Radioisotope Geochemistry
An Introduction

- Variety of naturally occurring and anthropogenic radioisotopes can be used to provide information on a wide variety of processes
- Can be used as tracers in same manner as stable isotopes but have the advantage of **time** as a parameter
- Their use requires certain assumptions be made – must be assessed on a case by case basis

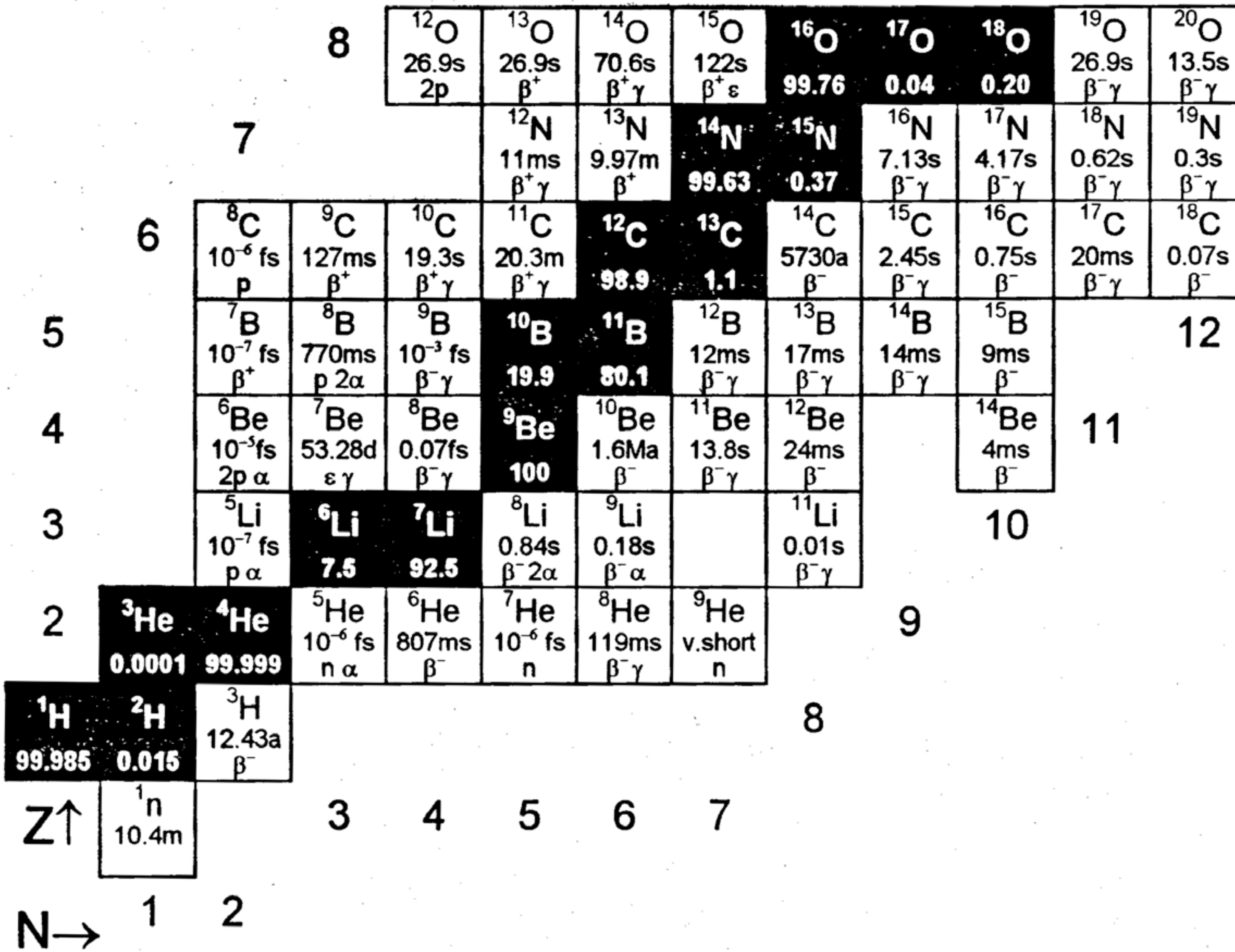


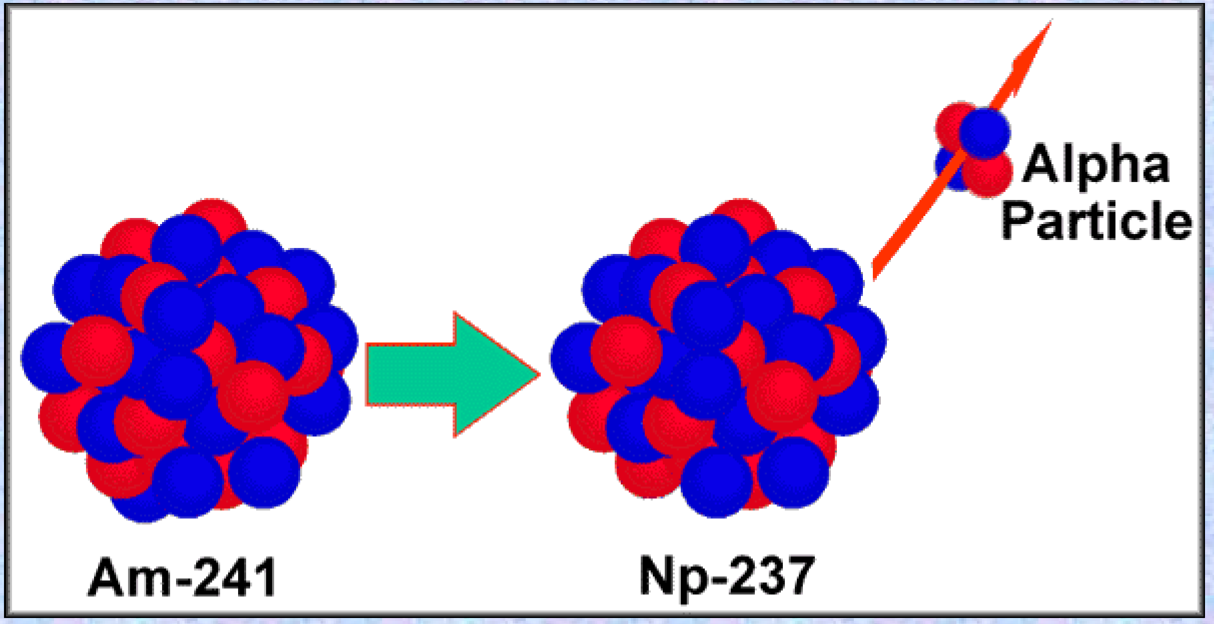
Fig. 1-2 Chart of the light element isotopes showing percent abundances of the stable isotopes (shaded black) and half-lives of radioisotopes (s = second, m = minute, d = day, a = year) with their principal and secondary decay modes, where α = alpha emission (2p and 2n), ε = electron capture, β^- = electron (beta) emission, β^+ = positron, γ = gamma emission, n = neutron emission, p = proton emission (after General Electric Ltd., 1989).

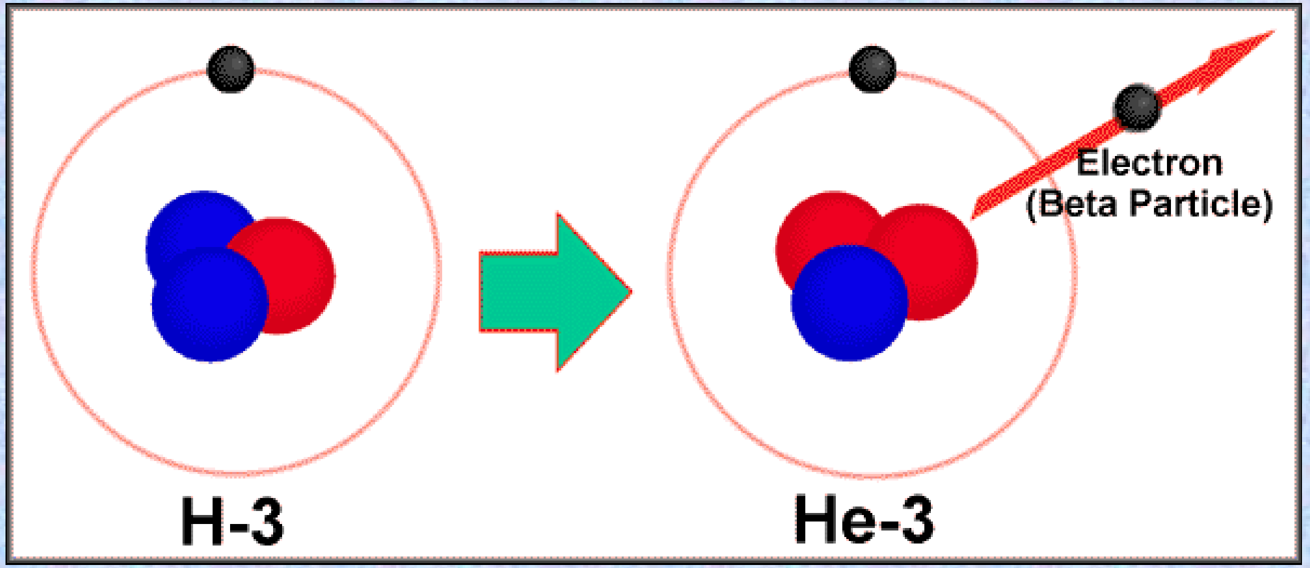
Examples of Processes

- Water Column Vertical and Horizontal Transport by
  - Mixing
  - Particle Scavenging and Sedimentation
  - Lateral advection
- Sediment Accumulation & Dating
  - Accumulation Rates
  - Mixing
  - Resuspension

Modes of Decay

- Alpha(α) – emission of two neutrons and two protons together as helium ion from nucleus
- Beta (β) – emission of electron *from nucleus* resulting in increase in atomic number of 1 in original nucleus
- Gamma (γ) – emission of photons from excited nuclei


Alpha
Particle
Am-241
Np-237


Electron
(Beta Particle)
H-3
He-3

Basic Equations of Decay

$$N = N_o e^{-\lambda t}$$

Where N_o = number of atoms at $t = 0$

N = number of atoms at time = t

λ= first order rate (decay) constant

λN = Activity (Bq (= dps) or dpm unit used)

Decay Equations (cont.)

