry or wet bone) normalizes the data to the mineral content of the bone, and eliminates variations caused by different pretreatment or condition (cleaning, drying, presence of fat, etc.)(Ho62). Although this normalized $^{226}Ra/Ca$ ratio has the dimension of $mass^{-1}$ we shall still refer to it in units of pCi/g, it being understood to be for a total radium content of 1 μCi. The analytical errors in the data are not given as they are negligible (2 to 5%) relative to the biological variability, which may amount to 25% or more.

$^{226}Ra/Ca$ ratios were determined for 459 bone samples from 66 females and 195 samples from 26 males, as summarized in Table 1. Of the 66 females 51 had acquired their burdens as dial painters, 11 had been injected with radium, two had ingested radium water, and two had ingested the nostrum Radithor. The 26 males, on the other hand, had somewhat different exposure histories; none had been dial painters, 12 had been radium chemists, and 9 had acquired radium by injection, three by ingestion of Radithor, and two by ingestion of radium water. The information on the subjects was taken from the CHR computer files, summarized in Ref. CHR81.

Comparison with Predictions of the ICRP Model

Consideration of the normalized ^{226}Ra/Ca ratio as a function of time after exposure may be useful in demonstrating changes in the distribution of radium in the skeleton. The ICRP model of alkaline earth metabolism (ICRP73) provides a framework for studying and predicting such changes (Ru78). The model consists of mathematical functions that describe the fractional retention, following intravenous injection of each of the four elements (Ca, Sr, Ba and Ra), in a number of compartments of bone as well as in the whole body, soft tissue and plasma. Of particular interest in the present context are the retention functions for radium in the whole body, and in the volumes of compact bone, and in cancellous (trabeculated) bone. We designate these R, R_{cp}, and R_{cn}, respectively. Compact and cancellous bone are defined in relation to the surface/volume ratio; the shaft of a long bone is associated with the compact bone of the model, and the whole of vertebra with cancellous bone, despite the presence of some trabecular bone in the former and of some cortical bone in the latter. The ICRP model (ICRP73) gives the fractional whole body retention function as

$$R = (1-p)e^{-mt} + p\varepsilon^{b}(t+\varepsilon)^{-b}[\beta e^{-r\lambda t} + (1-\beta)e^{-r\sigma\lambda t}] \qquad (1)$$

where ε is a small time related to the turnover of an initial pool,

b is the exponent of the power function,

λ is the rate of apposition and resorption in compact bone ($time^{-1}$),

σ is the ratio of the turnover rates of cancellous and compact bone,

β is the fraction of bone volume activity deposited in compact bone,

r is a factor which corrects for redeposition of activity in new bone at sites of resorption long after injection,

m is the rate constant of a small early exponential in R ($time^{-1}$),

p is the fraction of R not in the early exponential, and

t is the time after injection.

Further, for compact bone, the fractional retention of radium in the volume (the variable $R_{COMPVOLUME}$ in ICRP73) is

$$R_{cp} = p\beta\varepsilon^{b}(t+\Theta)^{-b}\ e^{-r\lambda t} \quad , \qquad (2)$$

and for cancellous (trabeculated) bone, the fractional retention ($R_{CANVOLUME}$ in ICRP73) is

$$R_{cn} = p(1-\beta)\varepsilon^{b}(t+\Theta)^{-b}e^{-\sigma r\lambda t} , \tag{3}$$

where Θ is a time-dependent factor

$$\Theta = \left[0.8\ f_c\ c\lambda\omega(1-R)/p\varepsilon^{b}\beta\eta k\right]^{-1/b} , \tag{4}$$

where c is the total mass of bone calcium, f_c is the ratio of total activity deposited in compact bone volume to that deposited in new compact bone, ω is the discrimination factor for radium relative to calcium from blood to new bone, η is the ratio of excretory plasma clearance of radium relative to that of calcium, and k is the endogenous excretion rate of calcium (urinary plus fecal). The effects of the retention of radium on bone surfaces are neglected here. They are substantial in the first days (< 100) after intake, but they can probably be neglected thereafter.

For compact bone the expression predicts the ratio

$$r = 10^{6}\ R_{cp}/0.8 \cdot c \cdot R \tag{5}$$

where 0.8 is the fraction of the skeletal calcium in compact bone; the factor 10^6 allows for the fact that our ratio is expressed as pCi ^{226}Ra/g Ca per <u>μCi</u> total ^{226}Ra content. The values for the body calcium content (c) have been taken as 750 g for females and 1100 g for males (ICRP75).[a] In an individual, the total calcium content depends on stature and age and, although the age of each of our subjects was known, in most cases we have no information on the height or weight. Therefore, no adjustment of the measured $^{226}Ra/Ca$ ratios could be made to allow for variations with stature.

The expressions for the model's predictions of the normalized $^{226}Ra/Ca$ ratio in compact bone then become, for females and males, respectively

$$r_{cpf} = 1667\ R_C/R \text{ and} \tag{5a}$$

$$r_{cpm} = 1136\ R_C/R\ . \tag{5b}$$

For cancellous bone the ratios for the respective groups are

$$r_{cnf} = 6667\ R_T/R, \text{ and} \tag{6a}$$

$$r_{cnm} = 4545\ R_T/R\ , \tag{6b}$$

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[a]While the parameters in the ICRP model were determined on the basis of a calcium body content of 1000 g, use of those from Reference Man of 750 and 1000 g for females and males, respectively, does not appear to distort the results significantly. These values are necessary to properly normalize the $^{226}Ra/Ca$ ratios in the samples to whole body content. The only explicit use of c in the parameters of the model is in determining the value of Θ (equation 4). Variation of this value over a factor of 5 or more had little effect on the values of r (equation 5) relative to the biological variabilities in the data.

since 0.2 of the total calcium content is considered to be in cancellous bone. In these expressions, no allowance has been made for the radium deposited on bone surfaces, but this is negligible for times of one year or more. Somewhat more serious may be the fact that the expression for R includes the soft tissue component which is estimated in a recent modification of the ICRP model by Schlenker et al.(Sc82) to be 14% of the total body content at one year, 5.4% at three years, and 1.7% at ten years, whereas most of our values for total body content were based on measurements of γ radiation from the exhumed skeleton with no soft tissue. The soft-tissue component would thus appear to cause only a moderate problem.

RESULTS AND DISCUSSION

The results for the Ra/Ca ratios were determined for compact bone, that taken from shafts of the femur, tibia and humerus between and including the distal and proximal metaphyses which include a substantial fraction of cancellous bone. The concentrations appear to be fairly constant in this region as was demonstrated earlier (Ru78) and shown in Figure 2. The other bones in the sampling were considered to be cancellous, such as vertebra, rib, condyle, and femur head.

