



Radioactivity in the Marine Environment

Uranium-Thorium Decay Series

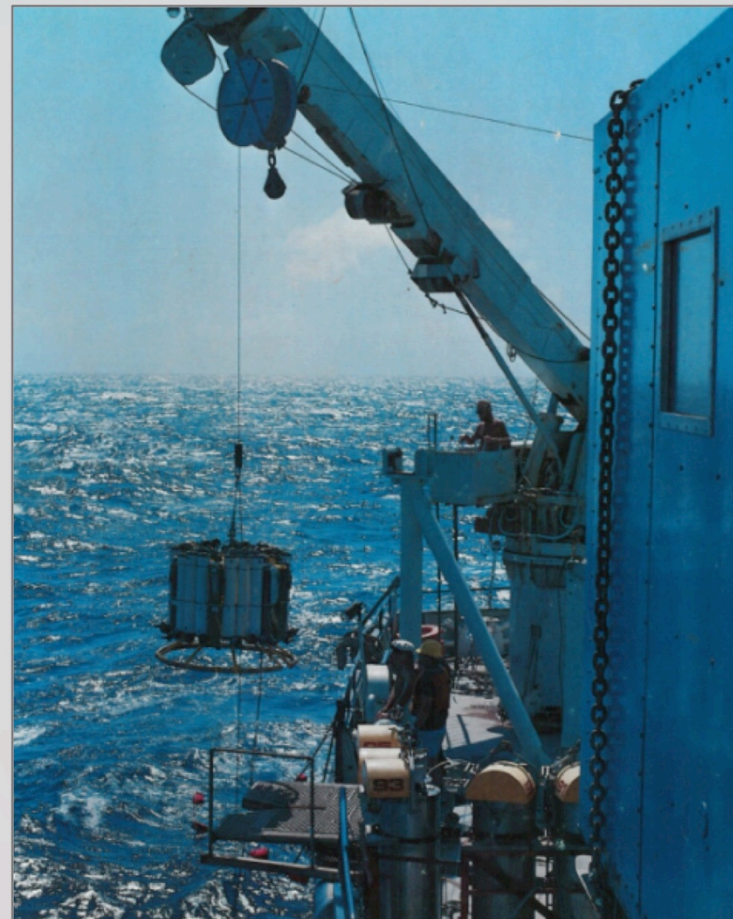
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Radionuclides in the Marine Environment

Radionuclides, of both natural and anthropogenic origins, can be used as **CLOCKS** of key processes (age and/or rates) in the oceans, mainly because they:

- Are ubiquitous in all compartments of the oceans
- Have a large range of half-lives (from seconds to billions of years)



Tracers in the Sea
Broecker and Peng, 1982

Three main sources of radionuclides to the Marine Environment:

1. **U-Th series radionuclides** – of primordial origin, occur naturally on land and in ocean, and produce a series of “daughter” radionuclides via radioactive decay.

Examples: ^{238}U , ^{234}Th , ^{232}Th , ^{210}Pb , $^{223, 224, 226, 228}\text{Ra}$ and ^{222}Rn .

2. **Cosmogenic Radionuclides** – continuously being created by cosmogenic rays that interact with materials in the atmosphere and on Earth.

Examples: ^3H , ^{14}C , ^7Be

3. **Anthropogenic radionuclides** – continuously being produced by humans since the 1940s.

Examples: ^3H , ^{14}C , ^{90}Sr , ^{137}Cs , ^{129}I , $^{238, 239, 240}\text{Pu}$

Note: Some radionuclides have both cosmogenic and anthropogenic sources (e.g., ^3H and ^{14}C).

The case of the Radioactive Daughter

For the naturally occurring radionuclides: ^{238}U , ^{235}U , and ^{232}Th , the half-lives of the parent nuclides are *much, much longer* than their daughter products.

Element	U-238 Series					Th-232 Series			U-235 Series			
Uranium	U-238 4.5×10^9 y		U-234 245500 y							U-235 7.0×10^8 y		
Protactinium		Pa-234 1.2 m									Pa-231 32800 y	
Thorium	Th-234 24.1 d		Th-230 75400 y			Th-232 1.4×10^{10}		Th-228 1.91 y		Th-231 25.5 h		Th-227 18.7 d
Actinium							Ac-228 6.1 h				Ac-227 21.8 y	
Radium			Ra-226 1600 y			Ra-228 5.75 y		Ra-224 3.7 d				Ra-223 11.4 d
Francium												
Radon			Rn-222 3.8 d									
Astatine												
Polonium			Po-218 3.1 m		Po-214 0.00014 s							
Bismuth				Bi-214 19.9 m		Bi-210 5.0 d						
Lead			Pb-214 26.8 m		Pb-210 22.3 y	Pb-206 stable		Pb-208 stable				Pb-207 stable

α Decay
↓ Z: - 2
↓ N: - 4

β Decay
↘ Z: + 1
↘ N: ± 0

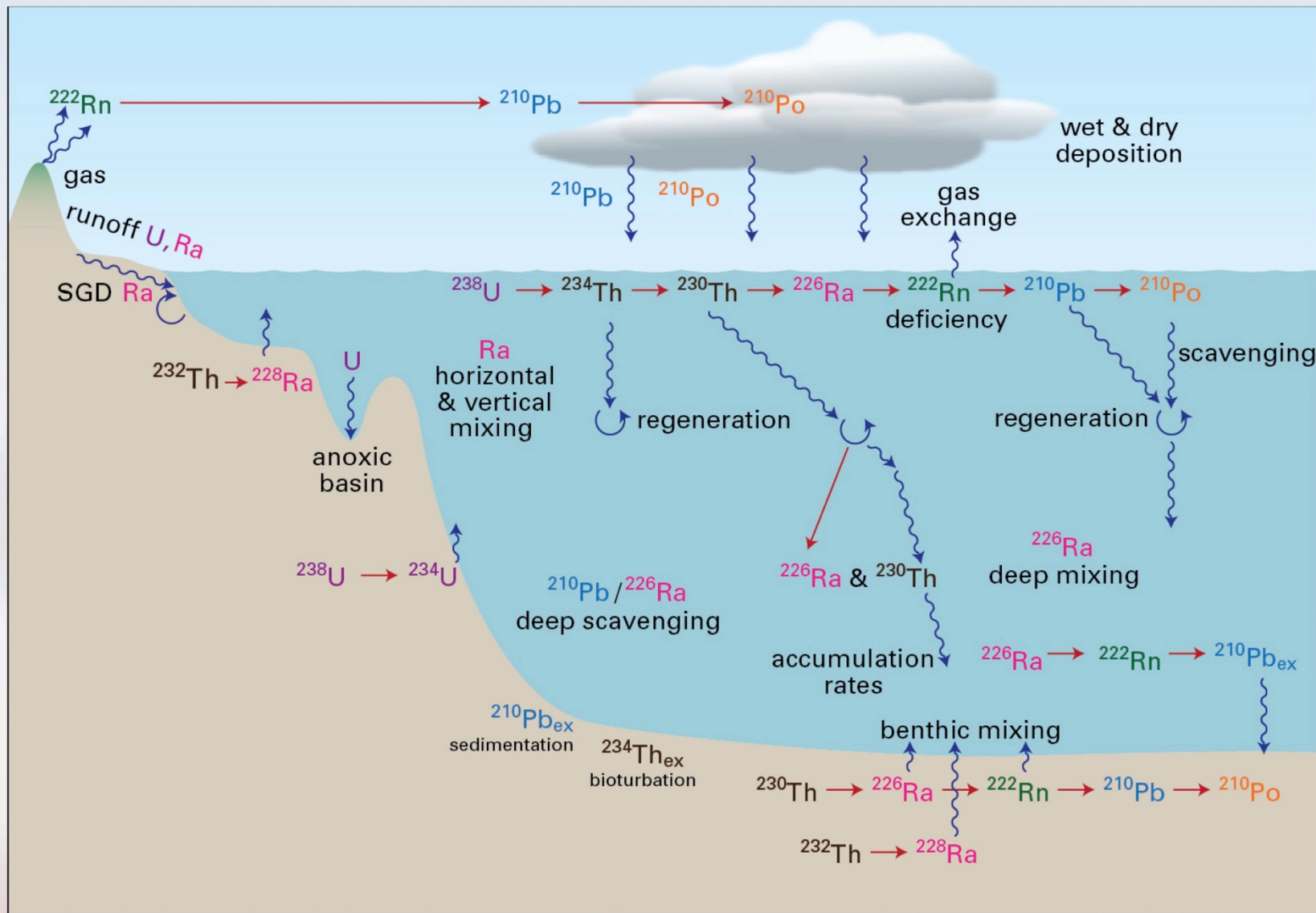
↓ Decay Series of
Short-lived
radionuclides

Rutgers van der Loeff (2014a)

Element Symbol Mass Number
U-238
 4.5×10^9 y
Half life

Particle Reactivity in Seawater	
	Low
	Medium
	High

U-Th series decay chains



Radionuclide distributions in the ocean are controlled by:

Reservoir	Process	Parameter (examples)	Tracer nuclides	Reference
Atmosphere	Aerosol Scavenging	Residence time	^7Be , ^{10}Be , ^{222}Rn , ^{210}Pb , ^{210}Po	Ralsbeck <i>et al.</i> , (1981), Bleichrodt, (1978) Martell and Moore (1974), Robbins (1978)
	Dry and wet dep.	Rates	^{131}I , ^{137}Cs , ...	Wiffen (1958), Stewart and Crooks (1958) Santschi <i>et al.</i> (1988a)
	Atmospheric circulation	Rates	^{14}C , ^{37}Ar	Loosli <i>et al.</i> (1973), Feely <i>et al.</i> (1966)
Soil	Mechanical and chemical erosion	Residence time in top soil	^{210}Pb , $^{239,240}\text{Pu}$, ^{137}Cs , ^{134}Cs	Lewis (1979), Simpson <i>et al.</i> (1976) Dominik <i>et al.</i> (1987)
	Water movement	Rates	^{10}Be , ^3H	Monaghan <i>et al.</i> (1983) Höhn and Santschi (1987), Santschi <i>et al.</i> (1987b)
Ocean and lake water	Horizontal	Diffusion	^{228}Ra , ^{222}Rn	Sarmiento and Roth (1980), Sarmiento <i>et al.</i> (1982), Moore and Santschi (1986), Broecker and Peng (1982)
	or vertical	coefficient	^3He , ^3H	Imboden and Emerson (1978), Roether <i>et al.</i> (1970), Imboden and Joller (1984), Li <i>et al.</i> (1984), Broecker and Peng (1982)
	diffusion	Rates	^{39}Ar , ^{85}Kr , ^{221}Pa , ^{230}Th	Loosli (1983), Schlitzer <i>et al.</i> (1985) Anderson <i>et al.</i> (1983), Bacon and Rosholt (1982), Broecker and Peng (1982)
	Horizontal	Rates	^{39}Ar , ^{85}Kr , ^{221}Pa , ^{230}Th	Loosli (1983), Schlitzer <i>et al.</i> (1985) Anderson <i>et al.</i> (1983), Bacon and Rosholt (1982), Broecker and Peng (1982)
	or vertical	Mechanisms	^{210}Pb , $^{239,240}\text{Pu}$	Carpenter and Beasley (1981), Beasley <i>et al.</i> (1982), Moore <i>et al.</i> (1981), Santschi <i>et al.</i> (1980b), Scott <i>et al.</i> (1983), Nyffeler <i>et al.</i> (1986, 1984)
	scavenging		^{10}Be , ^{234}Th , ^{238}Th	Mangini (1984) Santschi <i>et al.</i> (1979, 1980a, 1983a), Honeyman and Santschi (1989)
	Sediment	Rates	^{234}Th , ^{238}Th	Aller and Cochran (1976), Broecker and Peng (1982)
	resuspension	Mechanisms	$^{134,137}\text{Cs}$, ^3Be , ^{210}Pb	Robbins and Eadie (1982), Santschi (1987a), Santschi <i>et al.</i> (1987c)
	Gas exchange	Rates	^{222}Rn , ^{14}C , ^3H , ^3He	Broecker and Peng (1982), Torgerson <i>et al.</i> (1977, 1982)
	Evaporation	Rates	^3H	Herczeg and Imboden (1988)
Sediments	Bioturbation	Rates	^{234}Th , ^{210}Pb , ^{14}C	Aller and Cochran (1976), Broecker and Peng (1982), Krishnaswami <i>et al.</i> (1984)
	Sedimentation	Rates	$^{239,240}\text{Pu}$, ^{137}Cs , ^{230}Th , ^{231}Pa , ^{10}Be	Santschi <i>et al.</i> (1980b, 1983b) Armi <i>et al.</i> (1975), Broecker and Peng (1982)
			^{137}Cs , ^{134}Cs , ^{210}Pb	Wan <i>et al.</i> (1987), Robbin and Edington (1975), Robbins (1978)
	Diagenetic remobilization	Rates	^{234}U , ^{238}U , ^{230}Th , ^{234}Th	Osmond and Cowart (1982). Cochran <i>et al.</i> (1986), Cochran and Krishnaswami (1980), Colley <i>et al.</i> (1984), Colley and Thompson (1985)
	or fixation		$^{239,240}\text{Pu}$	Santschi <i>et al.</i> (1983b)
Groundwater	Sorption	Residence times	^{234}Th , ^{234}Ra , ^{226}Ra , ^{222}Rn , ^{210}Pb	Krishnaswami <i>et al.</i> (1982) Hussain and Krishnaswami (1980) Rama and Moore (1984)
	Rock-water interaction	Rates	^{14}C	Mazor <i>et al.</i> (1986)
	Mixing	Rates	^{36}Cl , ^{81}Kr	Fabryka-Martin <i>et al.</i> (1987), Lehmann <i>et al.</i> (1986)

Santschi and Honeyman (1989)

Table of radionuclides used to quantify geochemical processes, with more applications waiting to be discovered!

How do we address these oceanographic questions?

Choose the appropriate tracer(s), with 3 major constraints:

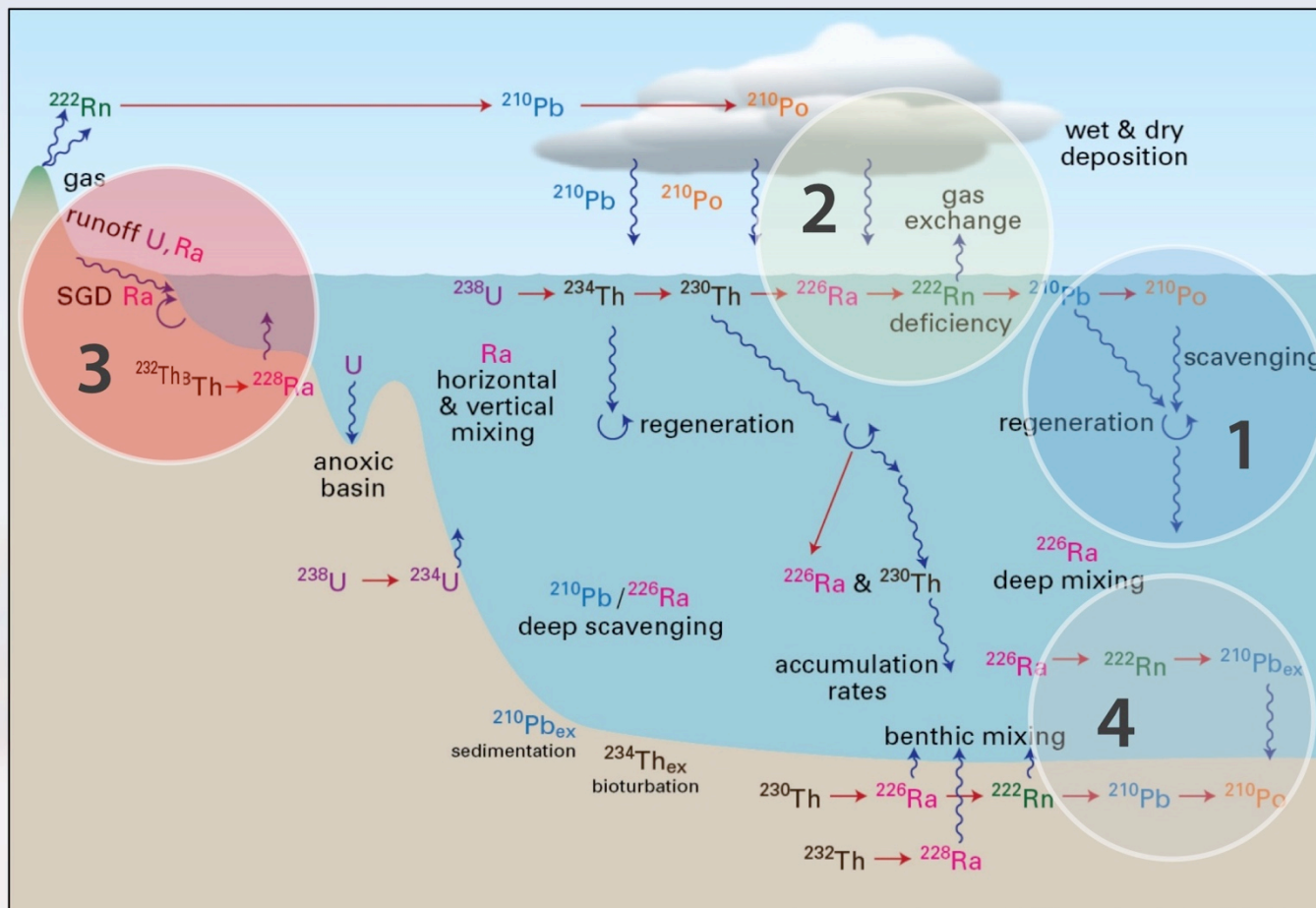
- **Source Term:** is the source function well resolved?
- **Biogeochemistry:** is it relevant? known?
- **Timescale:** is the half-life ($t_{1/2}$) of the radionuclide appropriate?

We need:

- **A Model:** For example:
- **Analytical Techniques**

$$\frac{\partial \mathbf{C}}{\partial t} = \frac{\partial}{\partial \mathbf{x}} \left(\mathbf{D} \frac{\partial \mathbf{C}}{\partial \mathbf{x}} \right) - \mathbf{v} \frac{\partial \mathbf{C}}{\partial \mathbf{x}} - \lambda \mathbf{C}$$

Case Study Applications



1. Scavenging
2. Air-Sea gas exchange
3. Tracing groundwater discharge
4. Sedimentation/Age Dating

1. Scavenging

- Many elements have much lower dissolved sea water concentrations than they should have based on continental weathering supply
- Fritz Haber (Nobel Prize in 1918)
- “Sorption” onto suspended and sinking phases leads to removal of inorganic and organic compounds (Goldberg, 1954) ****Note that here we are not differentiating between biological uptake and surface sorption.**
- Particles therefore act as scavenging sites for reactive elements (Fe, Cu, Pb, Th, Pa, etc.)
- Small particles (0.01 – 100 μm) provide increased surface area for adsorption of dissolved chemicals

