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DOSE RATES TO AQUATIC LIFE NEAR A URANIUM WASTE SITE

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MANAGEMENT PERSPECTIVE

We have calculated radiation dose rates to aquatic organisms living near an abandoned uranium mine on Lake Athabaska. Dose rates in sediment were over ten times the recommended maxima for humans. Internal dose rates to whitefish bone were somewhat higher than the maxima. We intend to prepare tables for general use for the calculation of dose rates from ingested radioactivity from various sources.

DOSES RADIOACTIVES POUR LA FAUNE AQUATIQUE PRÈS D'UN SITE DE DÉCHETS D'URANIUM

R.F. Platford et S.R. Joshi

PERSPECTIVE-GESTION

Nous avons calculé les doses de radiation pour les organismes aquatiques vivant près d'une mine d'uranium abandonnée sur le lac Athabaska. Dans les sédiments, les doses étaient dix fois supérieures au maximum recommandé pour les humains. Les doses internes des os de corégones étaient un peu supérieures au maximum. Nous prévoyons préparer des tableaux qui pourront être utilisés pour le calcul des doses de radioactivité ingérée provenant de diverses sources.

Abstract. Radiation dose rates to aquatic organisms have been estimated from radionuclide levels in fish, in water and in the sediments near an abandoned uranium waste site. The dose rates in the sediments were high enough ($>10^3$mSv y^{-1}) to pose some risk to any animals residing there. Whitefish received dose rates of >100 mSv y^{-1} from internal ^{226}Ra and could have received comparable dose rates from the sediments if they foraged near the lake bottom. The calculated dose rates to fish were comparable to those reported near an adjacent uranium mine (Sw83).

Résumé. Les doses de radiation pour les organismes aquatiques ont été évaluées en déterminant les teneurs en radionucléides dans les poissons, l'eau et les sédiments près d'un site abandonné de déchets d'uranium. Les doses dans les sédiments étaient suffisamment importantes ($>10^3$mSv an^{-1}) pour présenter des risques à tout animal y vivant. Les corégones ont reçu des doses de 226 Ra supérieures à 100 mSv an^{-1} et pourraient avoir reçu des doses comparables s'ils fourrageaient près des sédiments du fond du lac. Les doses calculées pour les poissons étaient comparables à celles observées près d'une mine d'uranium adjacente (Sw83).

INTRODUCTION

Our Department is responsible for assessing hazards which appear in the aquatic ecosystem. Most abandoned uranium mines have associated tailings which contain daughters of ^{226}Ra. These normally pose little problem for humans, but fish and burrowing organisms can be exposed to constant high radiation levels.

In late July 1983, we collected a series of aquatic samples from the area around Langley Bay, near the east end of Lake Athabaska. The bay contains a waste site from the Gunnar uranium mine which operated from 1955 to 1963 but has since been inactive. In the intervening 20 years, all of the radioactive daughter products of radium have had time to reach (secular) equilibrium, and thus to provide us with an indication of long-term dose rates to aquatic life in the area.

STUDY AREA

The sampling locations are shown in Fig. 1. The mine on the north shore of Beaverlodge Lake, which was operated by Eldorado Nuclear Ltd. from 1952 to 1982 was the subject of an earlier study similar to ours (Sw82; Sw83).

MATERIALS AND METHODS

Sample preparation and counting are described in detail in another paper (Jo8X). Fish organs were freeze-dried and ground, and their activity was determined by gamma ray spectrometry.

DOSIMETRY

We have calculated dose rates to biota from both water and sediments using the general procedure described by Woodhead (Wo76); we describe the calculations in detail, however, as some of our approximations differ from his.