Compact Bone

In 66 female cases results were obtained from 164 samples of compact bone from the shafts of the femur, tibia and humerus for an average of 2.8 sections per subject shown in Table 1. In 26 males the compact bone results are from 73 specimens, 65 from femur, and eight from tibia (also 2.8 sections per subject). The mean normalized Ra/Ca ratios for eacn subject are plotted in Figures 3a and 3b for the females and males, respectively, as functions of time since midexposure. They may be compared with the predictions for radium metabolism in the ICRP model (ICRP73) drawn as the continuous curves in these figures. The dashed curves represent this model as modified by Schlenker et al. (Sc82) to give a better fit of the model to soft tissue data (Eq. 4). Finally, a third curve (dotted) is a straight line, linear least squares fit to the data. Because of the large scatter of the data and because the initial points are not near zero, as predicted by the ICRP model, the latter curve, even though it is not physiologically correct (it does not pass through the origin), appears to fit as well as or better than the other curves. The slope does not differ significantly from zero ($P > 0.9$), but the intercept does ($P < 0.001$). The values of the parameters for these straight lines fitted to the various bone data are given in Table 2, part A.

While the agreements between the various model curves and the data are poor, that from the ICRP model appears to be an upper limit. The Schlenker et al. curve is essentially identical to that of the ICRP model, but because of its initial rapid rise, it appears to provide a slightly better fit to the earlier data. Similar results were obtained for the male compact bone data in Figure 3b, except that the curves provided better fits here than they did to the female data. The curves were generated in an essentially identical manner to those for the females except for the larger calcium content in the males. Neither straight line has a slope significantly greater than zero ($P \gg 0.05$ in Table 2).

Cancellous Bone

The cancellous bones have been taken as all the bones that are not in the bone shaft, such as the head, condyle and trochanter of the femur, and rib, vertebra and innominate. The mean Ra/Ca ratio for vertebral bone for a given subject normalized to the ratio for compact bone are similar to that for all cancellous bone. A fraction of the "cancellous" bone is vertebral bone, about 28% (84 of 295 samples) for females and 23% (22 of 122 samples) for males. The Ra/Ca ratios of vertebral and those for all cancellous bone, plotted in Figures 4a and 4b, are highly correlated, with slopes of 1.02 ± 0.15 ($P < 0.0001$) for females (intercept ~ 0.0) and 1.37 ± 0.08 for males ($P < 0.0001$). However, for the males the intercept is -0.41 ($P < 0.04$). With the latter curve forced through zero the slope is then also ~ 1.0.

A plot of Ra/Ca ratios for the cancellous bone data of the females vs. time since midexposure (Figure 5a) shows the curves to form lower limits to the data, compared to an upper limit for compact bone. For males the cancellous Ra/Ca ratios vs. the time since midexposure were fitted fairly well by the ICRP curve and even better by the modified Schlenker et al. curve (Figure 5b). The linear fit vs. time has a negative slope of (-35.7 ± 9.1)/yr ($P < 0.0007$) with an intercept of 2089 (Table 2.A).

The results for vertebrae in Figure 6 are very similar, but somewhat more scattered than those for the cancellous bone. Note that these are mean values for each subject, so that for the data for the females some high results on single vertebral ratios in Figure 6a are lost when combined with cancellous bone results in Figure 5a.

For the males the data for the vertebrae appear to be slightly more scattered than those of the cancellous bone and the fits are poorer (Figure 6b). The slope of the linear curve is -45.7 ± 10.5/yr ($P < 0.003$) and the intercept is 2434.

Because of the large scatter in the data neither the ICRP model nor the Schlenker et al. modification fit them particularly well, especially those from the females. The curves appear to fit the male data better, both for the compact and cancellous bone. The linear curves appear to fit the data as well as, and in some cases better than, the complex ICRP model and its modification.

The Ra/Ca ratios correlated significantly with time since midexposure ($P < 0.05$) only for cancellous bone ($P < 0.0007$) and vertebra ($P < 0.0003$) of males and only for vertebrae ($P < 0.026$) of females.

To attempt to further reduce the variabilities between individuals, we have divided the mean Ra/Ca ratios of cancellous bone by those in the compact bone in each subject. A plot of these ratios against the time from exposure to death shows no significant trend with time for the female subjects (Figure 7a), but a decrease with time for the males amounting to about -0.11/yr. (Figure 7b). There appear to be no differences in trends with time between the cancellous and vertebral bone in either case. Thus, contrary to the predictions of the ICRP model this analysis indicates that for females, the concentrations in cancellous bone do not decrease significantly with time since exposure relative to those in compact bone. For males the analysis indicates a correspondence with the model, i.e. a relatively faster decrease in the Ra/Ca ratios in cancellous than in compact bone.

Further, as suggested by A. T. Keane, the ratio for retention of radium in cancellous bone relative to that compact expected from the ICRP model is

$$R_{cn}/R_{cp} = \frac{c_{cp}}{c_{cn}} \frac{1 - \beta}{\beta} e^{-(1-\sigma)\lambda rt} \qquad (7)$$

where c_{cp} and c_{cn} are the respective quantities of calcium in compact and cancellous bone, and

$c_{cp}/c_{cn} = 4$,

$\beta = 0.608$,

$\sigma = 4$,

$\lambda = 0.015\ y^{-1}$, and

$r = 0.997$.

Thus,

$$R_{cn}/R_{cp} = 2.58e^{-0.04486t}$$

and it is independent of total calcium body content. It is plotted on Figs. 7a and 7b as the dashed curve. It does not appear to fit the data well for either sex, but the intercept at t - 0 $\approx$ 2.6 is close to that of the fitted lines. It is a more reasonable fit for the males than for the females in that for the former it decreases, as does the straight line, but more rapidly.

The normalized Ra/Ca ratios were checked for correlation with the variables available, the exposure time (midexposure to death), mean skeletal dose and the terminal ^{226}Ra body content. Only the exposure time and dose showed significant correlations with the Ra/Ca ratios and then only in some cases. (The ages at intake and death also showed correlations, but these were not independent variables; they correlated strongly with exposure time, also.) The correlations with mean skeletal doses were significant only for compact bone ($P = 0.014$) and vertebra ($P = 0.0046$) of females. Thus, the dose effect appeared to be present in the females and in the cancellous bone, especially the vertebra.

To further demonstrate a relationship between absorbed dose and Ra/Ca ratio, we compared observed Ra/Ca ratio to that expected based on the straight line fits to the data (in Figures 3, 5, and 6, which are summarized, and Table 2A). These are plotted against dose in Figures 8 (compact bone), 9 (cancellous bone), and 10 (vertebra). For all cancellous bone the slopes of the straight line fits do not differ significantly from zero for either sex, but they have positive slopes for female bone, about 0.016 per krad ($P = 0.023$) for compact bone and 0.011 per krad ($P < 0.0001$) for vertebra; for males the slopes did not differ significantly from zero ($P > 0.1$).

Thus, there appears to be some effect of dose on the bone, at least for the females; the Ra/Ca ratios increase relative to those expected from the linear fits of Ra/Ca vs. time since midexposure. This dose effect was not evident for males. The demonstration of a possible effect on the metabolism of radium in bone is of interest, although the data are inconclusive because of the wide scatter. The smaller effect on the compact bone (0.001 per krad for compact vs. 0.097 per krad for vertebra) may be due to the fact that this type of bone turns over more slowly than does cancellous bone. In a similar vein, that this effect is evident only in the females indicates that the turnover rates of bone in females may be greater than that in males.