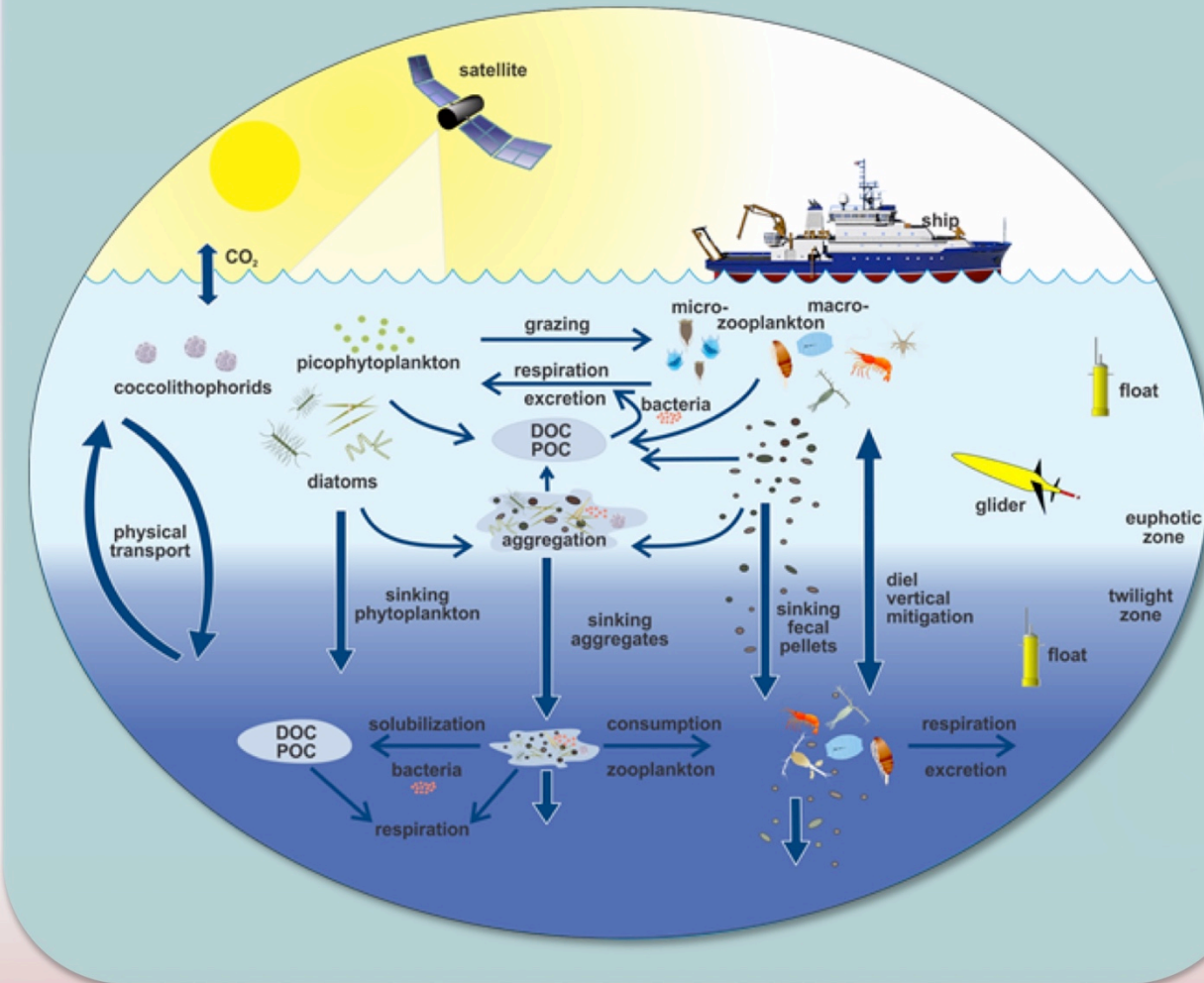


In the open ocean, particle production is primarily a result of biological processes.

Scavenging and the "Biological Pump"

Biological Pump = 5 to >12 Gt C y⁻¹

Turning off biological pump would increase atmospheric CO₂ by ~ 200 ppm.



These biological processes not only transfer organic matter to depth, but other particle reactive elements and compounds as well, such as trace metals (e.g., Hg, Zn, and Pb) and organic compounds like polychlorinated biphenyls (PCBs).

Geochemical Implications

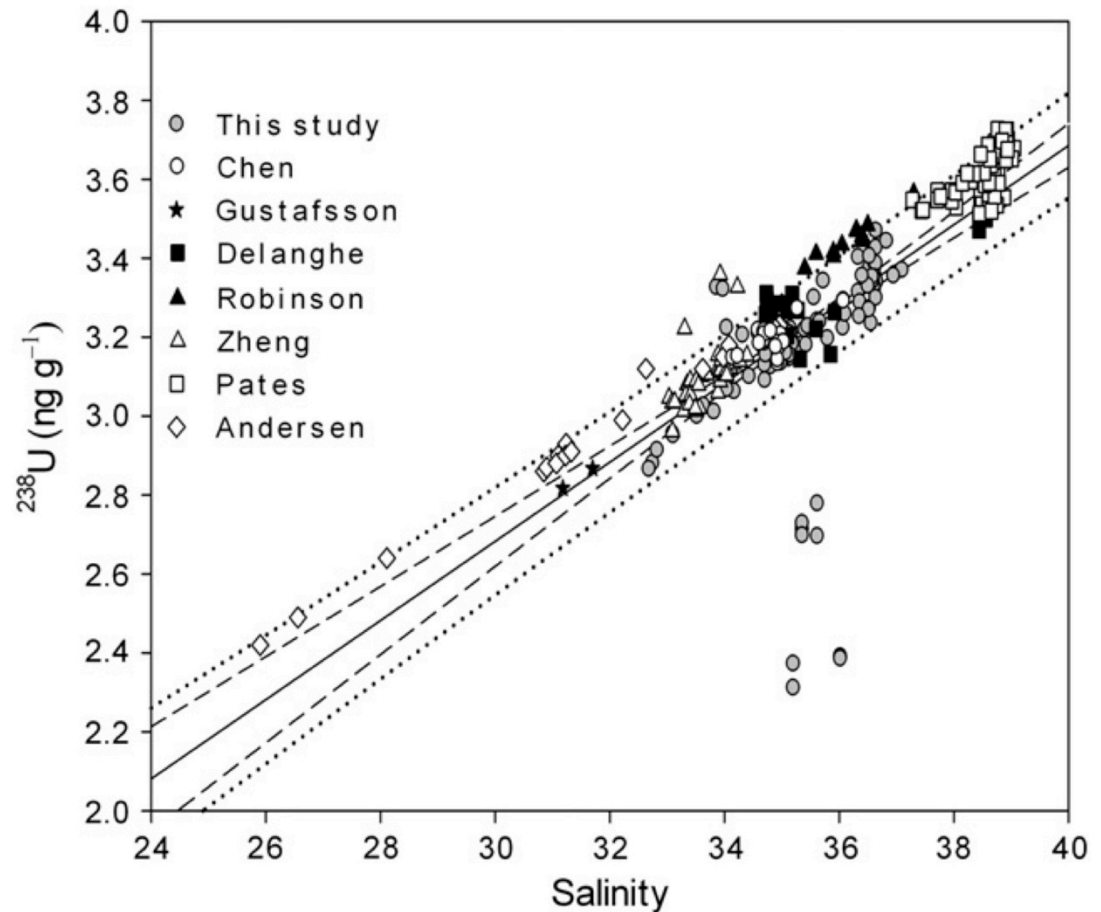


Many elements in the ocean are influenced by scavenging. Concentrations increasing with depth are diagnostic of scavenging and regeneration at depth (red boxes are not exhaustive).

U-Th series disequilibria

Uranium is soluble
and therefore
conservative in
seawater.

Its distribution can
be described by
measuring salinity.



$$\text{ng g}^{-1}: {}^{238}\text{U} (\pm 0.061) = 0.100 \times S - 0.326$$

$$\text{dpm L}^{-1}: {}^{238}\text{U} (\pm 0.047) = 0.0786 \times S - 0.315$$

Owens *et al.*, 2011

How does it work?

^{234}Th as a tracer for particle export



 = marine particle

 ^{238}U is conservative in seawater

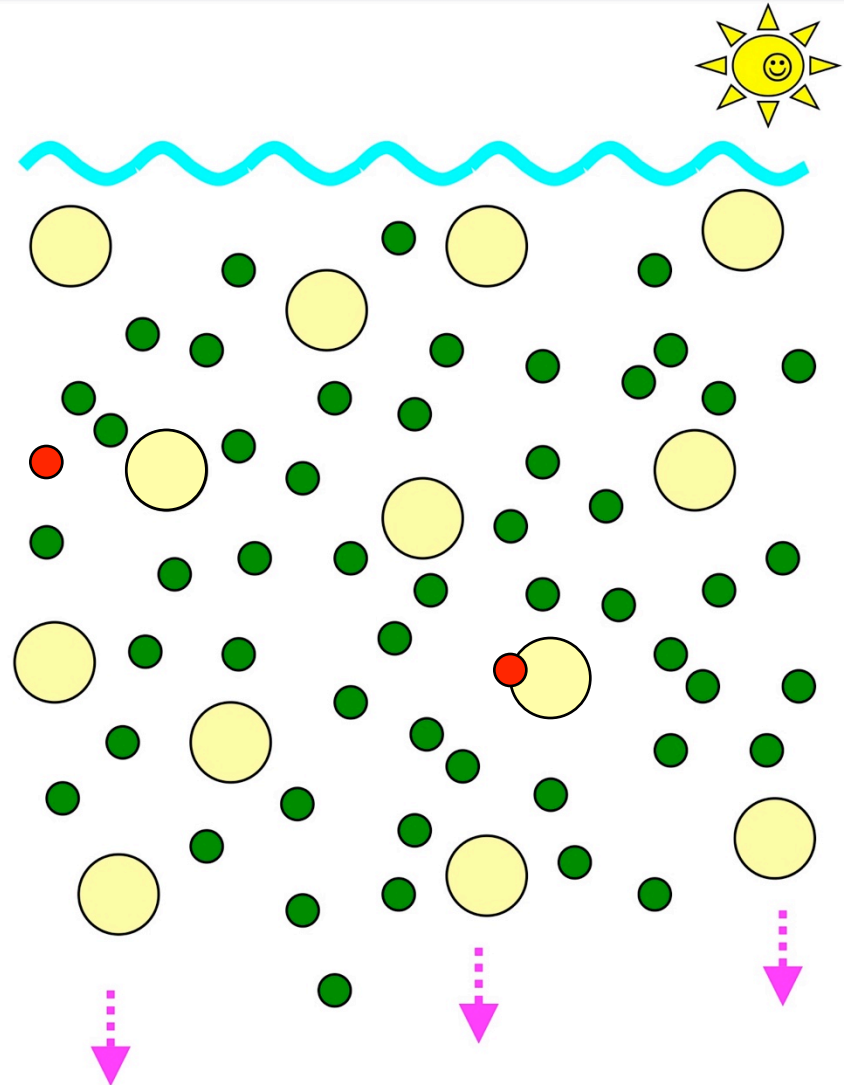
 ^{234}Th is highly particle-reactive

Secular equilibrium is expected
in an ocean without particles
(Activities should be the same...)