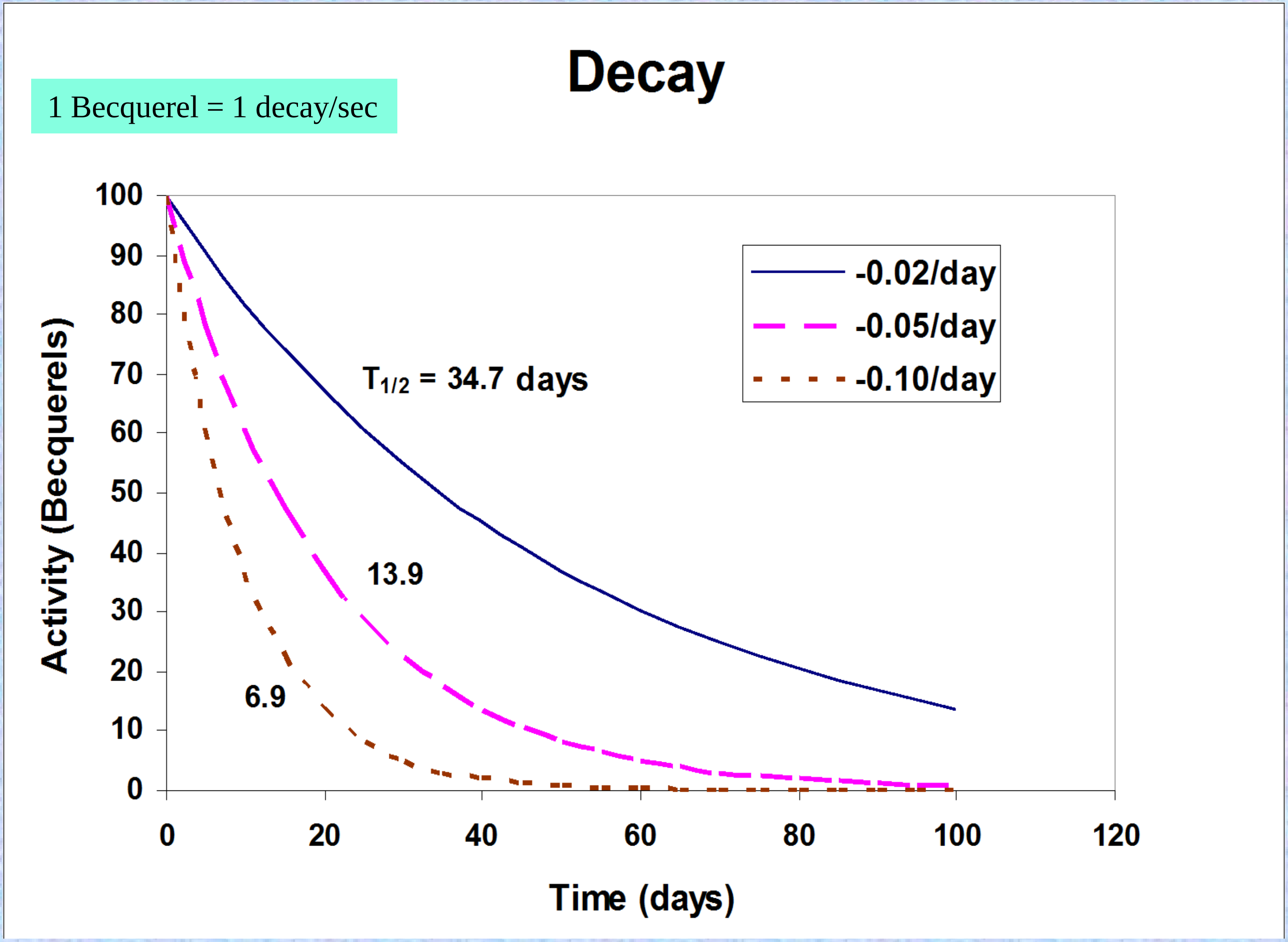
$$\ln N = \ln N_o - \lambda\, t$$

When $N = ½\, N_o$

$$t = t_{1/2}$$

i.e. $\ln 0.5 = -\lambda t_{1/2}$

$$t_{1/2} = 0.693/\lambda$$



Decay
1 Becquerel = 1 decay/sec
Activity (Becquerels)
Time (days)
-0.02/day
-0.05/day
-0.10/day
T1/2 = 34.7 days
13.9
6.9
100
90
80
70
60
50
40
30
20
10
0
0
20
40
60
80
100
120

Classes of Radioisotopes

1. Primordial
   - Parents produced by super-novae, long-lived
   - not produced on earth
   - e.g. U-series
2. Cosmogenic
   - Produced by interaction of cosmic rays with atoms in the atmosphere or land surface
   - Short to long-lived
   - e.g. ^{14}C
3. Artificial
   1. Man-made
   2. Purposefully or incidentally (nuclear bombs)
   3. e.g. ^{239}Pu

Production of ^{14}C in Atmosphere

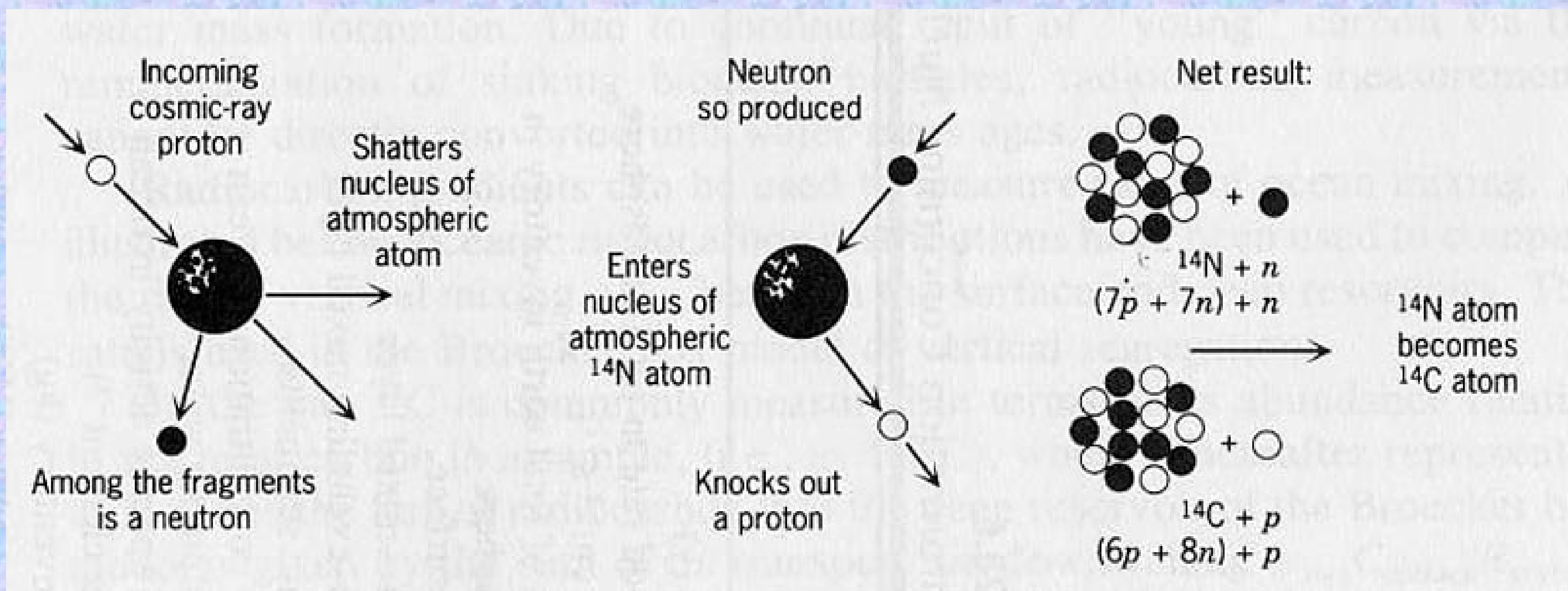
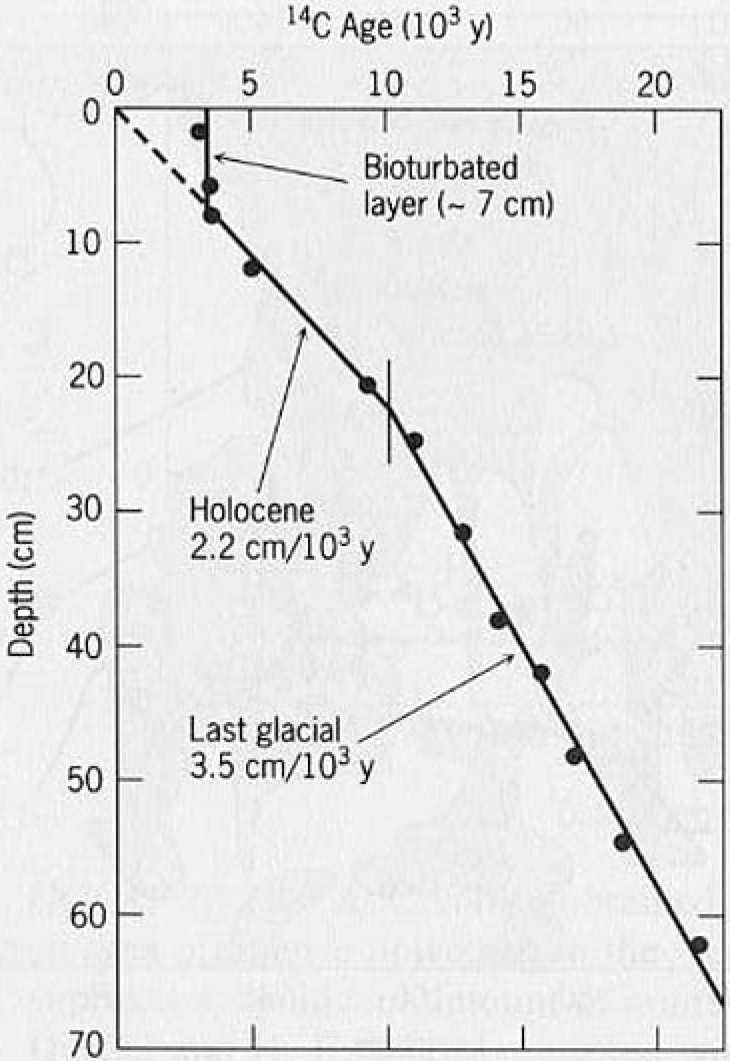