CONCLUSION

It appears that these results provide only a small amount of support for the ICRP model or for the modification of Schlenker et al., due mainly to the large scatter of the data. There is some evidence against the models or for the values of the parameters proposed for them (ICRP73), in that the curves for compact bone form upper limits to the data and for cancellous bone, lower limits. The data for males provide support for the models, possibly due to the larger value of skeletal calcium used. It is possible that a larger calcium value for the females would result in better fits. However, as noted earlier, the only parameter explicitly dependent on c is θ, and variations of this parameter did not produce appreciably better fits to the data.

Overall the $^{226}Ra/Ca$ ratios in cancellous vs. those in compact bone does not support the model for the female data (Fig. 7a), but it does for that of the males (Fig. 7b). Finally, the Ra/Ca ratios at early times appear to be greater than predicted by the models, except for the trabecular bone as fitted by the model of Schlenker at al. (Sc82), which shows a rapid initial rise.

Possibly additional analyses of the data might be found to correlate the data better, but at present this appears to be a very uncertain venture, due to the apparently large biological variability of these subjects in their metabolism of radium.

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Table 1. Number and types of samples.

Group	No. of Subjects		Number of Bone Samples: Compact[a]		Cancellous[a,b] (Trabecular)		Vertebra	
Females	66		164		295		84	
Dial painters		51		119		228		66
Medical cases		15		45		67		18
Males	26		73		122		28	
Medical cases		14		33		67		15
Chemists		12		40		55		13
	Total		237		417		112	

[a]See text.

[b]Includes vertebra

Table 2. Parameters of straight line least squares fit to the data.

A. Ra/Ca vs. Time since Midexposure

Type of Bone	Sex	Intercept	Slope yr^{-1}	P[a]
Compact				
	F	882	- 0.332	0.92
	M	748	0.839	0.80
Cancellous (with vertebrae)				
	F	1784	2.33	0.75
	M	2089	-35.69	0.0007
Vertebrae, only				
	F	2517	-19.2	0.026
	M	2434	-45.7	0.0003

B. Observed/Expected[b] Ra/Ca Ratios vs. Dose

			Slope, $Rad^{-1} \times 10^6$	
Compact				
	F	1.091	- 7.1	0.094
	M	1.045	-12.2	0.24
Cancellous (with vertebrae)				
	F	0.901	0.93	0.84
	M	1.20	25.8	0.29
Vertebrae, only				
	F	0.839	14.0	0.015
	M	0.899	22.4	0.22

[a]Probability that the slope is zero; probably significant if $P < 0.05$.

[b]"Expected" value from the best straight line fit to the data.

LEGENDS FOR TABLES

Table 1. Number and types of samples.

Table 2. Parameters of straight line least squares fits to the data.

LEGENDS FOR FIGURES

Fig. 1. The bones selected for examination in the standard sampling. The arrows identify the parts submitted for chemical analysis. (case 03-209) (from ref. Ru78).

Fig. 2. Summary of analytical results from 86 samples of femur from 26 females. PMT - proximal metaphysis, MS - midshaft, DMT - distal metaphysis, Dist - distal part of DMT, Cond - condyle (see figure 1) (from Ru78).

Fig. 3. Radium-calcium ratios in compact bone vs. time since mid-exposure for A. females, and B. males. The curves are:

________	ICRP-20 model (ICRP73),
- - - - -	modified ICRP-20 model by Schlenker et al. (Sc82),
....	straight line least squares fit to the data.

Fig. 4. Plot for each subject of the ratios of ^{226}Ra concentrations of vertebra to compact bone vs. those of cancellous bone to compact bone. The straight line is least squares fit to the data. A. females and B. males.

Fig. 5. Radium-calcium ratios in cancellous bone vs. time since mid-exposure. A. females and B. males. See figure 3 for description of curves.

Fig. 6. Radium-calcium ratios in vertebra vs. time since mid-exposure for A. females and B. males. See figure 3 for descriptions of curves.

Fig. 7. Plot for each subject of the ratios of ^{226}Ra concentrations in cancellous or vertebral bone to those in compact bone vs. time since mid-exposure, A. females and B. males. The solid line is a least squares fit to the data. The dashed line is that expected from the ICRP model.

■ cancellous/compact

o vertebra/compact

Fig. 8. Plot for compact bone from A. female and B. males of the ratio of the observed normalized Ra/Ca ratio to the expected ratio vs. average skeletal absorbed dose. Straight line least squares fit to the data. The "expected" value is from the linear fit in figure 3.

Fig. 9. Same as figure 8, but for cancellous bone, A. females and B. males. The "expected" values are obtained from the linear fit in figure 5.

Fig. 10. Same as figure 8, but for vertebrae for A. females and B. males. The "expected" values are obtained from the linear fit in figure 6.