Internal Dosimetry. Most of the internal dose arises from alpha radiation. We have assumed that any radioisotope in an organ is uniformly distributed in a volume approximated either by a sphere or by a long cylinder, and having unit density (except for bone, which is treated separately). The range of an alpha particle in $\mathrm{mg.cm^{-2}}$ is given approximately by (Ch80)

$$R = 0.173\ E_{\alpha}^{3/2}\ A_{w}^{1/3}$$

The most energetic alpha particle in the uranium decay series is the 7.69 MeV particle emitted by ^{214}Po. The weighted average atomic weight, A_w, of water is (2/3x1 + 1/3x16) = 6.0, giving a maximum alpha range in water of 0.067 mm. Most of the calculations in this paper are based on water as the absorbing medium, so the general expression for range in mm becomes

$$R_w = 3.14 \times 10^{-3} E_\alpha^{3/2}$$

For our purposes corrections to account for different types of soft tissue in fish will not affect dose calculations by more than 5% (Wh59).

In order to estimate radiation losses from the surfaces of small organs or animals we have repeated the calculations of Kononenko (Ko57, Sp68). For alpha and beta emitters we have arbitrarily assumed that surface radiation losses of 10% or less have a negligible effect on internal dose rates in a body. The minimum size for such a body is given in the following section, assuming the radioactivity is homogeneously distributed and isotropically emitted. See Appendix A for details of calculations.

The radiation field for a source of finite range is, in conventional polar coordinates (Wh59)

$$D = \frac{A}{2\pi r^2} \int_0^{2\pi} \int_0^{\pi/2} \int_0^{R} f(E) \cos\theta \, r^2 \sin\phi \, dr \, d\phi \, d\theta \qquad (1)$$

where D is the integrated dose rate from a source of specific activity A and having an energy distribution f(E). Kononenko (Ko57, Sp68) gives two important solutions to the equation. The first is the dose D_s from a hemispherical source to an adjacent body; the second is the similar dose rate D_p from a plane source. We use these solutions to calculate minimum organ sizes above which we can ignore a radiation loss (of 10%) through the organ wall. For the case of a sphere containing an alpha emitter with uniform rate of energy loss (LET), f(E) becomes $(1 - r/R_\alpha)$, where R_a is the range. Kononenko gives graphical solutions in terms of a characteristic radius, <u>a</u>, for curved solids, and the particle range R. It turns out that surface losses are fairly insensitive to <u>a</u>/R in this region, but even so, approximations in the integration of the dose field cause differences in the surface loss of a factor of three; because the loss is only about 10% of the total dose the calculations are still reliable enough to use for alpha particle doses. Detailed calculations of surface losses are given in Appendix A and are summarized in Table 1. The dose rates were derived from the expressions given in Appendix B.

Rough calculations of dose rates to the various compartments of the whole Langley Bay ecosystem showed that any organisms living in the sediments should have received the highest dose rates, mainly from alpha radiation. The more important radioisotope activities are given in Table 2 and the corresponding dose rates are given in Table 3, and are plotted schematically in Fig. 2. The target labelled "worms" is intended to include any organisms dwelling in the sediment and which have a homogeneously distributed specific activity, more or less similar to that of the sediment itself. Doses of 10-100 Sv can inactivate bacteria, so that the dose rate of 10^3-10^4 mSv y^{-1} (or 1-10 Sv y^{-1}) given in Table 3 is just enough to have an adverse effect on some organisms. In reality, the ^{226}Ra derived dose rate could be higher, depending on the persistence of its ^{222}Rn daughter, because there are four succeeding alpha emitters in secular or transient equilibrium with the mother isotope.

The internal emitters in whitefish are confined primarily to the gut contents and the bone. The transit time through fish gut at 5-10°C is about 100 hours (Mo67) but the fish sampled were presumably eating a steady diet of contaminated food, which consisted mainly of snails so that the activities given in Table 2, and therefore the dose rates given in Table 3 are probably realistic steady state values. Dose rates to the gut contents are probably not hazardous, although the radiation to the gut walls might be. The first layer of cells in the gut walls will be irradiated at about half the dose rate of the gut contents, because of the geometry, and the cell dimensions.