 Particulate matter

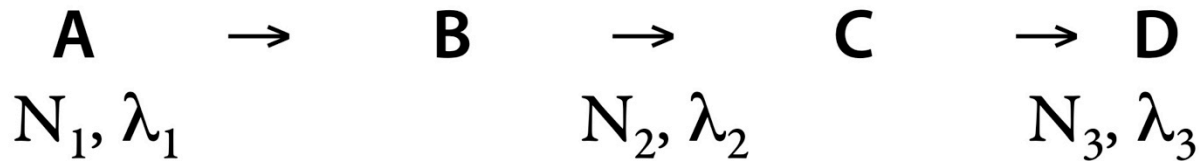
 ^{238}U

 ^{234}Th

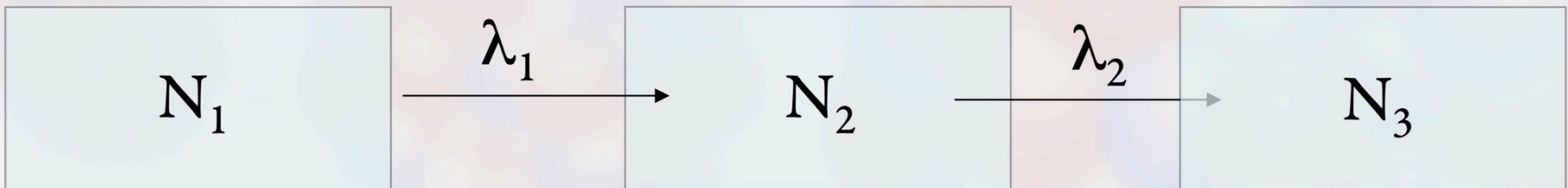


^{238}U Decays to ^{234}Th : The Case of the Radioactive Daughter

The Bateman Equations



$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$



$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 [e^{-\lambda_1 t} - e^{-\lambda_2 t}] + N_2^0 e^{-\lambda_2 t}$$

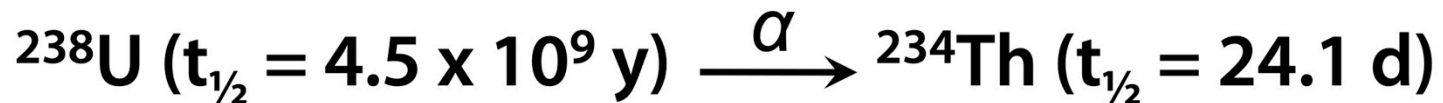
Bateman, 1910

Secular Equilibrium

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \rightarrow N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 [e^{-\lambda_1 t} - e^{-\lambda_2 t}] + N_2^0 e^{-\lambda_2 t}$$

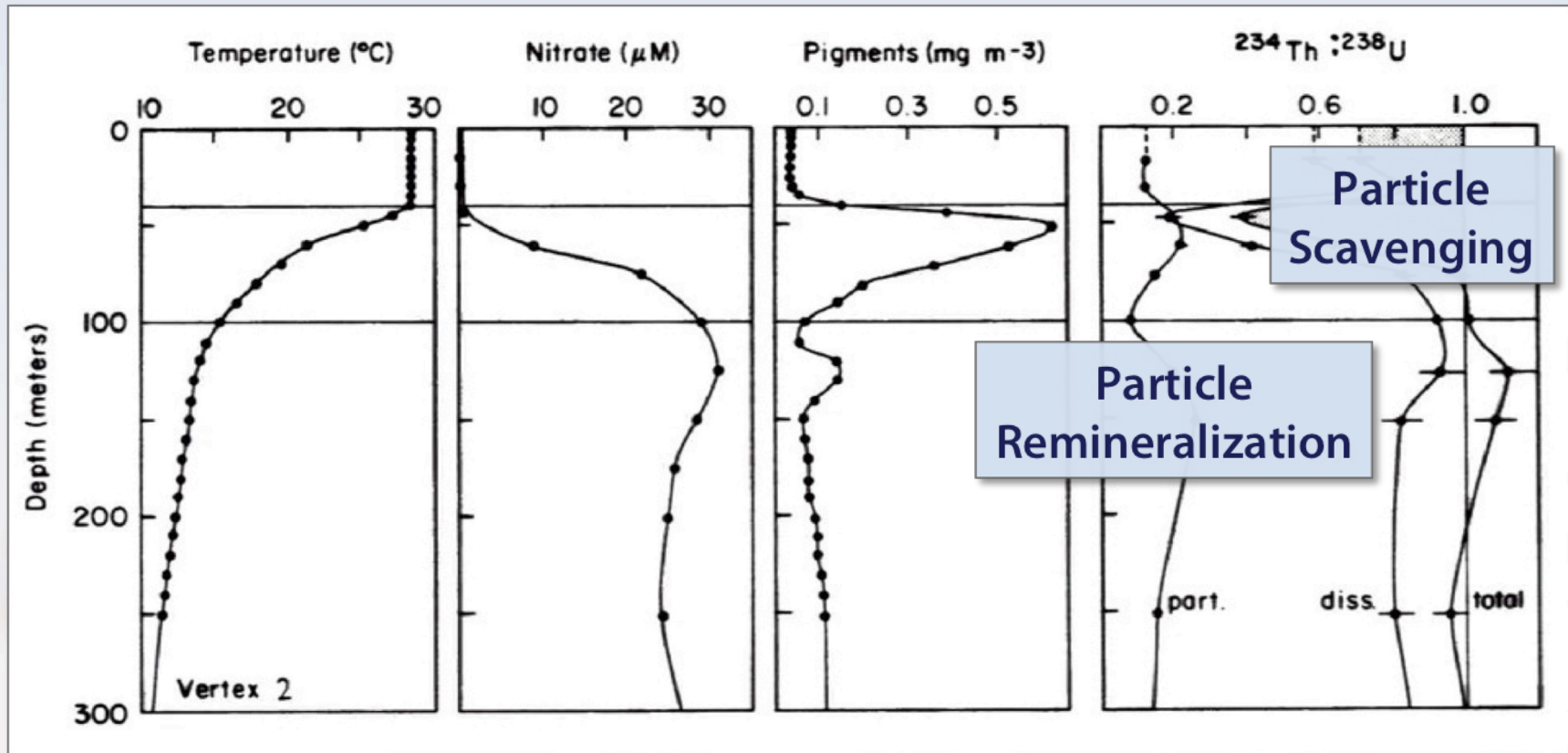
When $\lambda_1 \ll \lambda_2 \rightarrow \lambda_1 N_1 = \lambda_2 N_2$

For the naturally occurring radionuclides ^{238}U , ^{235}U , and ^{232}Th , the half-lives of the parent nuclides are much longer than their daughter products:



- The number of parent (N_1) atoms therefore remains ~ constant
- ^{238}U and ^{234}Th in equilibrium = same activity, A , where $A = N\lambda$

Radionuclide distributions in the ocean are controlled by:



Coale and Bruland (1987)

See decrease in ^{234}Th activity correlated with increasing biomass as evidenced by pigment concentrations.

Large scale differences are well-captured

Hawaii $^{234}\text{Th}/^{238}\text{U} \sim 1$
Low Particle Flux to Depth

NW Pacific $^{234}\text{Th}/^{238}\text{U} < 1$
High Particle Flux to Depth

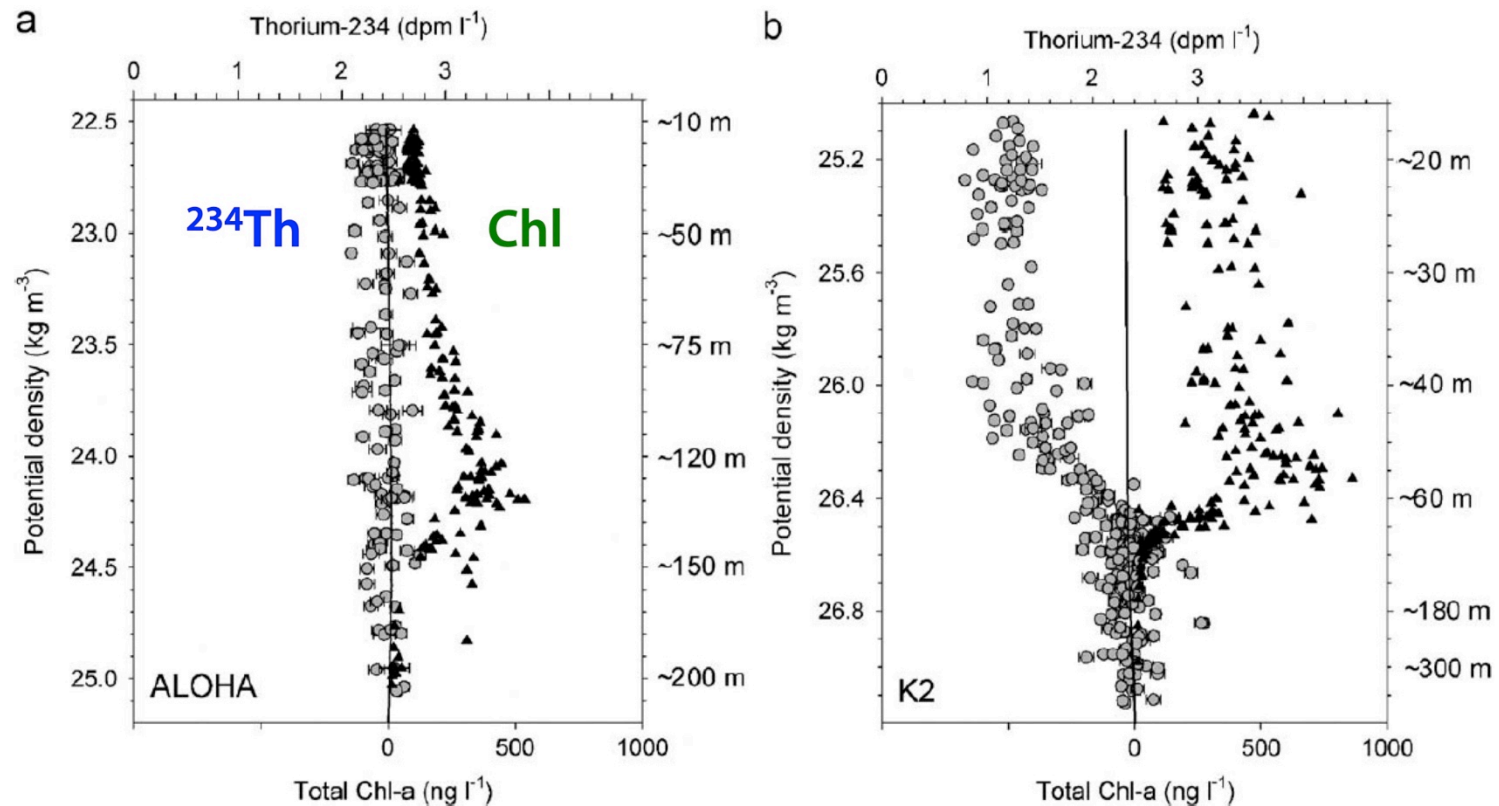
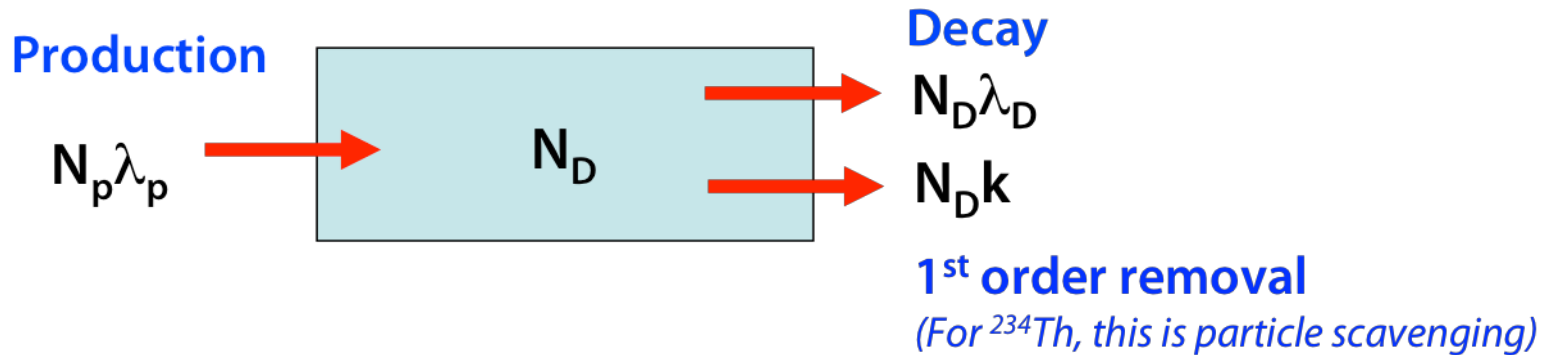