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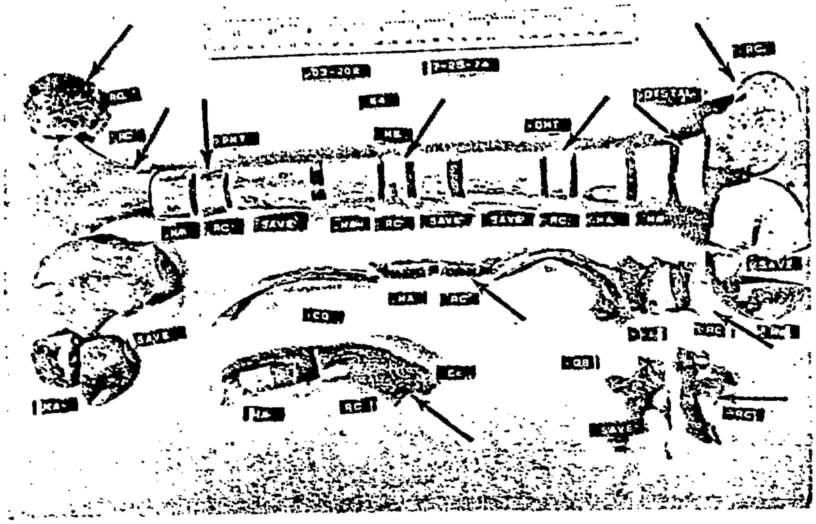
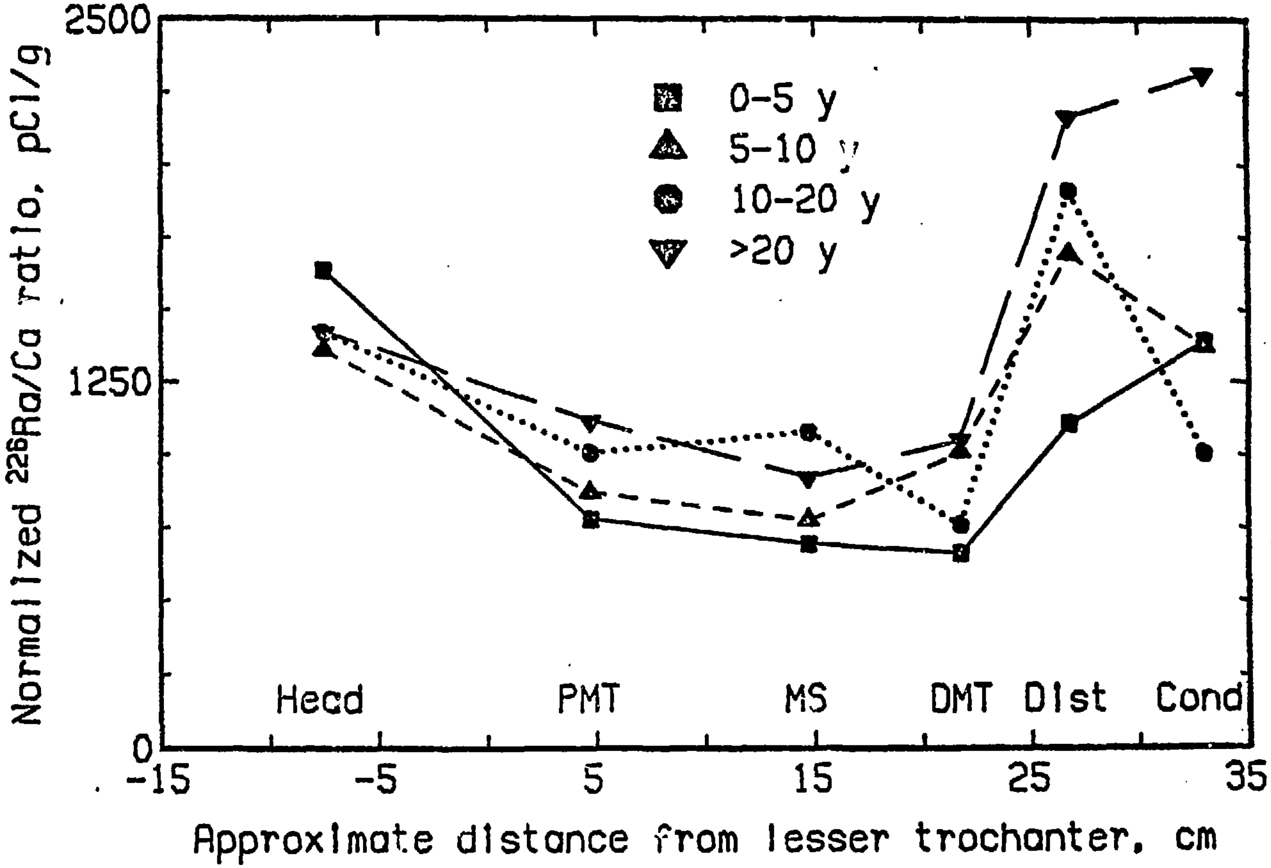
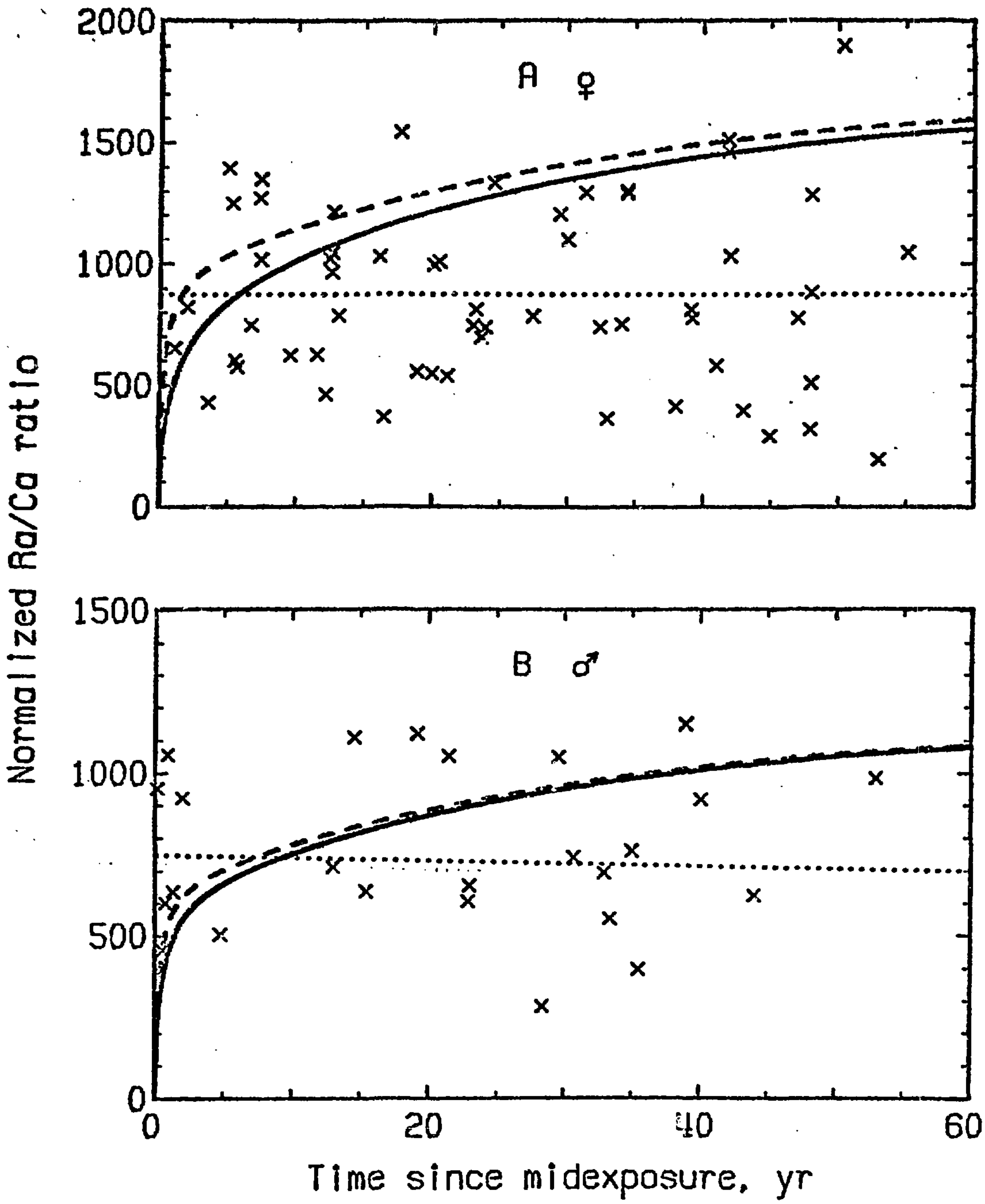


Fig. 1



2500
1250
0
-15
-5
5
15
25
35
Normalized 226Ra/Ca ratio, pCi/g
Approximate distance from lesser trochanter, cm
0-5 y
5-10 y
10-20 y
>20 y
Head
PMT
MS
DMT
Dist
Cond