The dose rate to the bone of about 100 m Sv y^{-1} is about one tenth the 30 day LD_{50} for fish (Ch80) and could pose a risk over several years. Surprisingly, the Whitefish accumulated the largest levels of alpha emitters whereas Swanson (Sw83) found White Suckers to accumulate the largest levels in Beaverlodge Lake, as one would expect from their feeding habits. She found levels about as high in White Suckers as we found in our Whitefish, with similar calculated dose rates.

ACKNOWLEDGEMENT

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Table 1. Minimum organ sizes for negligible surface radiation losses. From Appendix A.

Spherical Shape

Radiation	Energy	Range in Water	Diameter
^{238}U (γ)	4.20 Mev	2.8 cm[a]	>25 cm[b]
^{226}Ra (α)	4.80	33 µm	>140 µm[c],40µm[d],55µm[e]
(γ)	0.186	6 cm	70 cm
^{214}Po (α)	7.69	67 µm	>300 µm,80µm,110µm
^{214}Bi (β)	1.67	7.5 mm	33 mm
^{210}Pb (β)	0.024	10 µm	40 µm
^{210}Pb (γ)	0.046	2.7 cm[a]	25 cm

[a] Half-thickness for gamma rays.
[b] 95% absorption diameter from Loevinger (1969).
[c] From Kononenko (1957).
[d] From Woodhead (1976).
[e] Geometrical estimate (Appendix A).

Table 2. Radioisotope activities in Langley Bay biosphere during 1983[a] (1978 values in brackets).

Station	1	2	3	4	5	6	7	9
Water - Units of Bq per kilogram								
^{234,238}U	0.004	0.004	0.05	0.19	2.51			
^{226}Ra			0.20	0.41	1.7			
^{210}Pb	<0.04	<0.04	<0.04		4.1			
Sediments - Units of Bq per gram dry weight								
^{234,238}U	2.0	1.2	3.3	1.9	1.9		7.1	
^{228}Th	0.27	0.22	0.45	0.23	0.32		0.22	
^{226}Ra	39	11	14	10	3		0.6	
	(9)	(14)	(37)	(48)	(0.6)		(0.3)	
^{210}Pb	43	24	23	35	12		6	
	(14)	(16)	(48)	(27)	(3)		(1)	
Organ	Gut Content	Bone	Liver	Gut	Muscle	Kidney	Female Gonads	Male Gonads
Whitefish - Units of Bq per kilogram wet weight[b].								
^{228}Th	11	2						
^{226}Ra	358	185	3	5	4	3	6	2
^{210}Pb	1030	151	15	5	4	29	6	1

[a] A complete set of our data will be published separately. This table contains representative values.

[b] These values were at least five times the control values of fish taken from the open lake.

Table 3. Dose rates to various biota in Langley Bay (in mSv y^{-1}). From Appendix B.

Origin	Target	Significant Isotope	Dose Rate	Comments
Sediments	"Worms"	^{226}Ra (α)	$10^3 - 10^4$	Inactivates bacteria
Sediments	"Worms"	^{214}Bi (β)	-100	
Internal	Fish gut	^{226}Ra (α)	200	From gut contents
	Fish bone	^{226}Ra (α)	100	
Sediments	Whole fish	^{226}Ra (γ)	100	Bottom feeding fish

APPENDIX A. RADIATION FIELDS NEAR SMALL BODIES

The dose rate from a homogeneous radioactive sphere is given by the polar integral.

$$D = \frac{A\ d\ S}{4\pi\ r^2} \int_{\theta=0}^{2\pi} \int_{\phi=0}^{\pi} \int_{r=0}^{R} f(E) \left[r^2 \sin\phi\ dr\ d\phi\ d\theta\right]$$