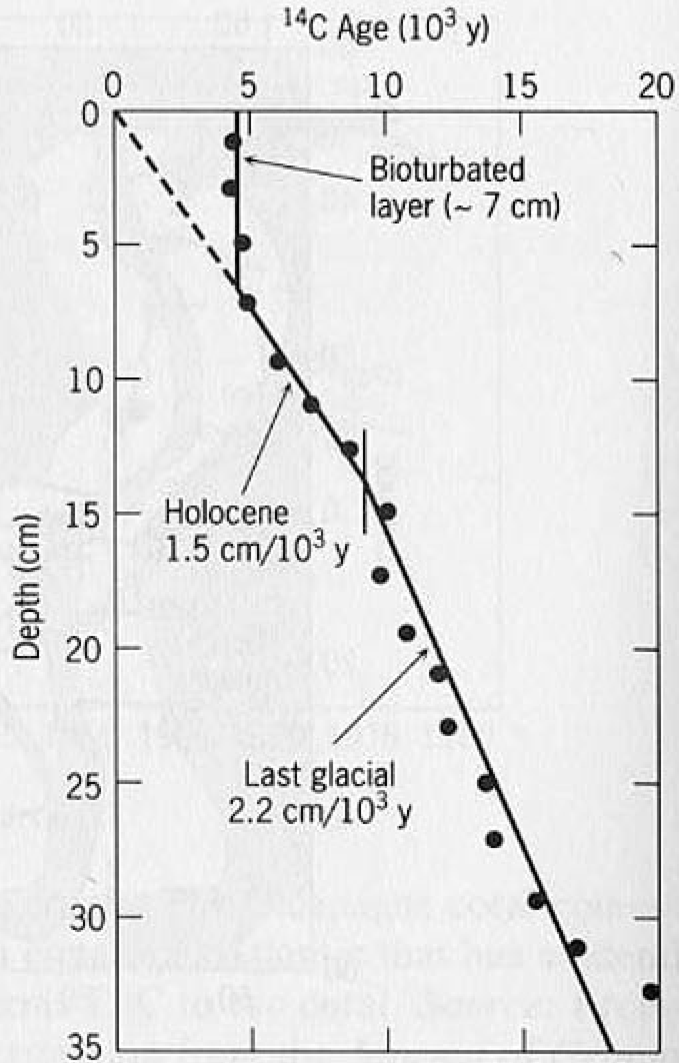
TABLE 28.6
Basic Information Concerning Nuclides Produced by Cosmic Rays

	Nuclide					
	^{3}H	^{7}Be	^{10}Be	^{14}C	^{26}Al	^{32}Si
Half-life (y)	12.3	0.145	2.5×10^{6}	5680	7.4×10^{5}	500
Production rate in total atmosphere (atom cm^{-2} s^{-1})	0.25	0.081	0.045	2.5	1.4×10^{-4}	1.6×10^{-4}
Fraction of total earth inventory in						
Atmosphere	0.072	0.71	3.9×10^{-7}	0.019	1.4×10^{-6}	2.0×10^{-3}
Land surface	0.27	0.08	0.29	0.04	0.29	0.29
Ocean—mixed layer	0.35	0.2	5.7×10^{-6}	0.022	1.4×10^{-5}	0.0035
Ocean–excluding mixed layer	0.3	0.002	10^{-4}	0.92	7×10^{-5}	0.68
Oceanic sediments	0	0	0.71	0.004	0.71	0.028
Average concentration in ocean (10^{-3} dpm kg $water^{-1}$)	36	—	10^{-3}	260	1.2×10^{-5}	2.4×10^{-2}
Average specific activity in ocean (dpm g $element^{-1}$)	3.3×10^{-4}	—	1600	10	0.0012	0.008
Global inventory (kg)	3.5	3.2×10^{-3}	4.3×10^{5}	7.5×10^{4}	1.1×10^{3}	1.4
Global inventory (MCi)	35	1.1	6.4	340	0.020	0.023

Source: From *Chemical Oceanography*, vol. 3, J. D. Burton (eds: J. P. Riley and G. Skirrow), copyright © 1975 by Academic Press, Orlando, FL, p. 140. Reprinted by permission. Data from *The Handbook of Physics, 2E*, E. U. Condon and H. Odishaw, copyright © 1967 by McGraw–Hill, Inc., New York, pp. 9.277, 9.285, 9.319. Reprinted by permission.



^{14}C Age (10^3 y)
0
5
10
15
20
Depth (cm)
0
10
20
30
40
50
60
70
Bioturbated
layer (~ 7 cm)
Holocene
2.2 cm/10^3 y
Last glacial
3.5 cm/10^3 y



^{14}C Age (10^3 y)
0
5
10
15
20
Depth (cm)
0
5
10
15
20
25
30
35
Bioturbated
layer (~ 7 cm)
Holocene
1.5 cm/10^3 y
Last glacial
2.2 cm/10^3 y

Hughen et al. Science 2004; Cariaco Basin sediment ^{14}C

1) Annual laminations in this anoxic basin allows for ‘tree-ring’ year counting for much of the record