A ♀
B ♂
2000
1500
1000
500
0
Normalized Ra/Ca ratio
0
20
40
60
Time since midexposure, yr

Fig 2

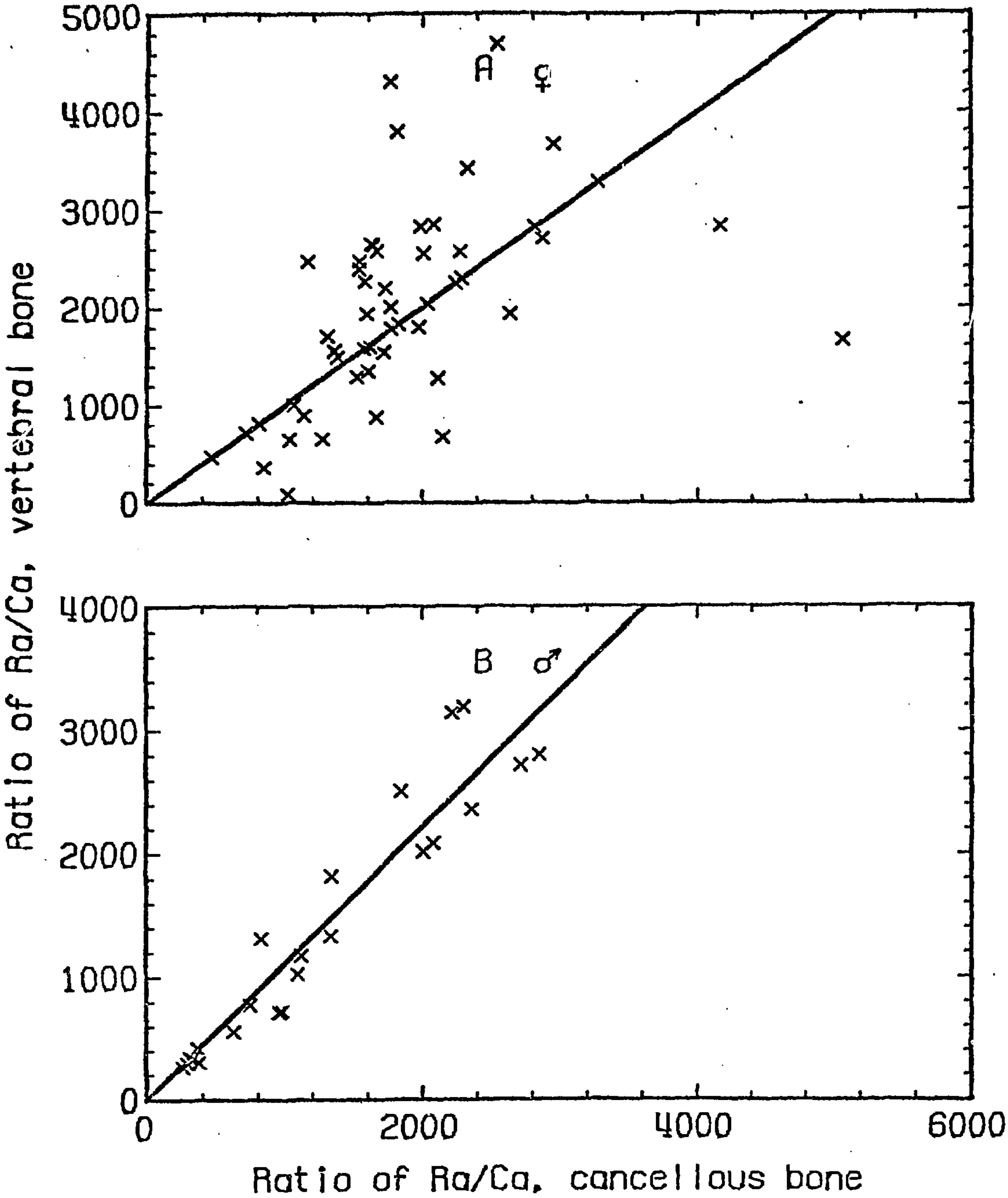
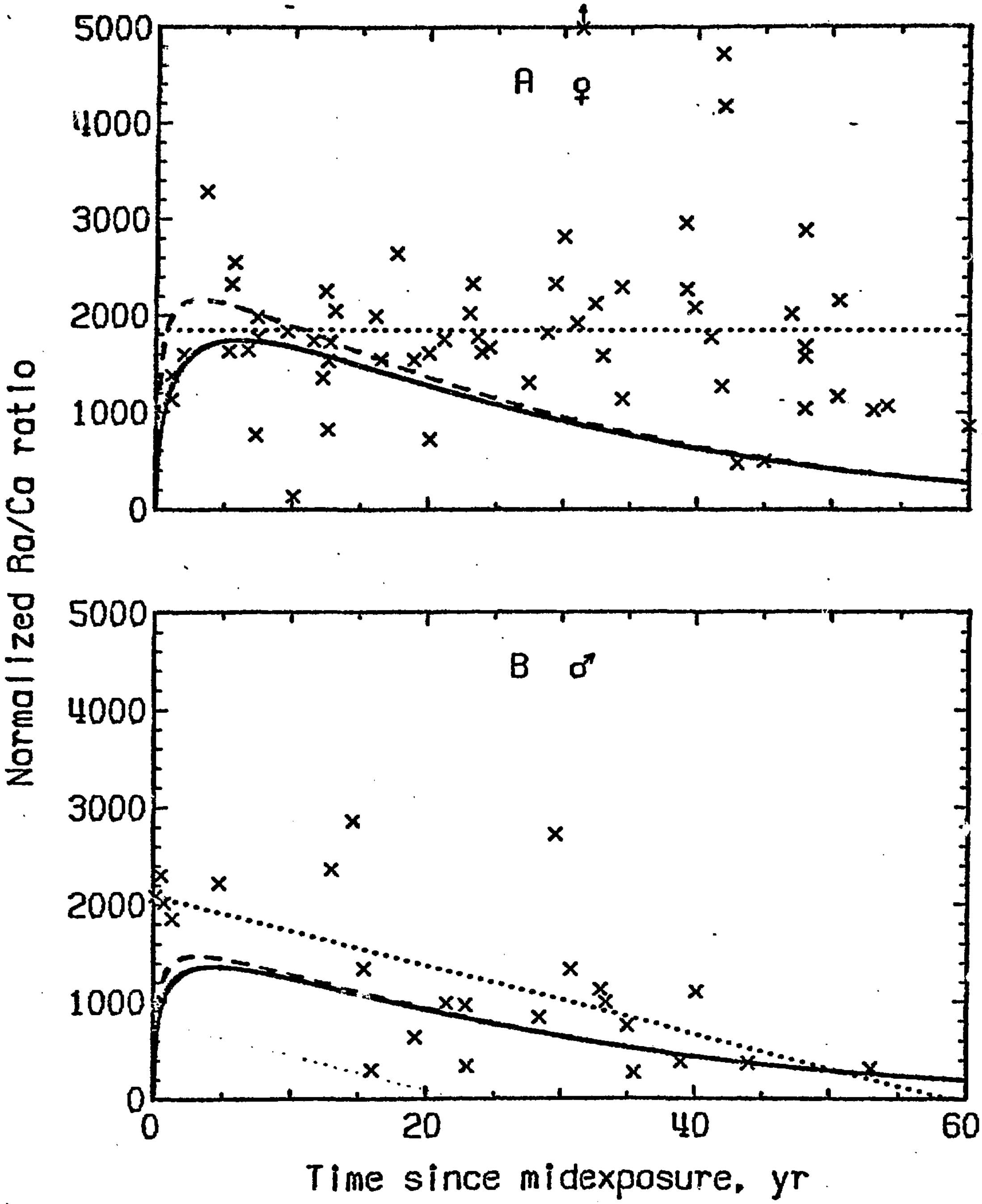
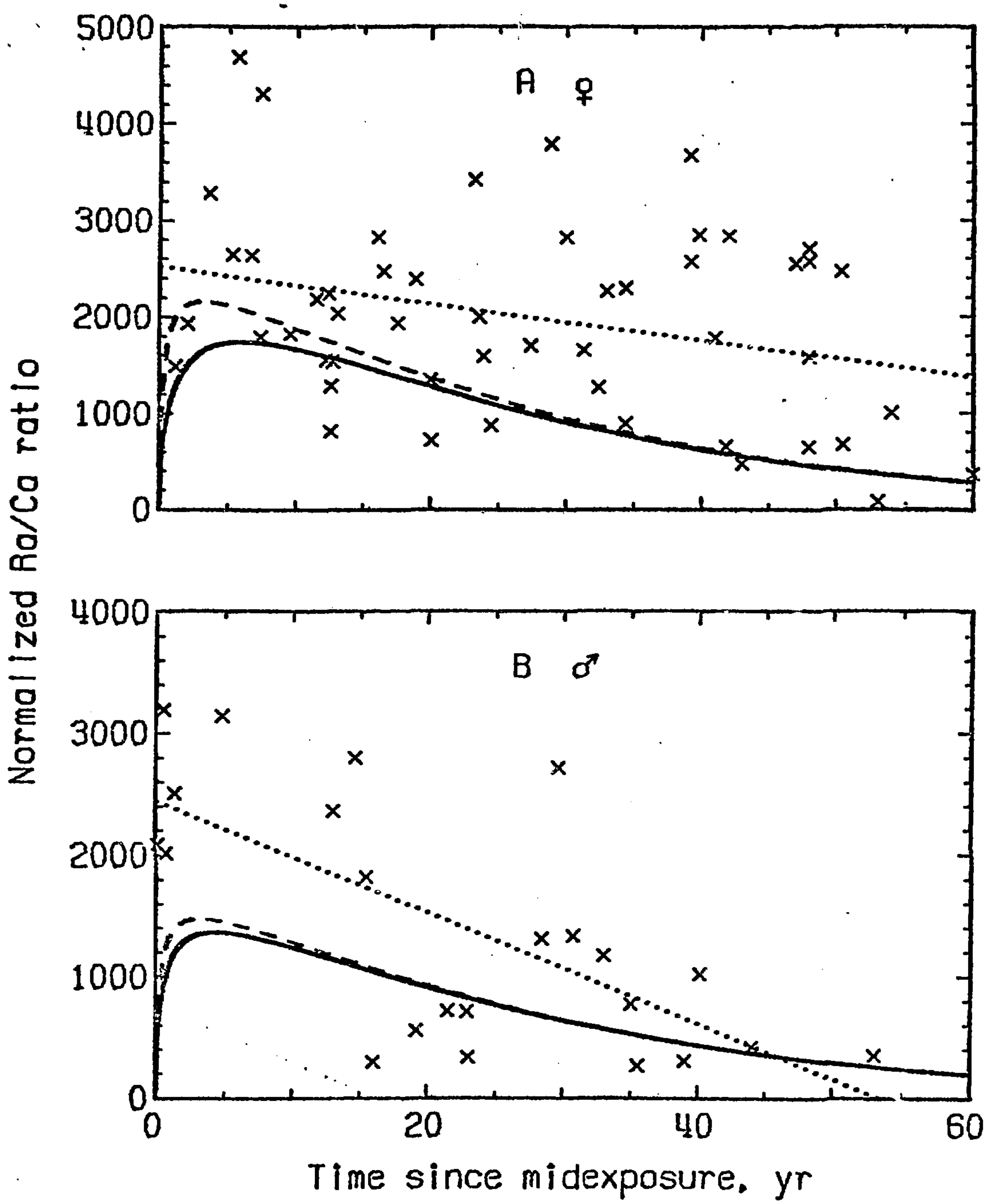