Fig. 1. Total ^{234}Th activities (dpm l^{-1}) and total chlorophyll- a (ng l^{-1}) vs. potential density (kg m^{-3}) for (a) ALOHA and (b) K2. Thorium-234 data are plotted as gray circles with error bars shown if greater than the symbol size, using the upper X-axis and plotted relative to ^{238}U (black line) near 2.3 dpm l^{-1} . Total chl- a data are offset to run from 0 to 1000 ng l^{-1} starting at the point along the lower X-axis, where $^{234}\text{Th} = ^{238}\text{U}$, to emphasize ^{234}Th removal associated with the euphotic zone. Density scales (Y-axis—left) differ at the two sites, and for convenience, approximate water depths are shown along the right Y-axis specific to each site.

Buesseler et al. (2009)

The Basic Model

Particle reactive ^{234}Th is sourced from conservative ^{238}U in seawater, and ^{234}Th is lost via radioactive decay AND particle attachment (scavenging) and sinking.



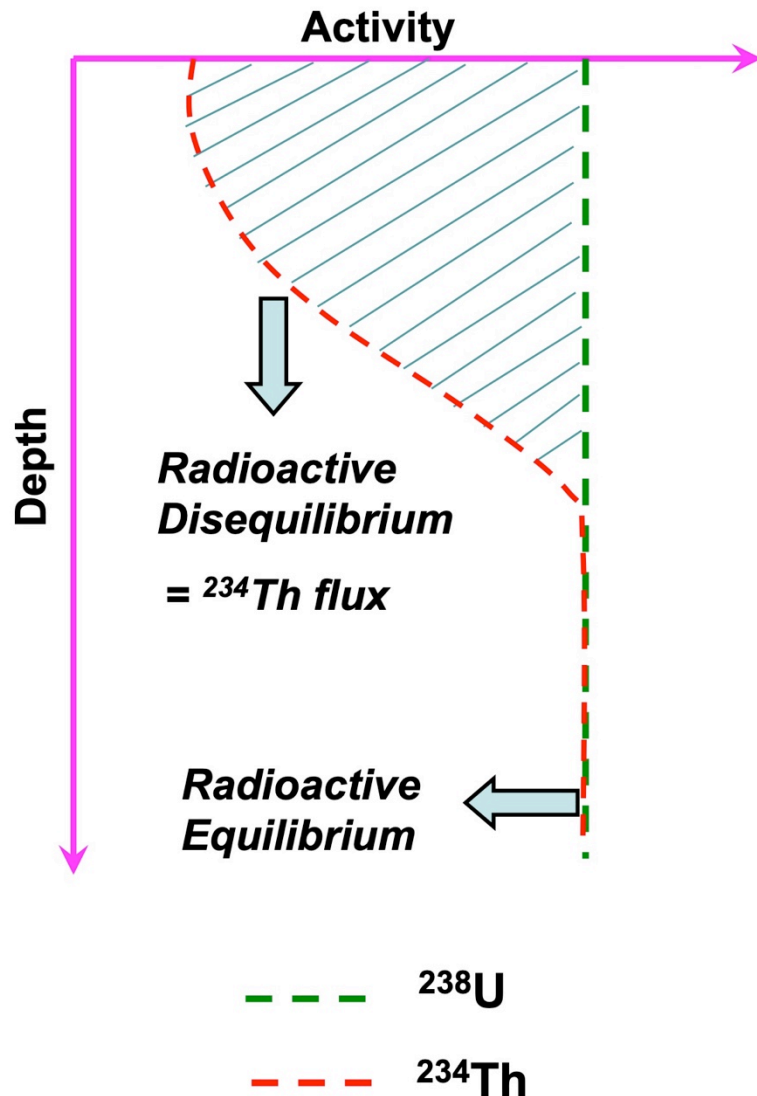
$$N_p \lambda_p = N_D \lambda_D + k N_D \quad \text{Remember that } A = \lambda N$$

$$A_p \lambda_D = A_D \lambda_D + k A_D \quad \text{where } k A_D \text{ is the } A_D \text{ "Flux"}$$

$$k = \lambda_D (A_p / A_D - 1) \quad \text{where } k \text{ is the scavenging coefficient}$$

$$\tau_D = 1/k = \text{Residence time of } N_D \text{ with respect to scavenging}$$

Particle Export using $^{238}\text{U} : ^{234}\text{Th}$



Assumes Steady State and minimal physical processes:

$$^{234}\text{Th flux} = \lambda_{\text{Th}} \int (A_{\text{U}} - A_{\text{Th}}) dz =$$

$$(\text{dpm m}^{-2} \text{ d}^{-1})$$

Where:

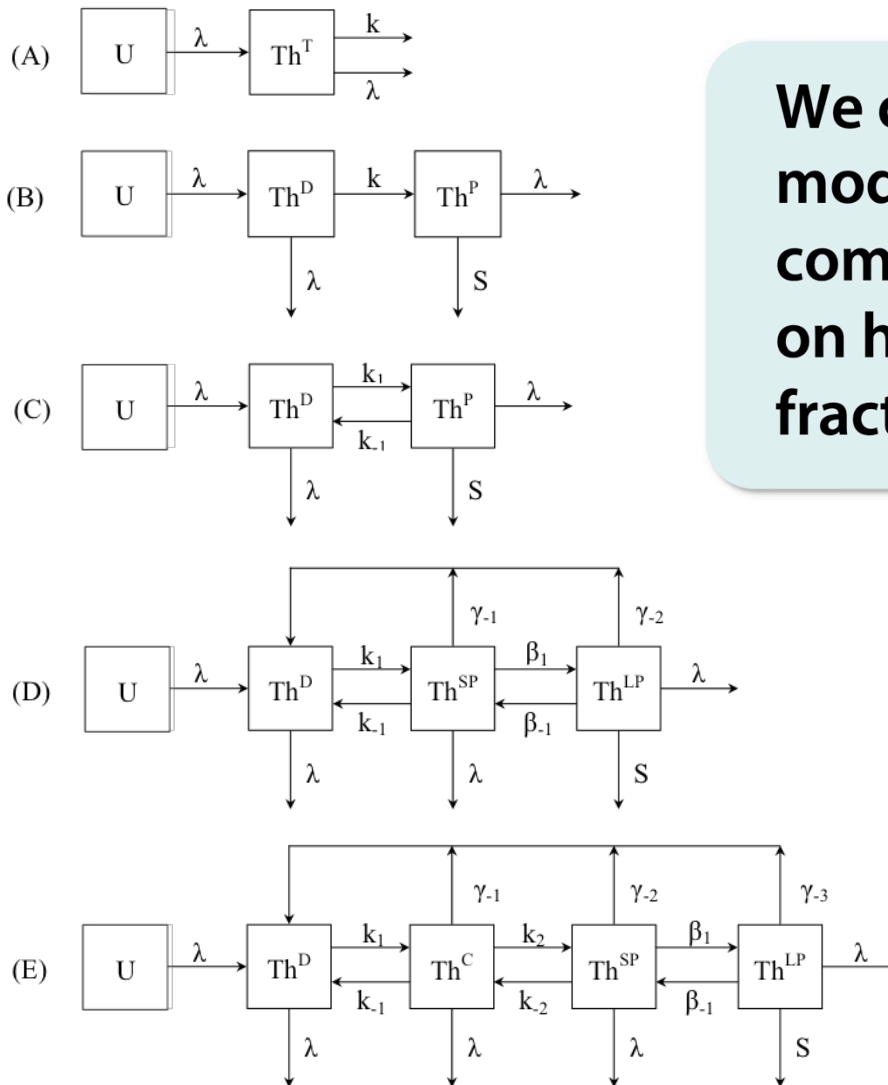
λ = Radioactive Decay Constant for ^{234}Th (d^{-1})

A_{U} = Activity of ^{238}U (dpm L^{-1})

A_{Th} = Activity of ^{234}Th (dpm L^{-1})

z = Depth interval in water column.