We have assumed a constant energy loss rate along the alpha particle track of length R_α (although it is about 50% higher at the end of the track than the beginning). Our assumption gives the integrand $f(E)=1-4/R_\alpha$; A is the specific activity of the sphere and S is the area through which the radiation escapes, i.e. the surface of the sphere. Kononenko's solutions (Ko57) are presented in a semilogarithmic graph for small spheres and cylinders. The cylindrical solution was estimated from the more exact solution for a planar source. The Woodhead report (Wo76) gives a different set of integrals, the solutions of which agree with Kononenko's for small spheres having $r \leq R_\alpha$. Their limits for larger spheres, however, are different in that Kononenko predicts a limiting surface loss fraction of 0.125 at large r, whereas Woodheald gives a fraction of 0.125 at $r/R_\alpha \sim 1.2$.

We have used a simple truncated sphere approximation in which we calculate the frustum corresponding to 0.9 of a sphere's volume. This radius is 0.61 of the sphere radius. Make the frustum plane of this sphere coincident with the surface of a solid emitter. Then 90% of any isotropic radiation from the centre of the sphere will be absorbed in the sphere volume. By rough averaging any plane sheet with thickness larger than about $1+0.61xR_{\alpha}$ will also absorb 90% or more of the dose. A curved sheet folded back on itself to form a cylinder would have to be somewhat larger. Our factor of 1.6 R_{α} can be compared with Woodhead's value of 0.9/0.750 or 1.2 R_{α}. The agreement among the three sets of surface loss estimates is quite adequate for our purposes, working as we do with organs much larger than 100 μm.

APPENDIX B. CALCULATION OF DOSE RATES

Alpha Radiation

Provided that the organ or animal is larger than the size given in Table 1, we assume that at least 90% of the alpha energy is dissipated internally. In SI units, one MeV is 1.6×10^{-13} J. One Bq kg^{-1} is 3.15×10^{7} dpy and the annual dose rate in mSv y^{-1} is thus $5.04 \times 10^{-3}\ E_\alpha \times Bq\ kg^{-1} \times RBE$.

We have assumed a value of 20 for the relative biological effectiveness for alpha particles (and of unity for beta and gamma rays). Thus the dose rates to fish organs and organisms in the sediments are given by

m

$$\cancel{M}Sv\ y^{-1} = 0.101\ E_\alpha \times Bq\ kg^{-1}$$

It was assumed that the radiation field in the sediments was homogeneous throughout any organisms therein.

Beta Radiation

The most energetic beta particle emitted in the radium decay chain is that from ^{214}Bi, and it has a range of almost one cm. The only compartment which receives a beta particle dose comparable to that from alpha particles is the sediments and any organisms living there. Even near the sediment-water interface, where half the radiation escapes, the internal dose rate to the sediments is about $5\times10^{-3} \times 1/2 \times 1/3\ E_{\beta} \times \mathrm{Bq\ kg^{-1}}$. (The average beta energy is about one third the maximum energy) so that

$$\mathrm{mSv\ y^{-1}} = 8 \times 10^{-4}\ E_{\beta} \times \mathrm{Bq\ kg^{-1}}$$