Ratio of Ra/Ca, vertebral bone
5000
4000
3000
2000
1000
0
A ♀
4000
3000
2000
1000
0
B ♂
0
2000
4000
6000
Ratio of Ra/Ca, cancellous bone

Fig. 4a b

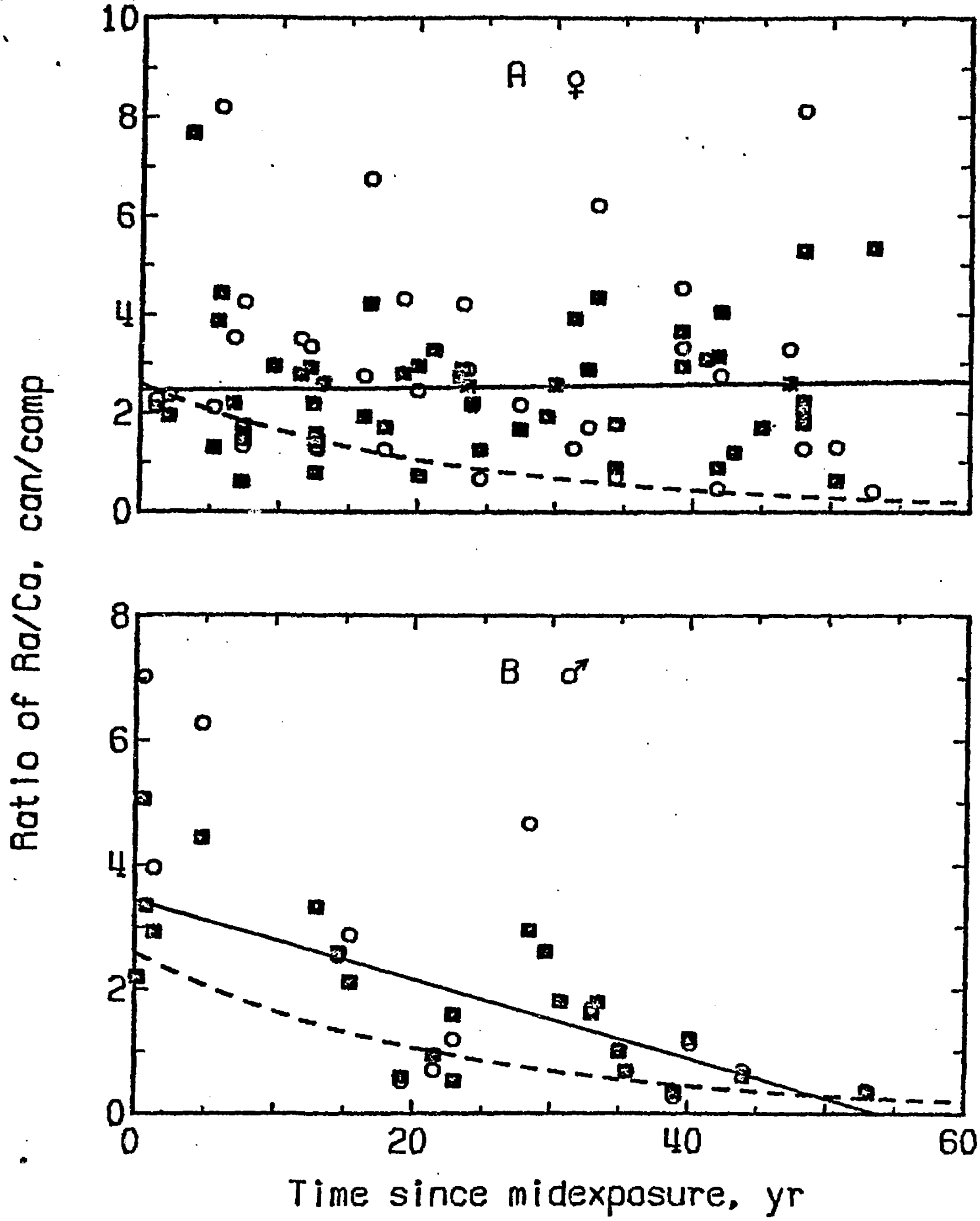


A ♀
B ♂
5000
4000
3000
2000
1000
0
Normalized Ra/Ca ratio
0
20
40
60
Time since midexposure, yr