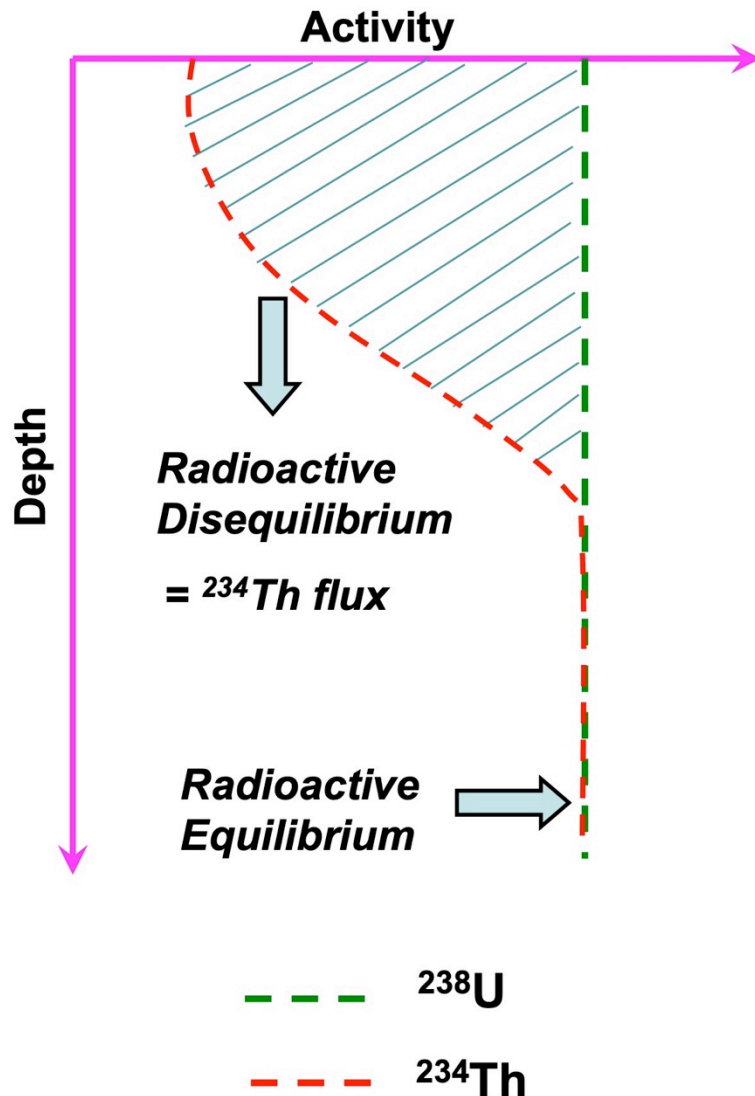
Particle Export using $^{238}\text{U} : ^{234}\text{Th}$



Cochran and Masqué (2003)

We can make these box models even more complicated depending on how many particle size fractions are measured.

Particle Export using $^{238}\text{U} : ^{234}\text{Th}$



Buesseler et al. (2006)

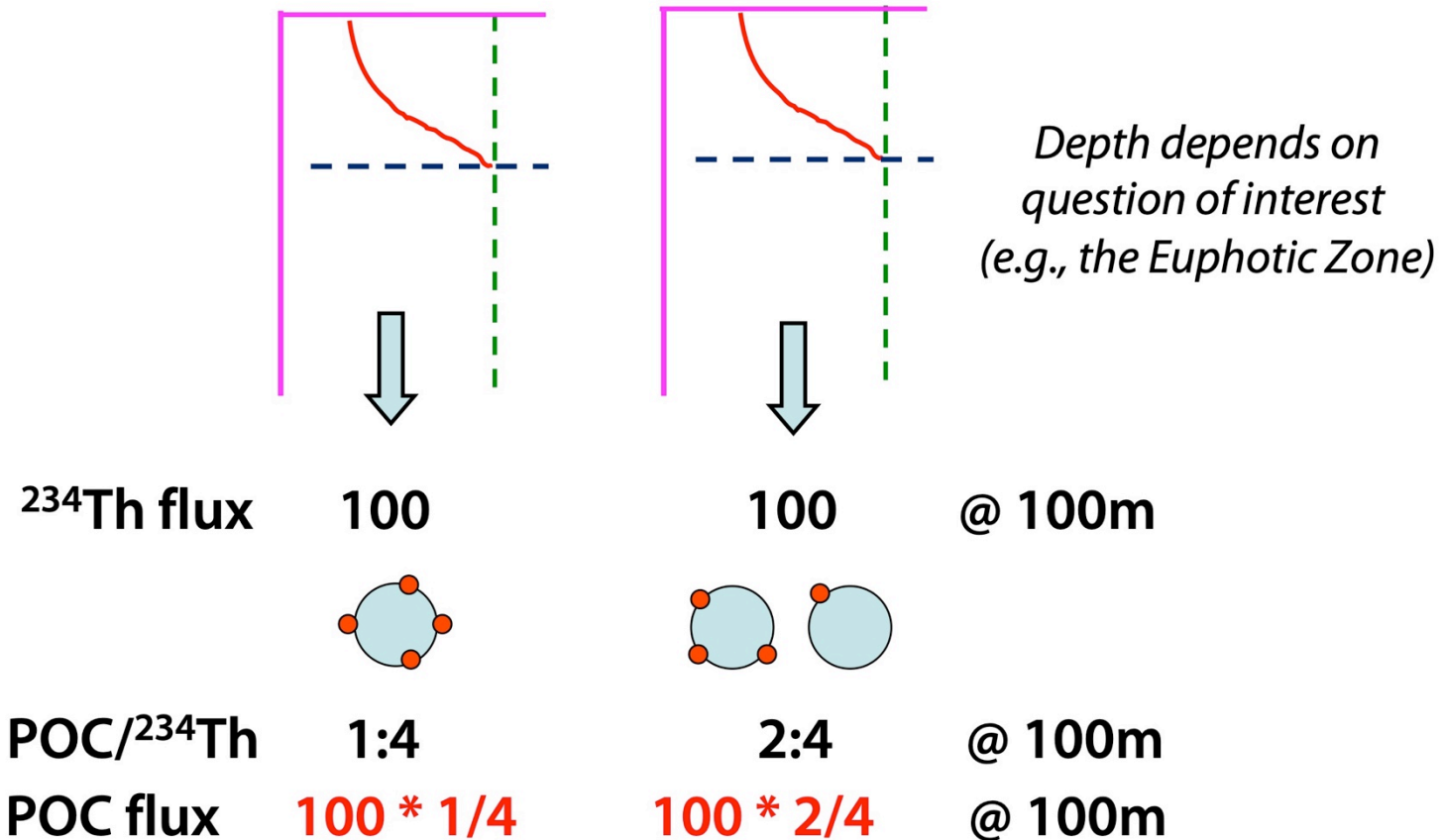
$$^{234}\text{Th flux} = \lambda_{\text{Th}} \int (A_{\text{U}} - A_{\text{Th}}) dz$$

Carbon (other element
or compound) Flux =

$$^{234}\text{Th flux} \times [C/^{234}\text{Th}]_{\text{part}}$$

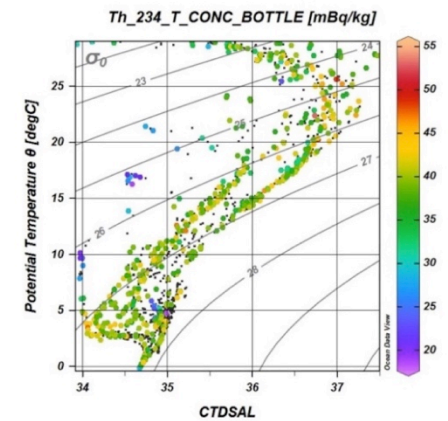
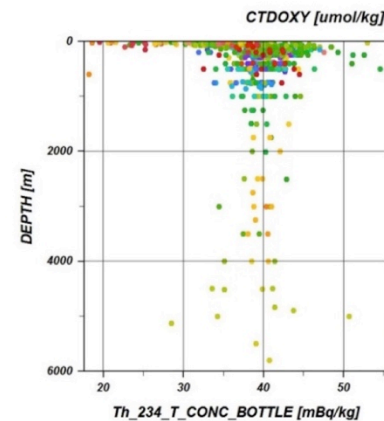
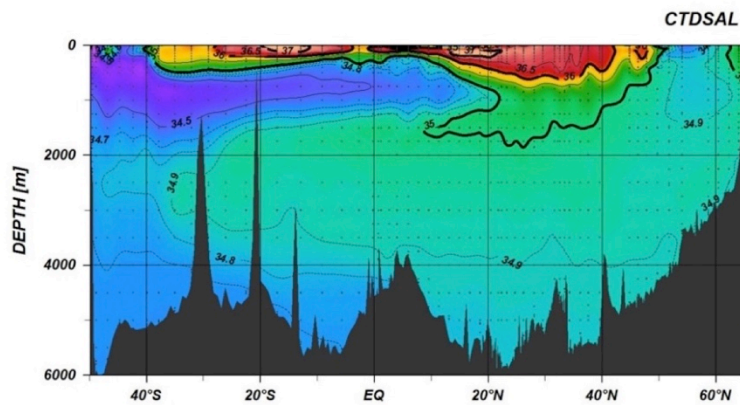
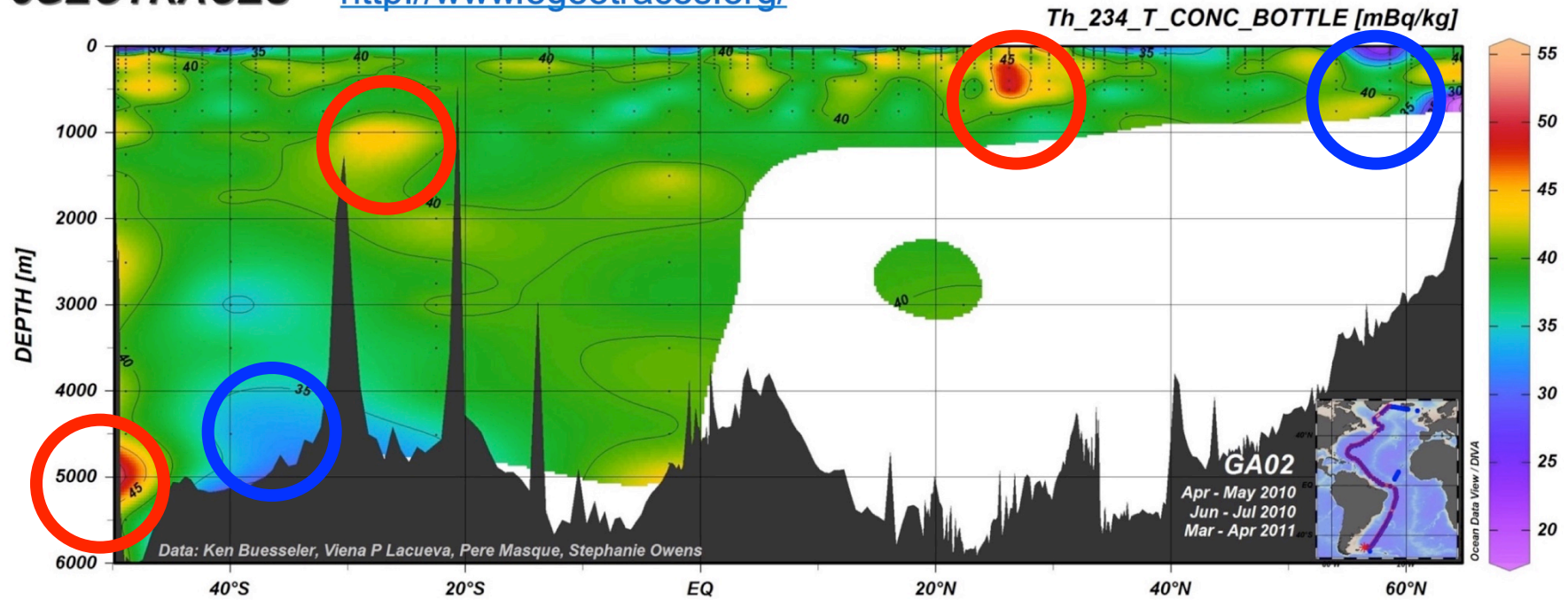
Estimate of particulate organic carbon (POC) or other element/compound flux from ^{234}Th flux

$$\text{Flux} = ^{234}\text{Th flux} \cdot [\text{POC}/^{234}\text{Th}]_{\text{sinking particles}}$$