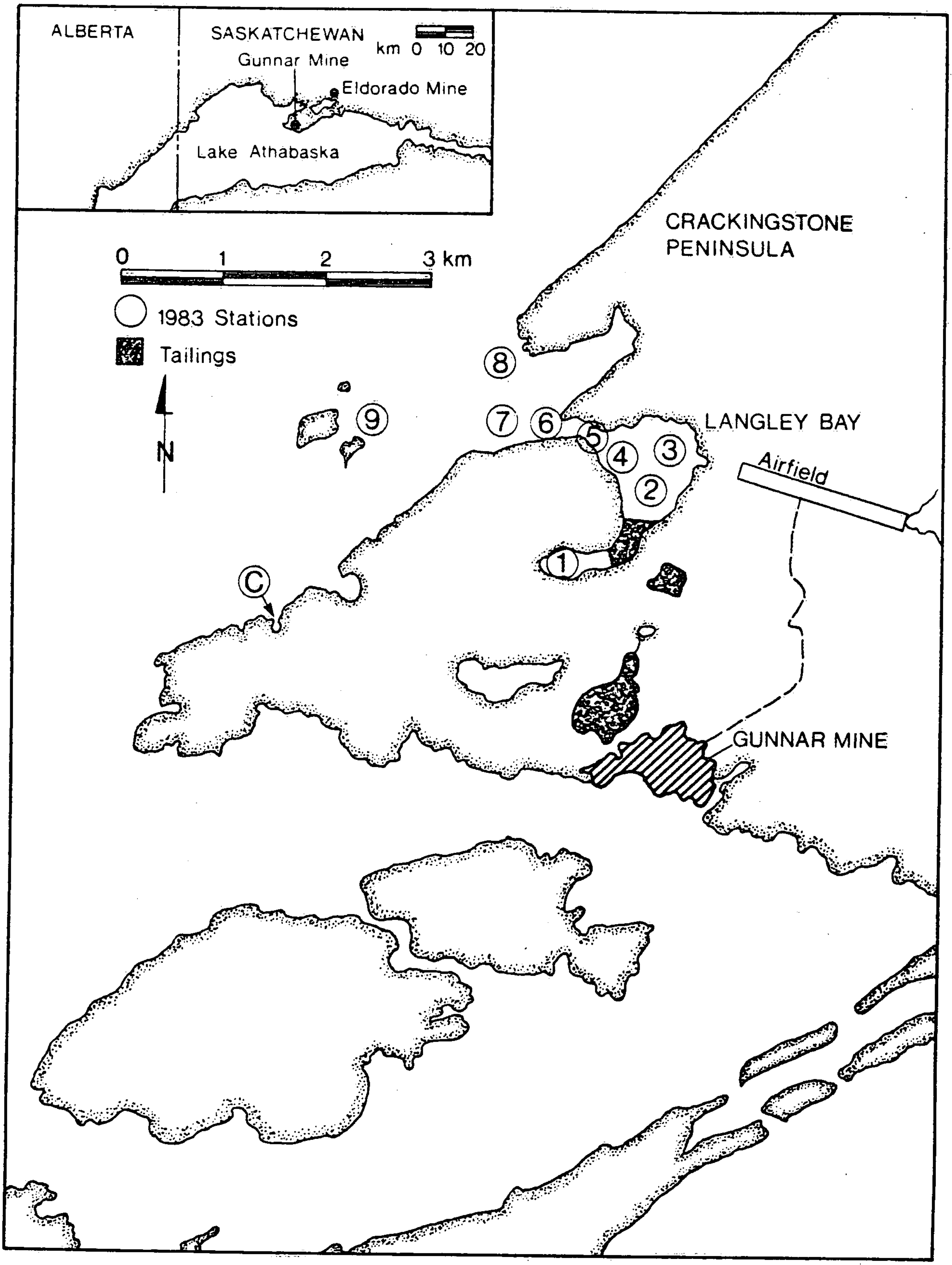
The gamma radiation contributed less than 1% to the total dose rate in the sediments, but it did however contribute a calculable whole body dose rate to fish within a few cm of the bottom. The dose rate was calculated by assuming the bottom to be a plane, collimated source of gamma rays. A thin source will radiate half its energy to the overlying water and half of that will be absorbed in a few cm. Thus, the water dose near the bottom will be given by the expression

$$\mathrm{mSv\ y^{-1}} \approx 10^{-3}\ E_{\gamma} \times \mathrm{Bq\ kg^{-1}}$$

FIGURES

Fig. 1 The Gunnar Uranium Mine and associate tailings system, Lake Athabaska, Saskatchewan. c indicates the control site.

Fig. 2 Dose rates to organisms in the Langley Bay area as a function of their size and location.