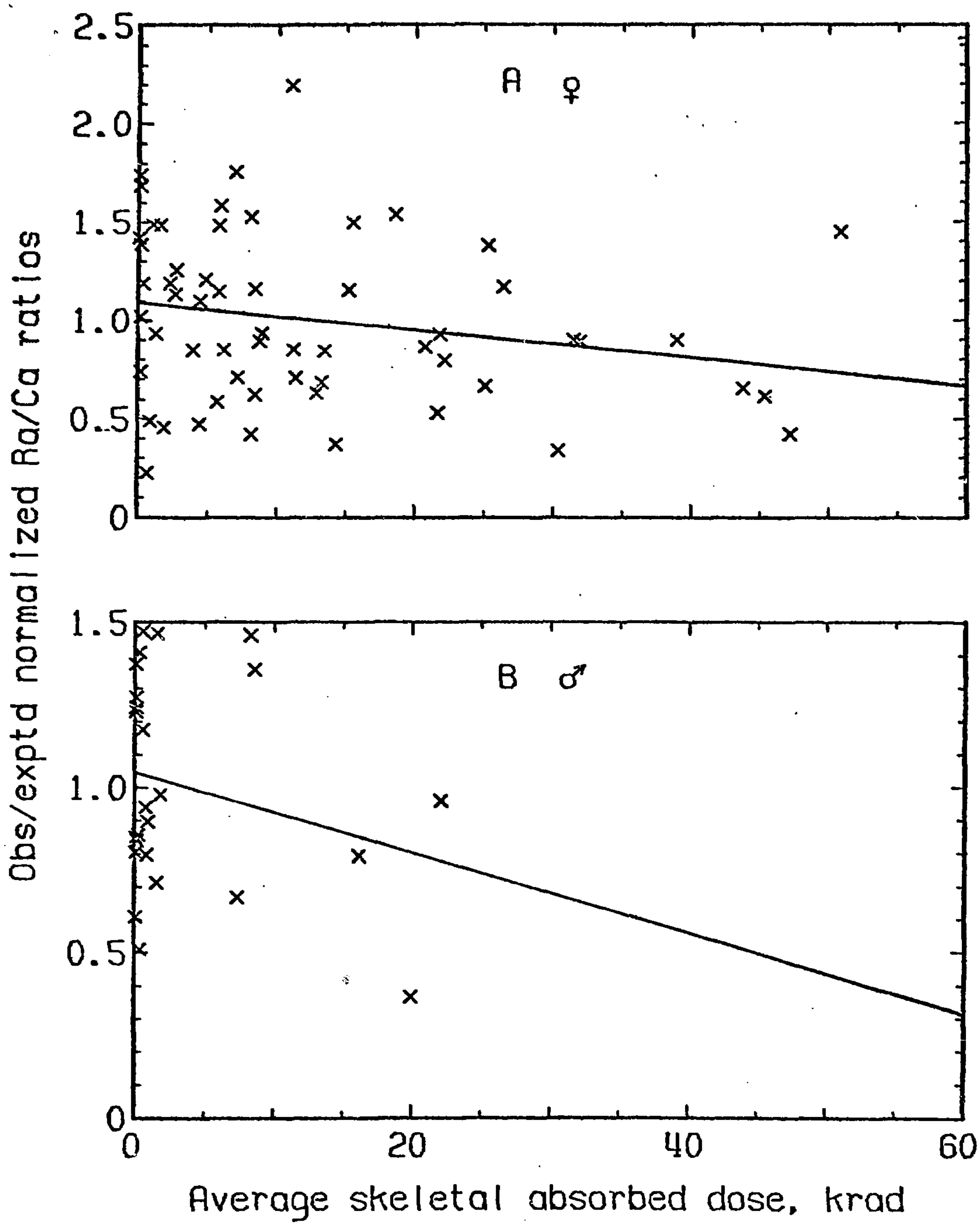
RBH MAY 13 1982

Fig 6a, b



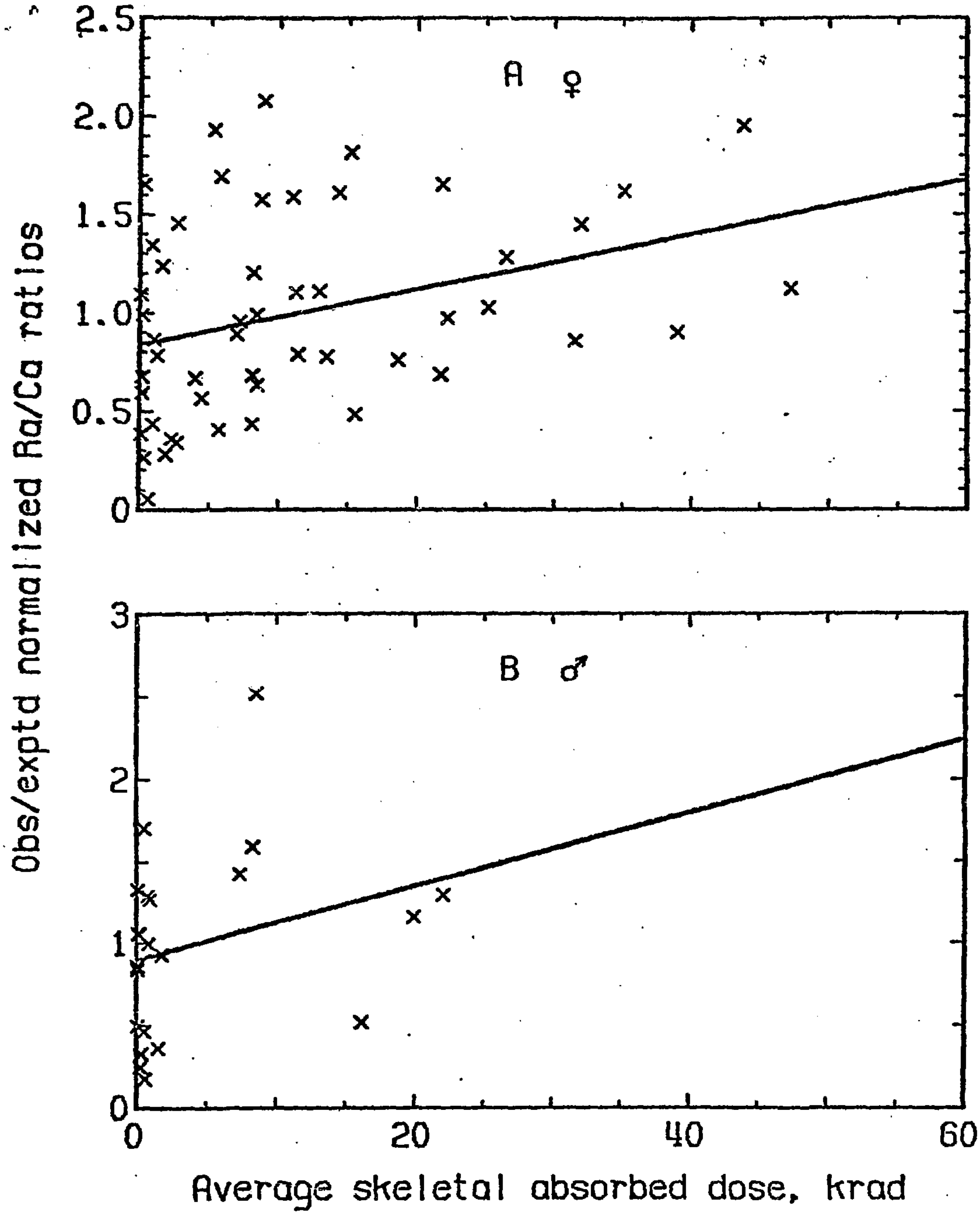
A ♀
B ♂
Ratio of Ra/Ca, can/comp
Time since midexposure, yr
0
2
4
6
8
10
20
40
60

Fig 7a,b