Buesseler et al. (2006)

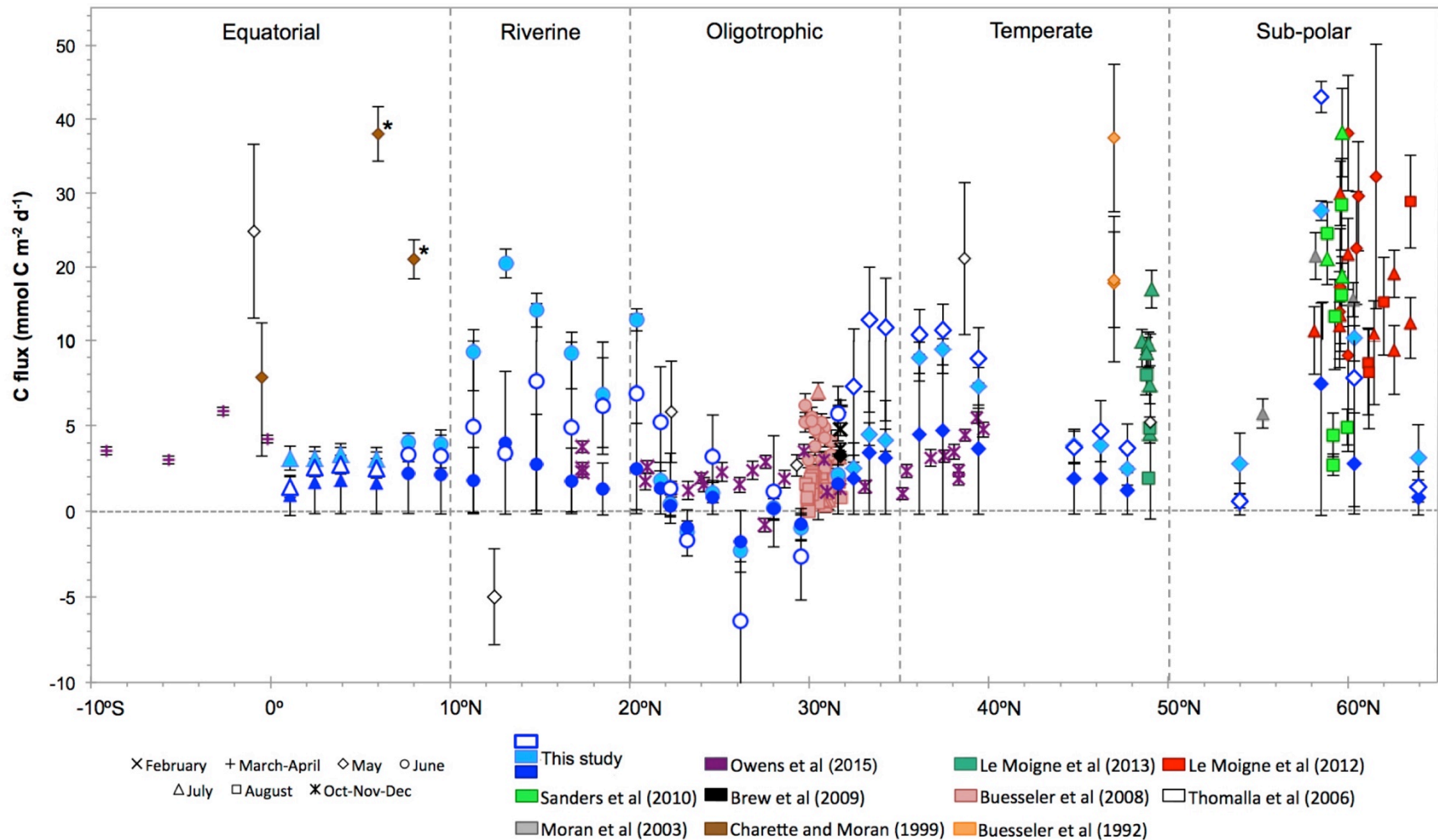
eGEOTRACES

<http://www.egeotrac.es.org/>

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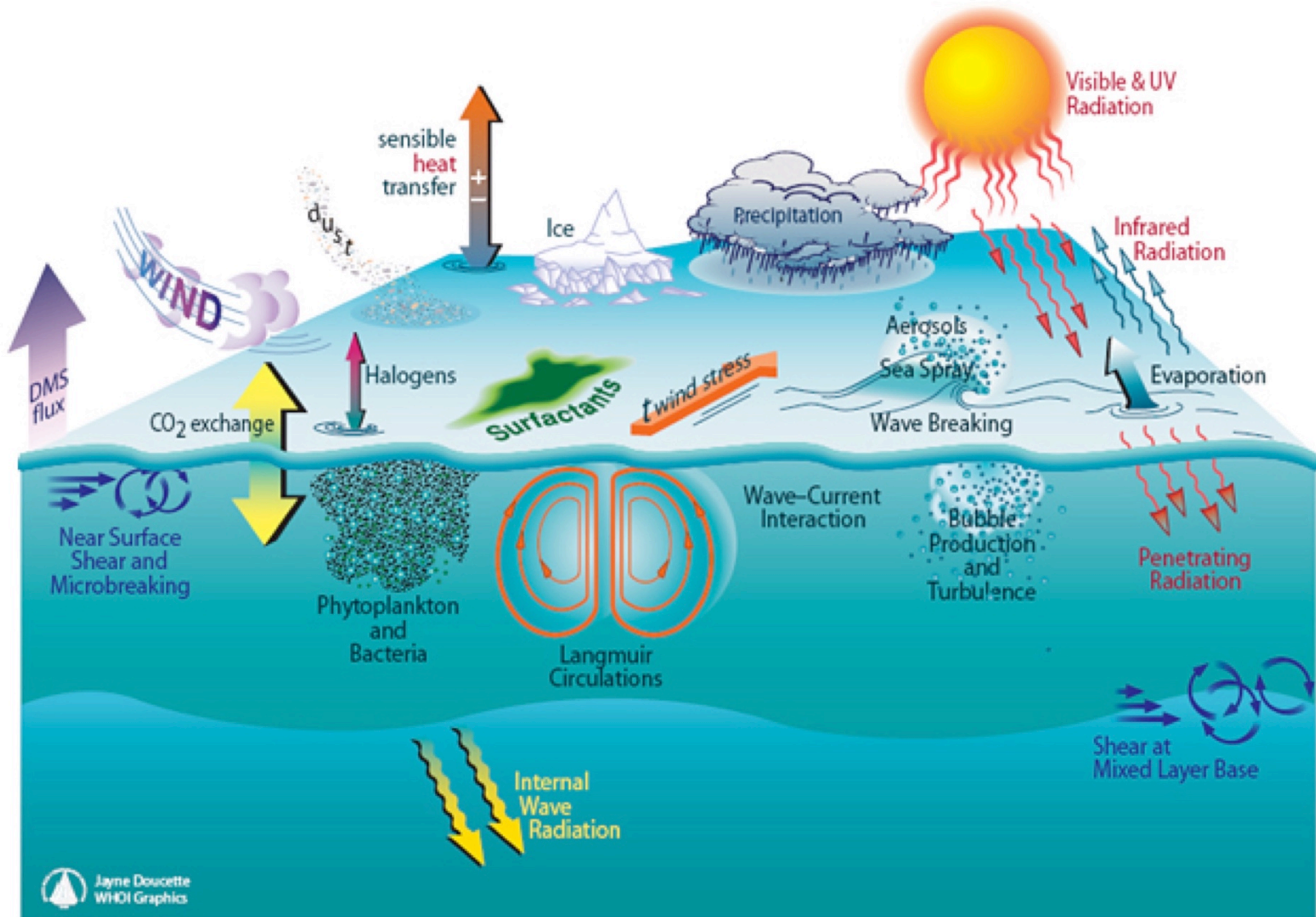
Compilation of Carbon Export fluxes at 100m in the Atlantic Ocean

From the Equator to the Arctic...

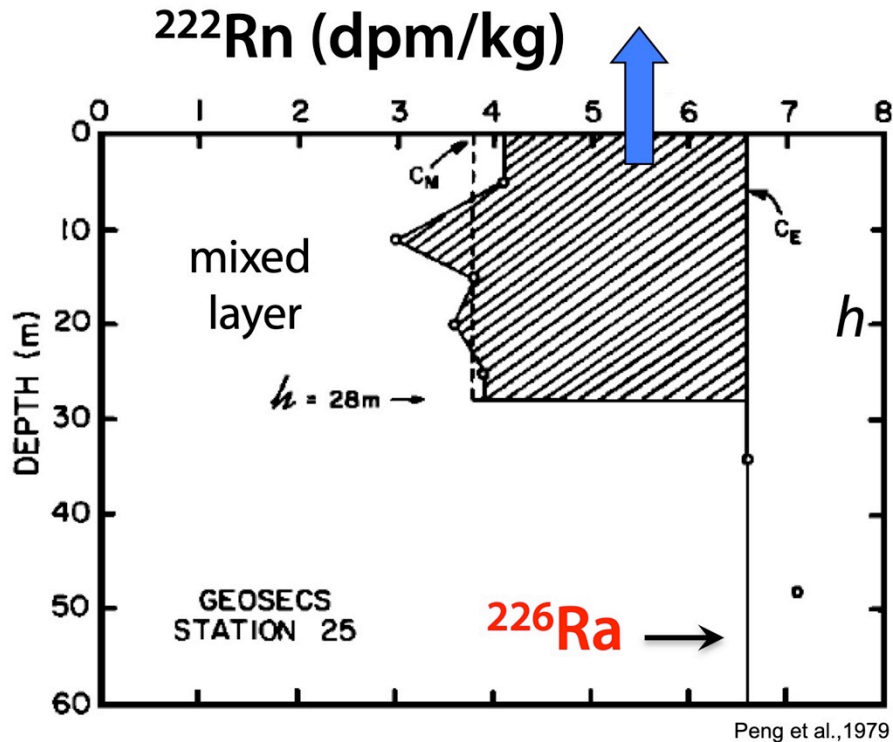


Puigcorb  et al. (2017)