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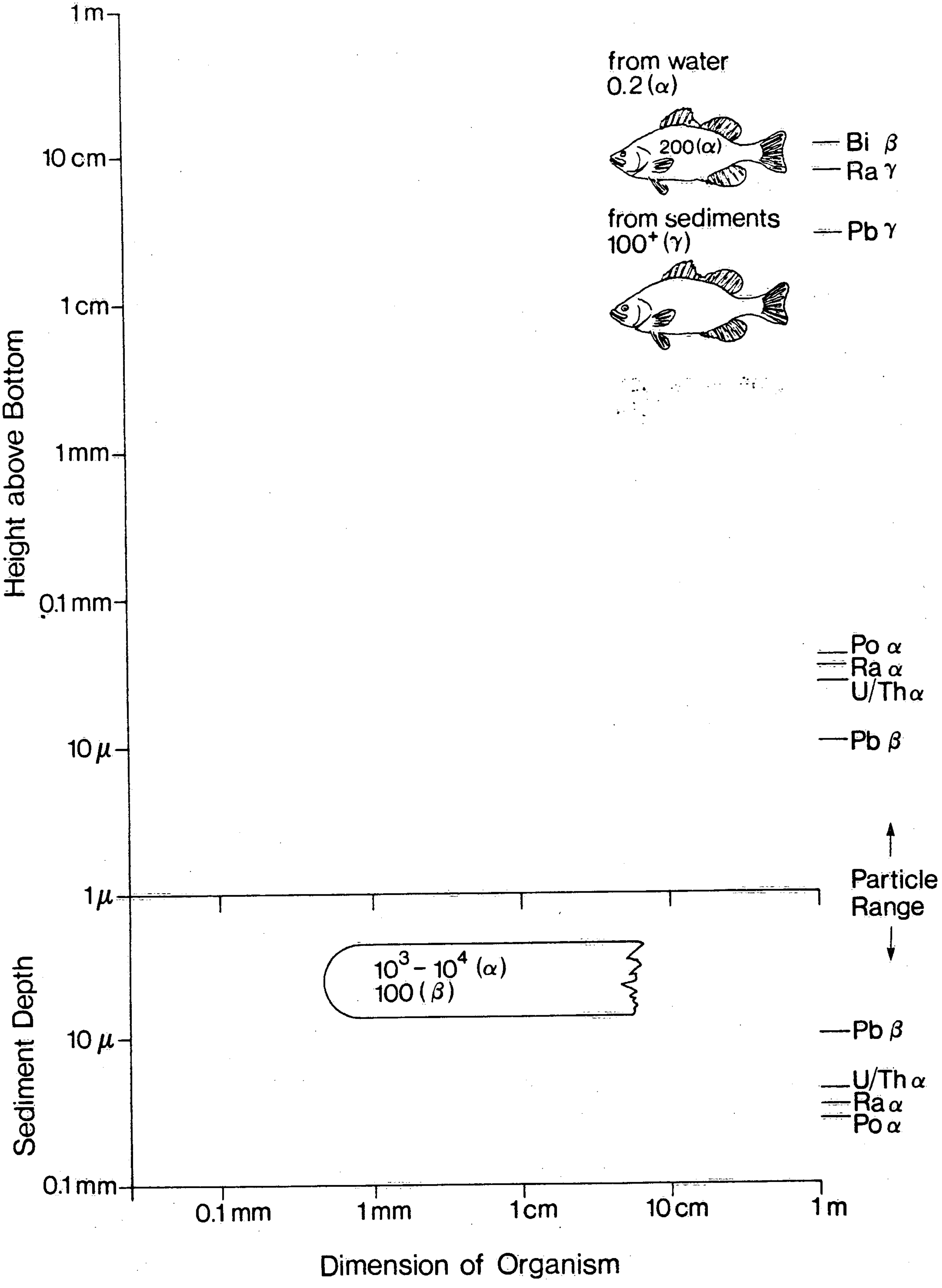

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