RBH MAY 13 1982

Fig 89b



A ♀
B ♂
Obs/exptd normalized Ra/Ca ratios
Average skeletal absorbed dose, krad

Fig 9a–b

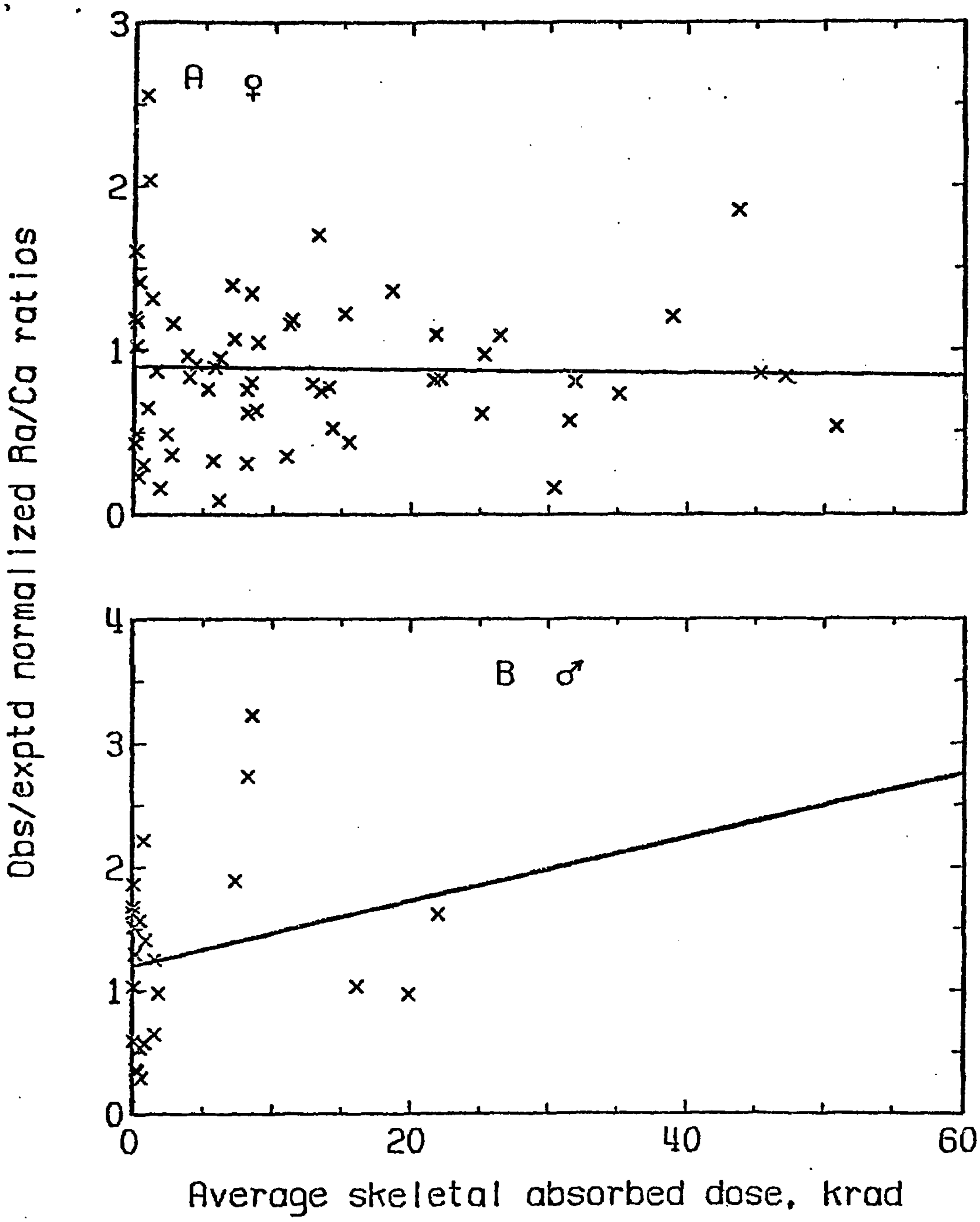


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Fig 10a,b