2. Difference between calendar year and ^{14}C age due to:

– ^{14}C production rate in atmosphere

–Deep mixing rate

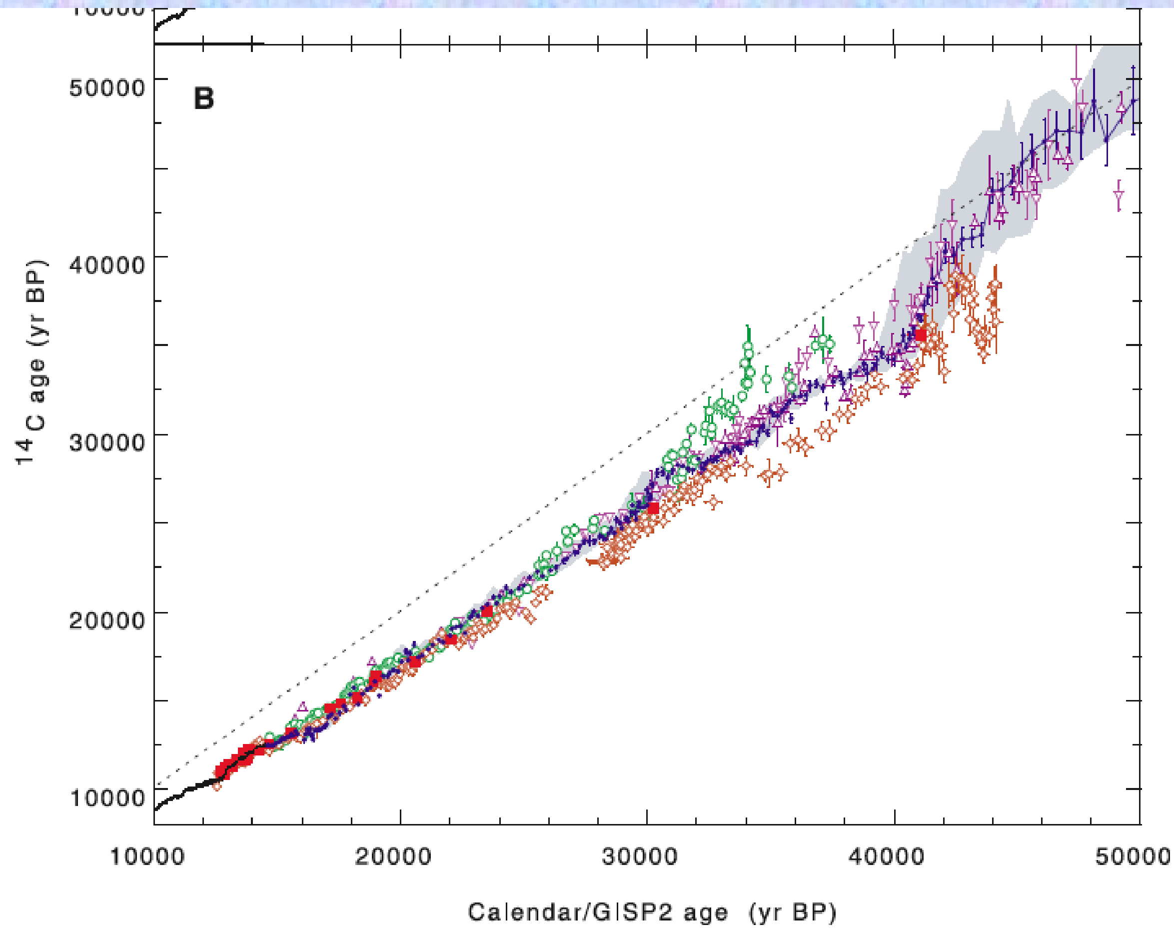


Fig. 2. Radiocarbon calibration data from various sources. (**A**) Calibration data from Cariaco leg 165, holes 1002D and 1002E (blue circles), plotted versus GISP2 calendar age (*12*) assigned by correlation of detailed paleoclimate records (*17*) (SOM Text and fig. S2). The thin black line is high-resolution calibration data from Intcal98 tree rings (*2*, *3*) joined at ~12 cal. ka B.P. to the Cariaco PL07-58PC varve chronology (*13*). Red squares are paired ^{14}C-U/Th dates from corals (*5*). Replicate measurements, including overlap between 1002D and 1002E, have been averaged. Light gray shading represents the Cariaco calibration curve shifted within limits of calendar age uncertainty. Dashed line shows equal ^{14}C-calendar ages. Error bars are 1 σ. (**B**) Cariaco site 1002 data set plotted versus other published ^{14}C calibration data. Symbols are the same as above, with additional data from Lake Suigetsu varves (*6*) (open circles), Bahama speleothem U/Th (*7*) (open diamonds), and North Atlantic cores PS2644 (*9*) (upside-down triangles) and SO82-5 (*10*) (triangles) correlated to GISP2. Error bars for all records are 1 σ.

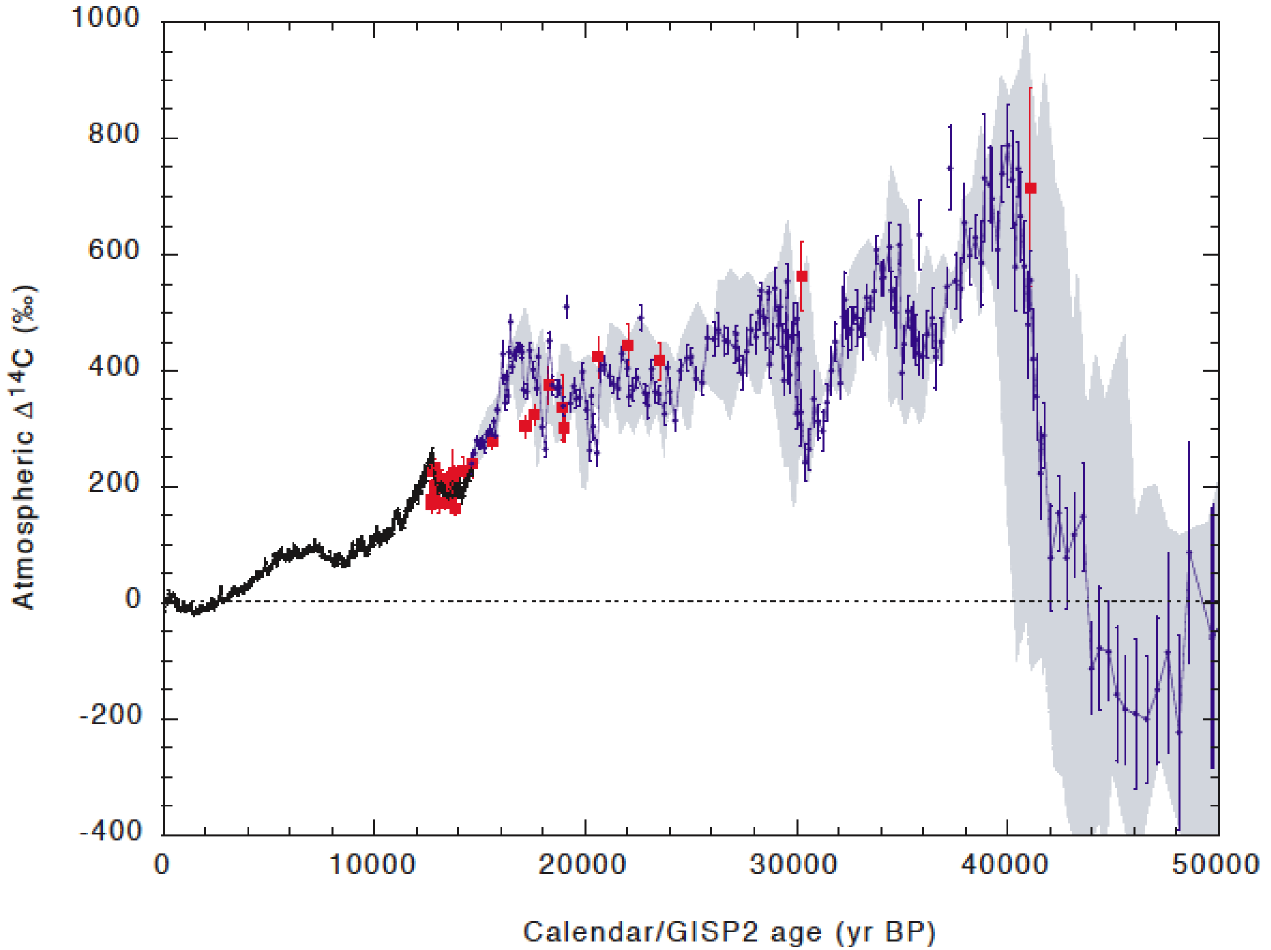
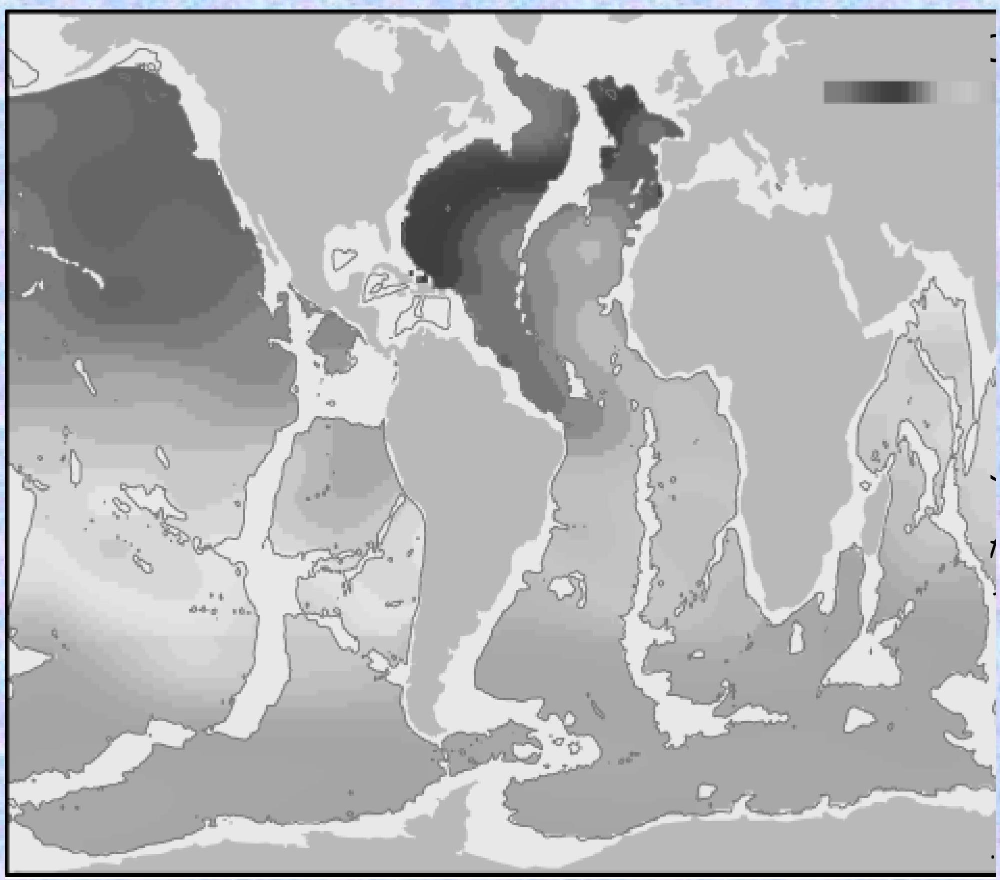
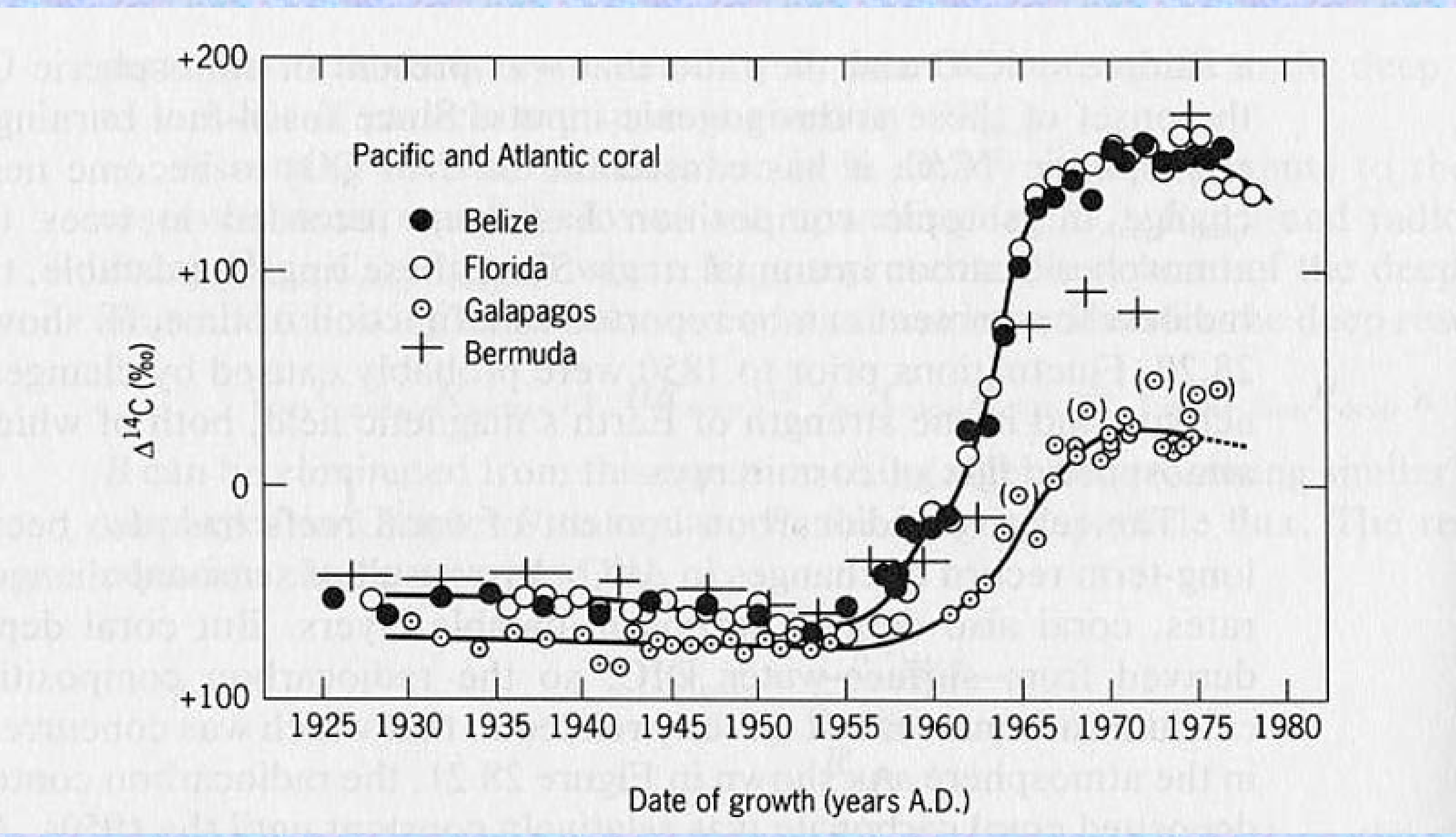


Fig. 3. Atmospheric $\Delta^{14}C$ for the past 50 cal. ka B.P. Symbols are the same as those in Fig. 2A. Cariaco error bars represent independent uncertainty in $\Delta^{14}C$ due to 1-σ ^{14}C age error. Light gray shading shows additional uncertainty in Cariaco $\Delta^{14}C$ due to calendar-age error that is not independent from sample to sample, but rather would shift sections of the curve together within the limits of the shading. Dotted line is modern preindustrial atmospheric $\Delta^{14}C$, defined as 0‰. Upper and lower limits were determined by adding and subtracting 1–σ errors to the calendar age and recalculating $\Delta^{14}C$ with the use of the new calendar ages.



th of
3000 m, determined during the
WOCE program in the 1990s.
Courtesy of Robert Key, Princeton
University; Key *et al.* (2004). (See

^{14}C also an 'artificial' radioisotope as a result of nuclear bomb testing



U-Th Decay Series

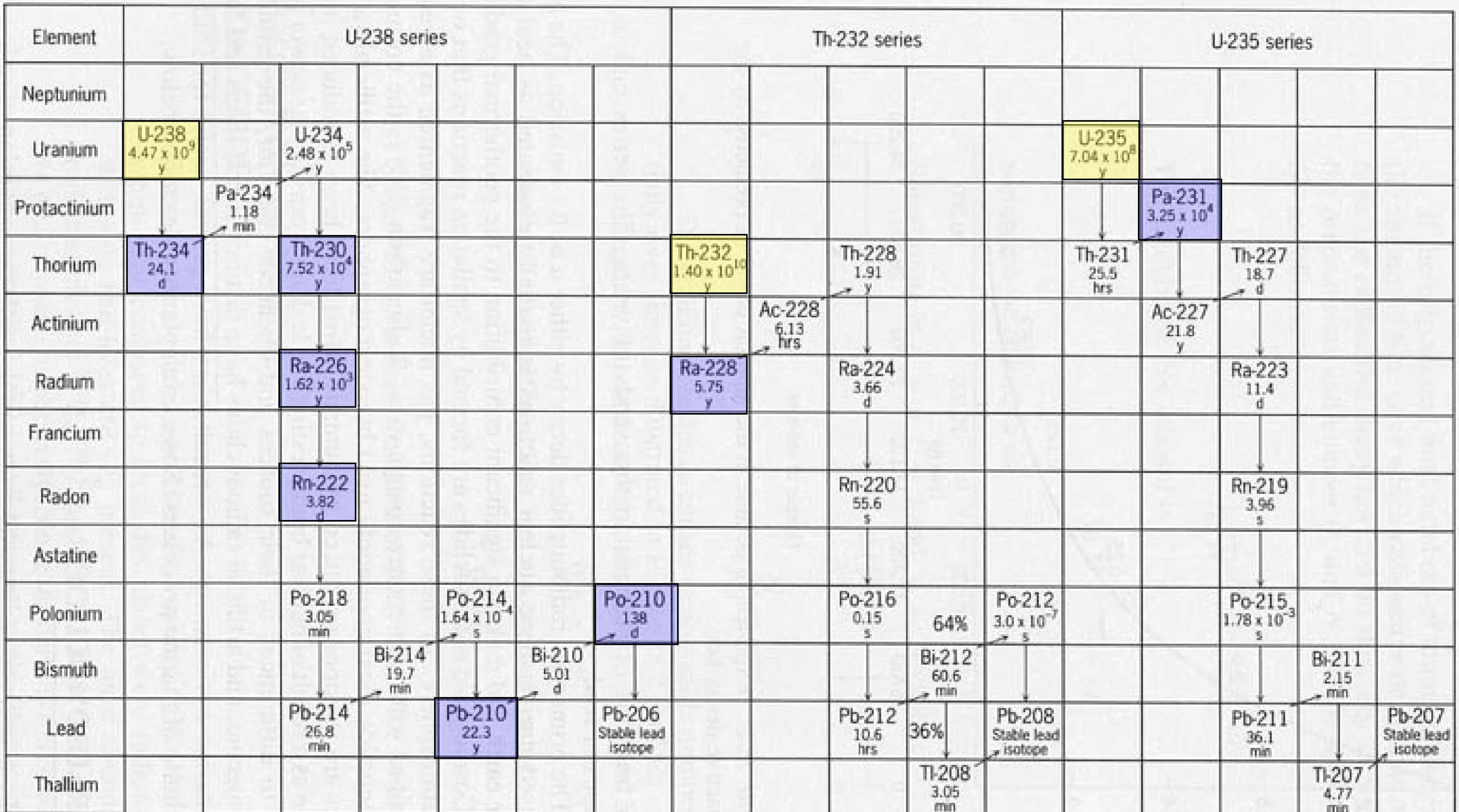
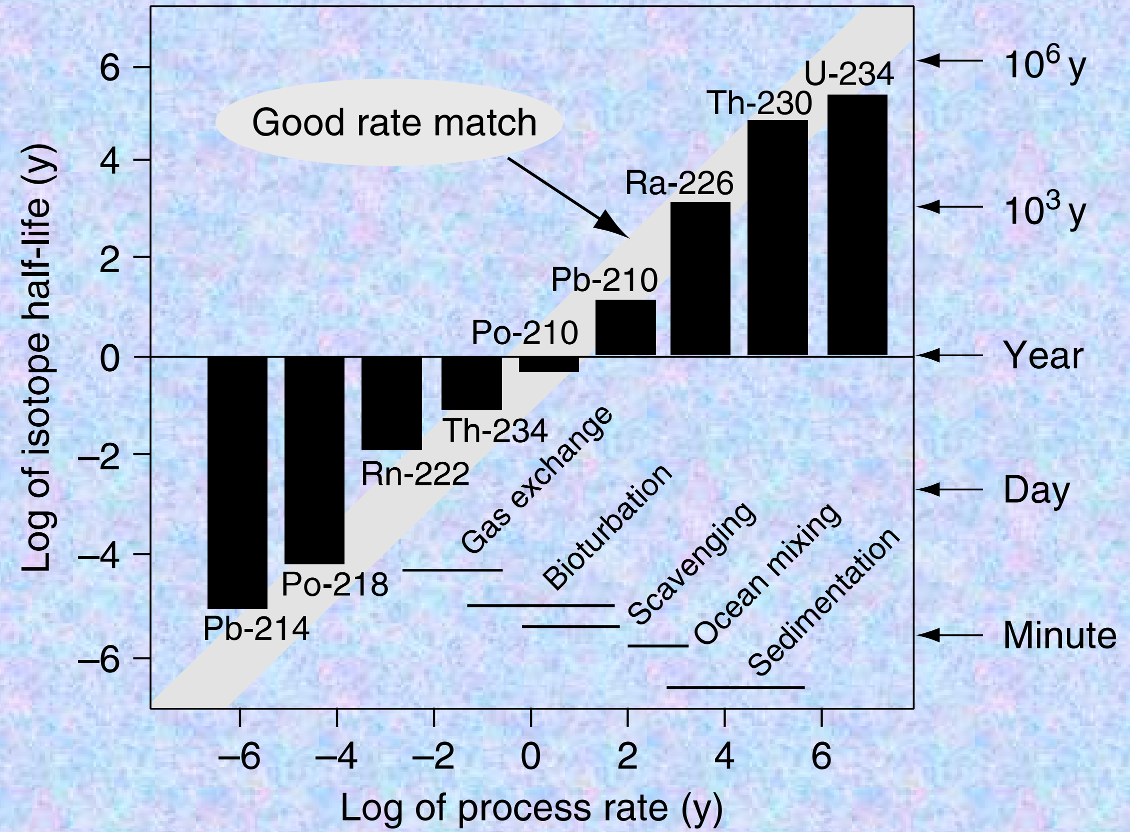


Figure 5.19. The relation between the half life of a radioisotope (ordinate) and the characteristic timescale for marine processes (abscissa). The shaded area indicates the range where the two lifetimes are a good match.



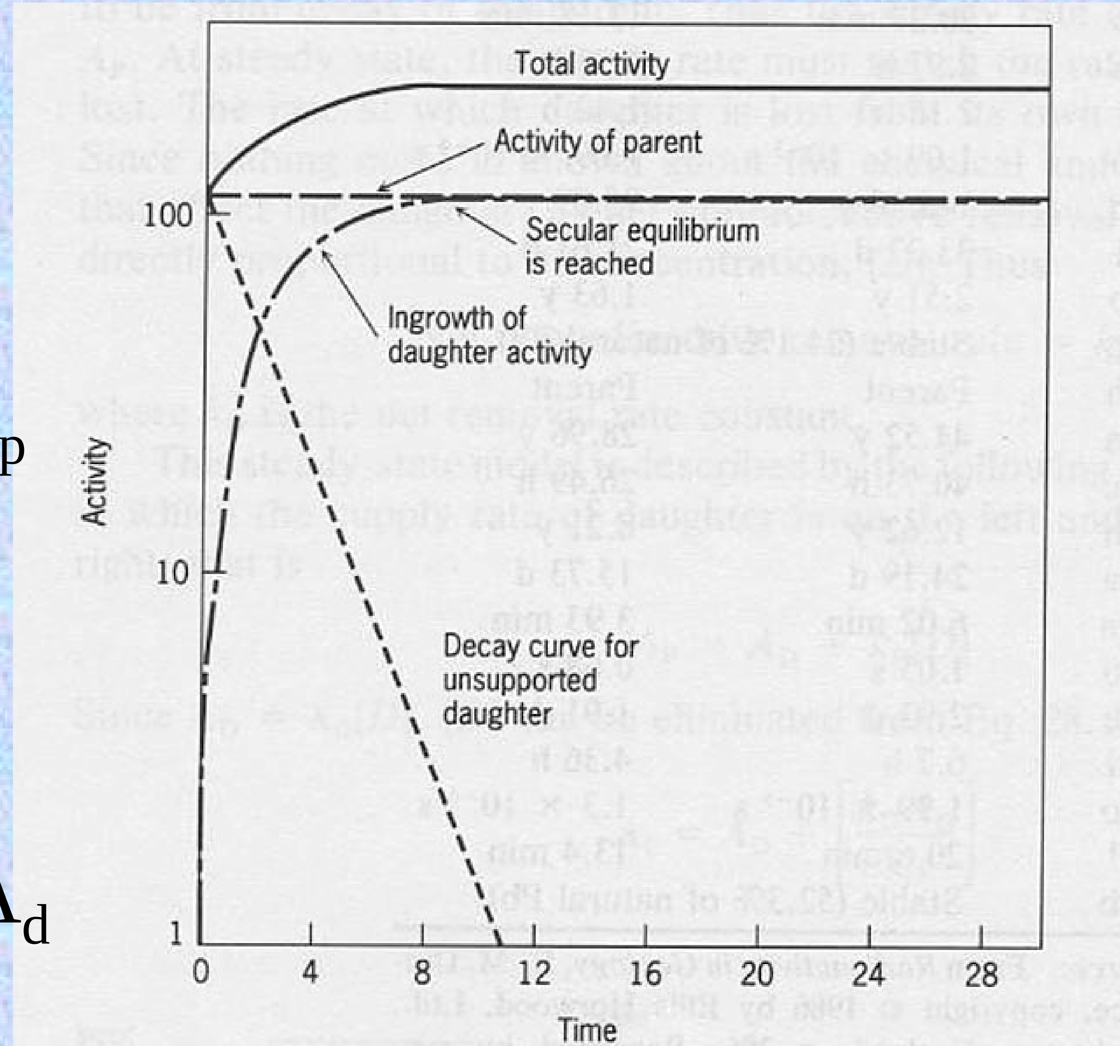
Secular Equilibrium

P

$\lambda_P[P] = A_p$

D

$\lambda_D[D] = A_d$



At steady state; $\lambda_P[P] = \lambda_D[D]$ or $A_p = A_d$

Scavenging

P

[e.g soluble long-lived ^{238}U with constant A in the Ocean]

$\lambda_P[P] = A_p$

D

Scavenging (F)

$F_D[D] = F$

[e.g adsorption of Th onto sinking particles]

$\lambda_D[D] = A_d$

At steady state; $A_p = A_d + F$

Scavenging and Decay

- Both first order processes
- If decay dominant then $A_P = A_D$
- Where both are important $A_D < A_P$
- Under steady state conditions the activity ratio can be used to estimate the scavenging rate and hence scavenging residence time

Equations for Scavenging Rate

$$\lambda_P[P] = \lambda_D[D] + F_{\mathbf{D}}\mathbf{[}D\mathbf{]}$$

$$A_D/A_P = \lambda_D/(\lambda_D + F_{\mathbf{D}})$$

Where λ_P = decay constant for parent

λ_D = decay constant for daughter

[ ] = atom concentration

$F_{\mathbf{D}}$ = scavenging rate constant

Equations for Scavenging Rate (cont.)

Solving for $F_{\mathbf{D}}$:

$$F_{\mathbf{D}} = [(1 - A_D/A_P)/(A_D/A_P)] \quad \lambda_D$$

or

$$\tau_{1/2} = [(A_D/A_P)/(1 - A_D/A_P)] \, t_{D_{1/2}}$$

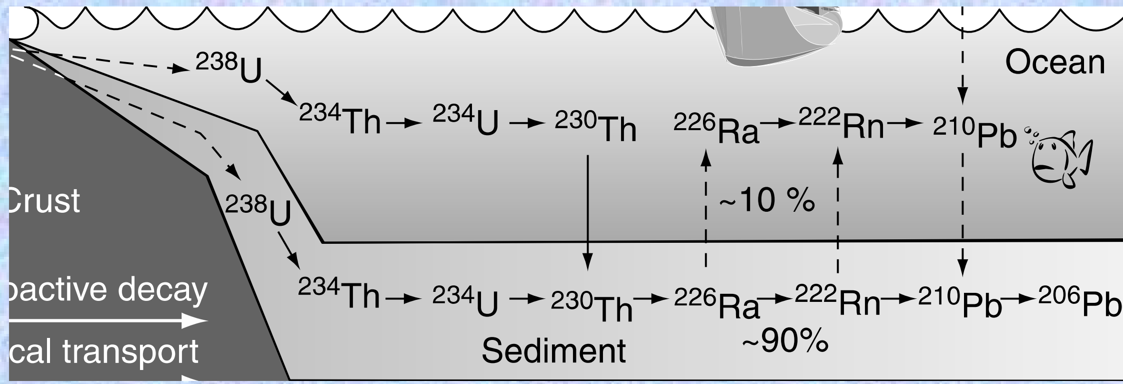
Where $\tau_{1/2}$ = scavenging "half-life"

$t_{D_{1/2}}$ = half-life of daughter

Table 4-4. Typical activity ratios for daughter-parent pairs in various water types.

	Estuaries	Coastal	Surface Ocean	Deep Sea
$^{210}Pb/^{226}Ra$	-	-	>1*	0.4-1.0
$^{230}Th/^{234}U$	-	-	$<3 \times 10^{-5}$	3×10^{-4}
$^{228}Th/^{228}Ra$	0.01	0.05	0.2	0.5-1.0
$^{234}Th/^{238}U$	0.2	0.6	>0.9	~ 1
$^{231}Pa/^{235}U$	-	-	-	2×10^{-3}
$^{210}Po/^{210}Pb$	-	-	0.5	1.0

*Although ^{210}Pb is being removed from surface water by particles, it has an additional source. Radon atoms escaping to the atmosphere from continental soils decay to ^{210}Pb. These ^{210}Pb atoms are incorporated into aerosols and are brought back to the earth's surface by rain and aerosol impact. The flux of these atoms to the sea surface exceeds by about a factor of 10 the in situ production by radiodecay of ^{226}Ra in the upper 200 meters of the ocean.



4). See also Table 5.5.

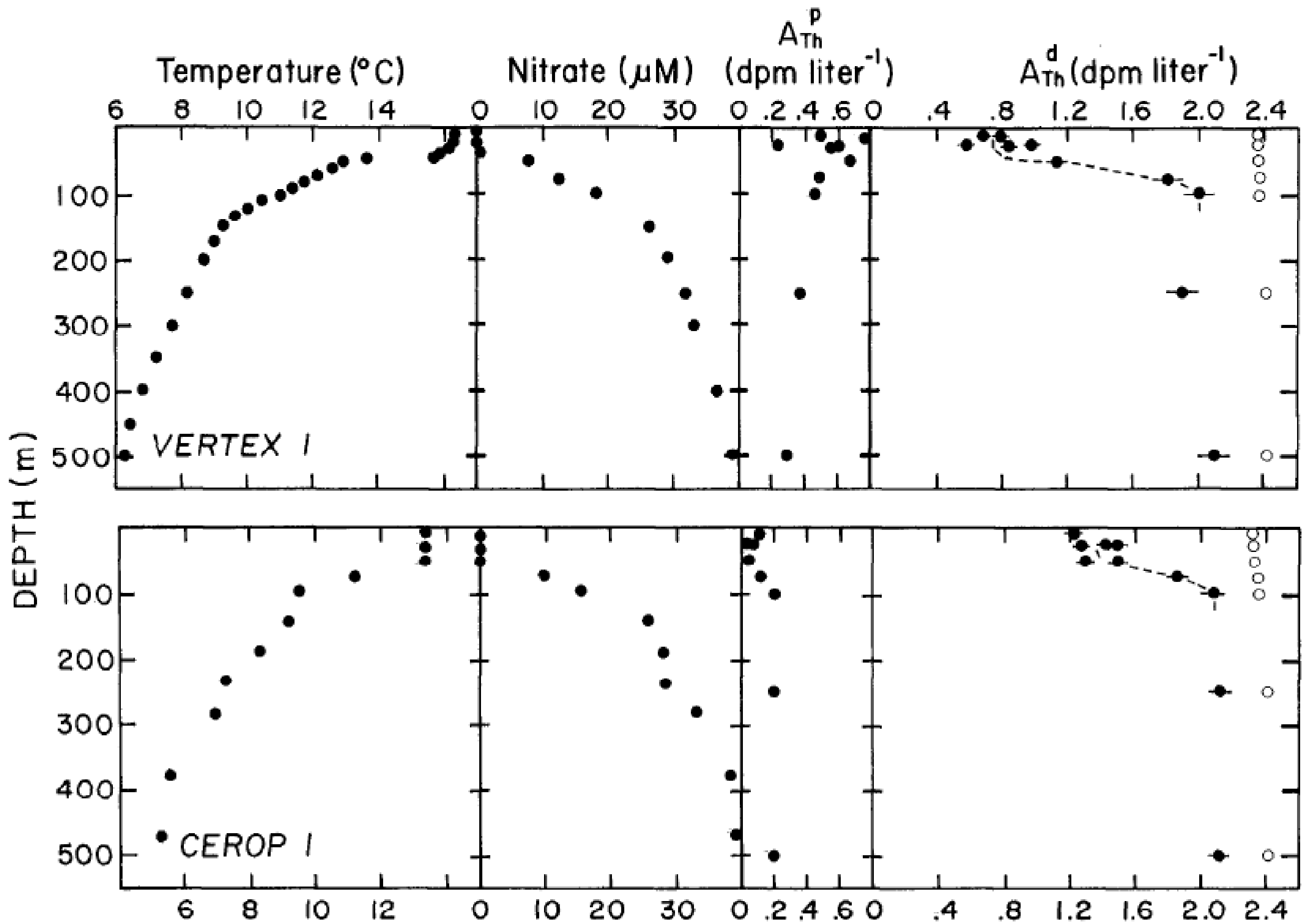


Fig. 2. Vertical profiles of temperature, nitrate, particulate and dissolved ^{234}Th activity (A^p_{Th} and A^d_{Th}) for VERTEX I and CEROP I, II, and III. ^{238}U activity—O.

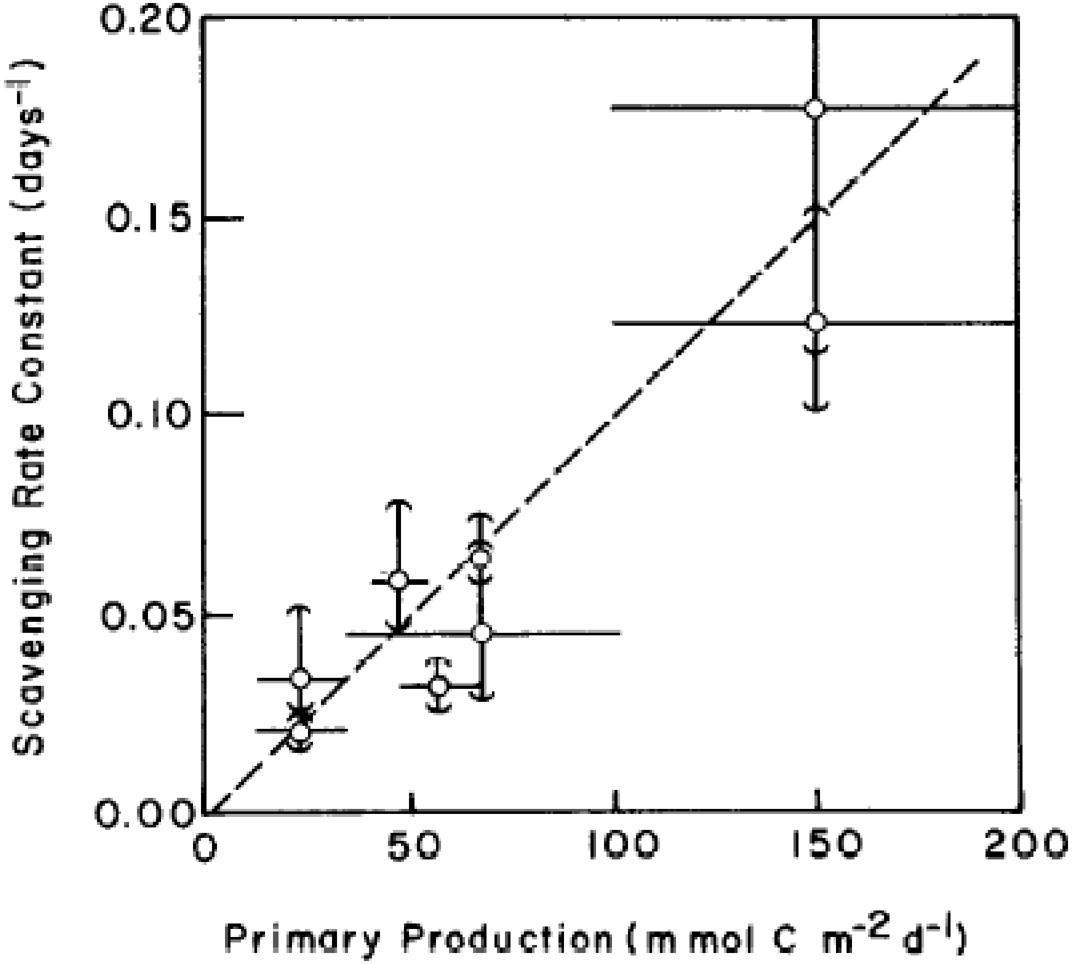
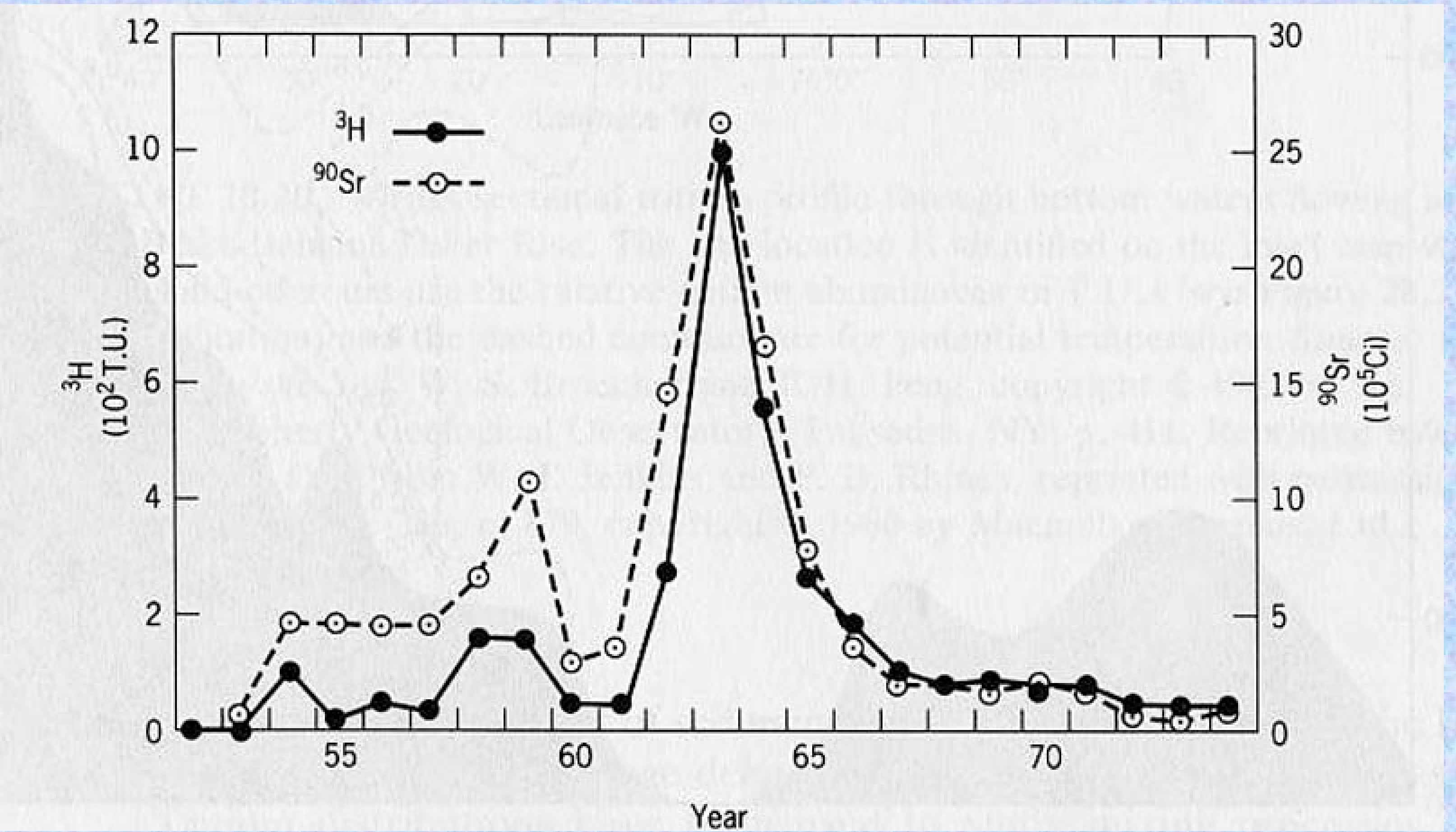
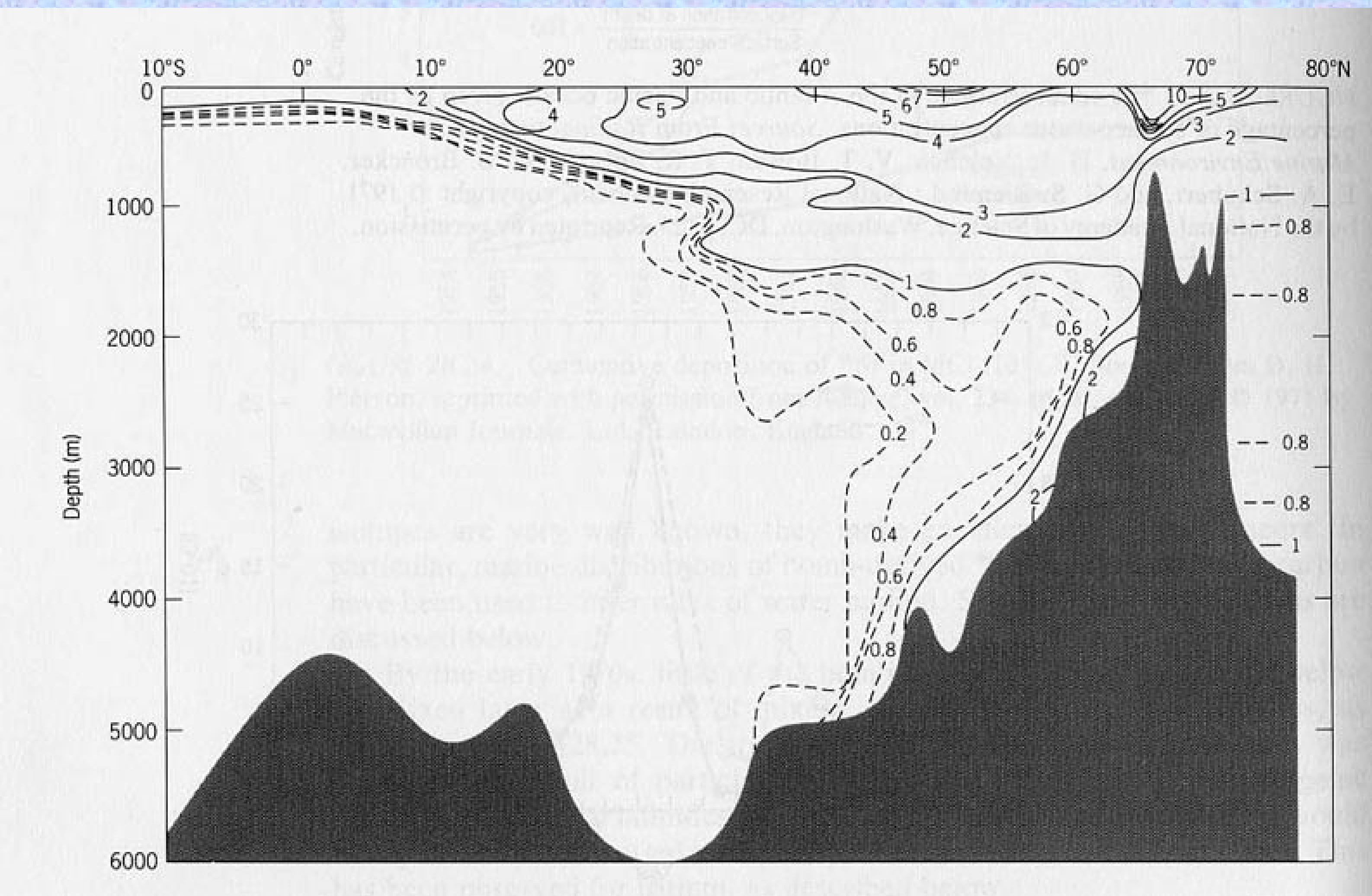


Fig. 3. Model-derived first-order dissolved ^{234}Th scavenging rate constant vs. primary production.



^3H
^{90}Sr
^3H (10^2 T.U.)
^{90}Sr (10^5Ci)
Year
0
2
4
6
8
10
12
0
5
10
15
20
25
30
55
60
65
70



10°S
0°
10°
20°
30°
40°
50°
60°
70°
80°N
Depth (m)
0
1000
2000
3000
4000
5000
6000

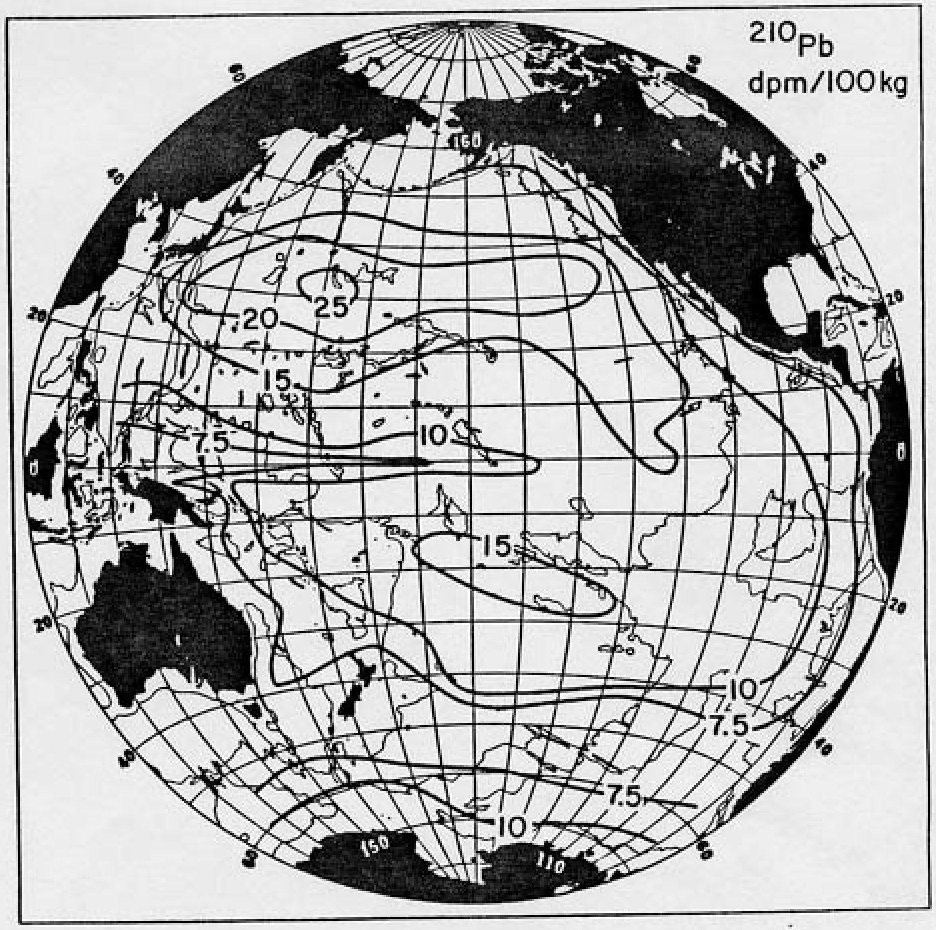


Figure 4-16. Map showing the distribtuion of ^{210}Pb concentrations (in dpm/100 kg) in the surface waters of the Pacific Ocean. The results were summarized by Nozaki, Thompson and Turekian (151).