2. Air-Sea Gas Exchange



Air-Sea Gas Exchange Using ^{226}Ra : ^{222}Rn



$$^{226}\text{t}_{1/2} = 1600 \text{ y} \quad ^{222}\text{t}_{1/2} = 3.8 \text{ days}$$

$$k^*(^{222}\text{Rn}_{\text{water}} - ^{222}\text{Rn}_{\text{air}}) = 0$$

$$^{222}\lambda \int (^{226}\text{Ra}_{\text{water}} - ^{222}\text{Rn}_{\text{water}}) dh$$

Where:

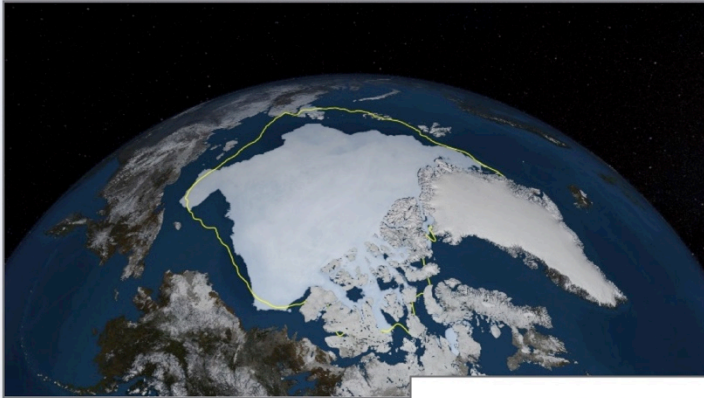
k = Gas transfer velocity (m/d)

^{226}Ra and ^{222}Rn = Activity measured in seawater or air

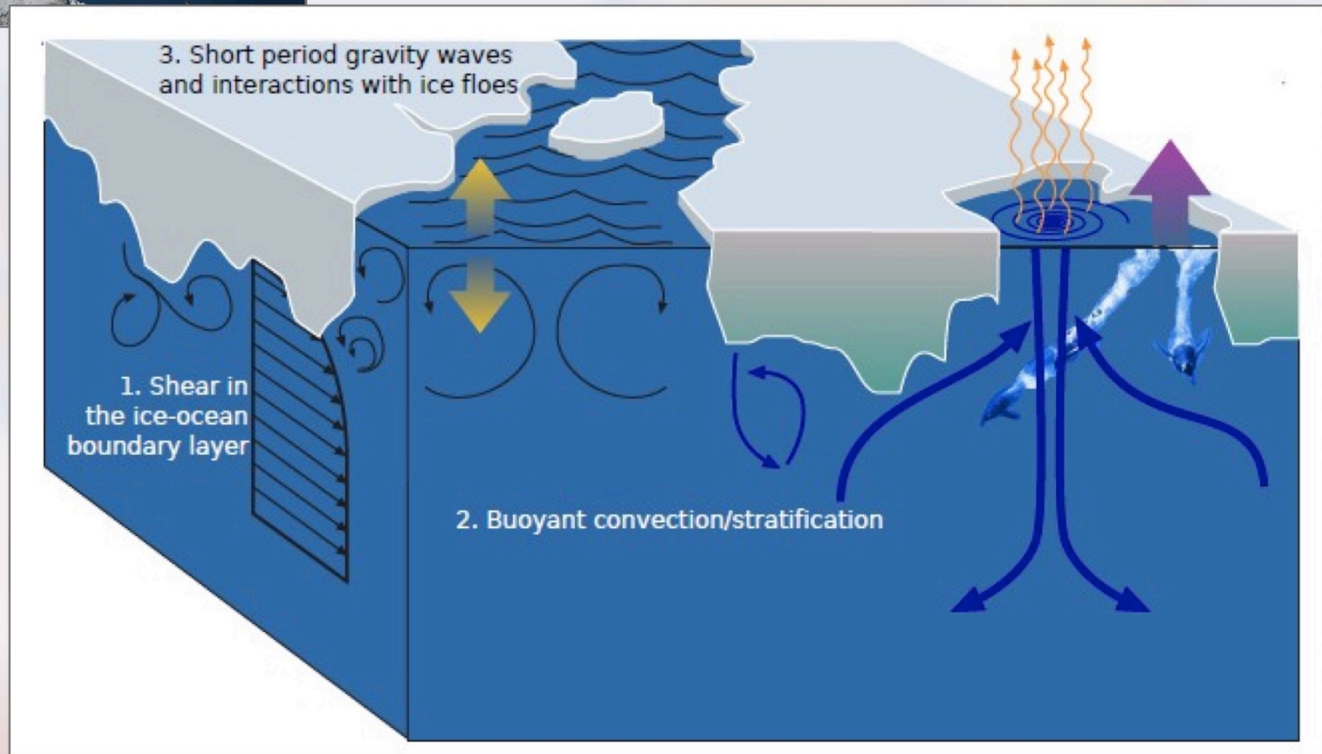
h = Depth of disequilibrium

$$k = \left(\frac{^{226}\text{Ra}_{\text{water}}}{^{222}\text{Rn}_{\text{water}}} - 1 \right)^{222} \lambda h$$

Application in the Arctic Ocean – Sea Ice



The relationship between ice cover and gas exchange in partially ice-covered regions can have a large effect on calculated annual CO₂ fluxes to and from the atmosphere.

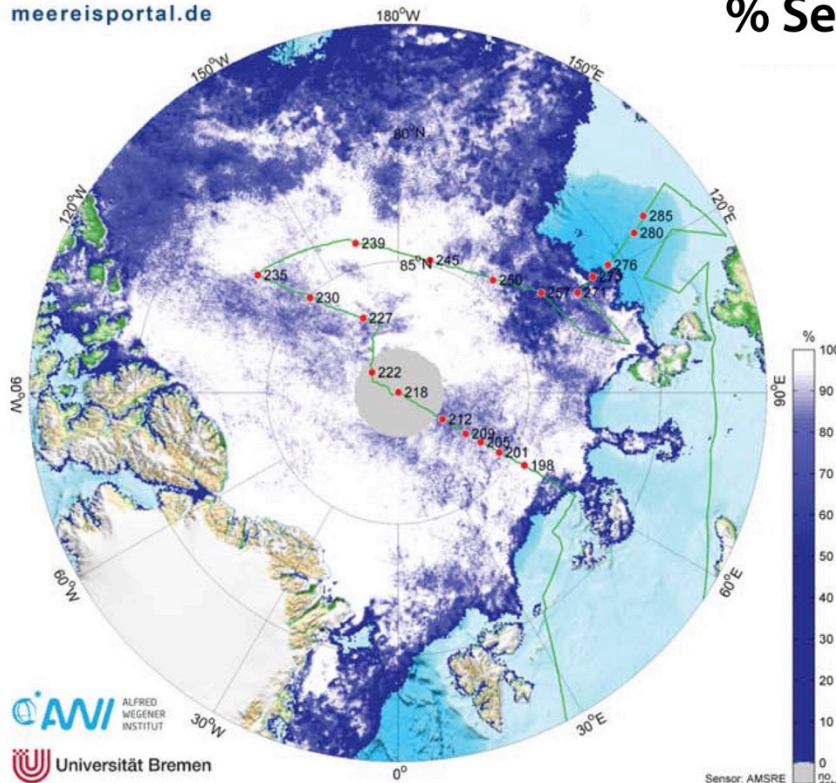


Loose et al., 2014

ARK XXIV/3 TransArc 2011

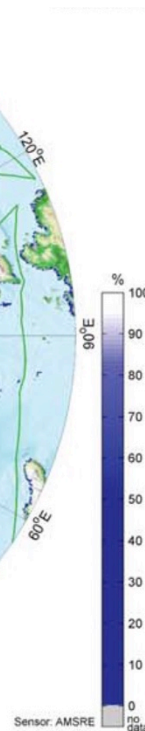
This map depicts station locations of ^{222}Rn and ^{226}Ra samples collected during varying ice conditions in the Arctic Ocean.

meereisportal.de



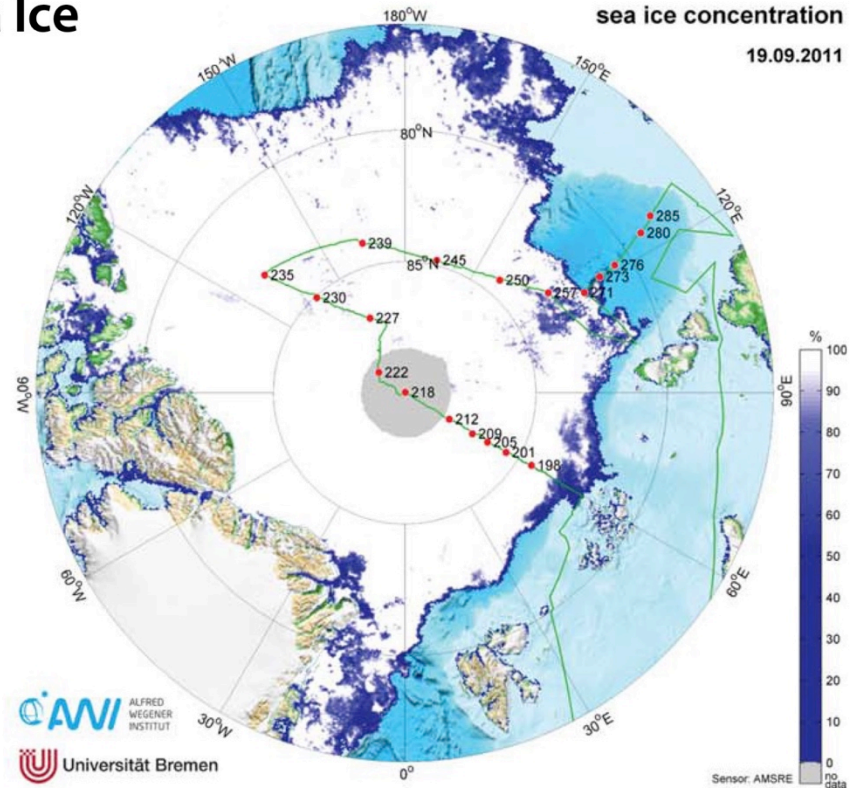
August 13, 2011

% Sea Ice



sea ice concentration

19.09.2011



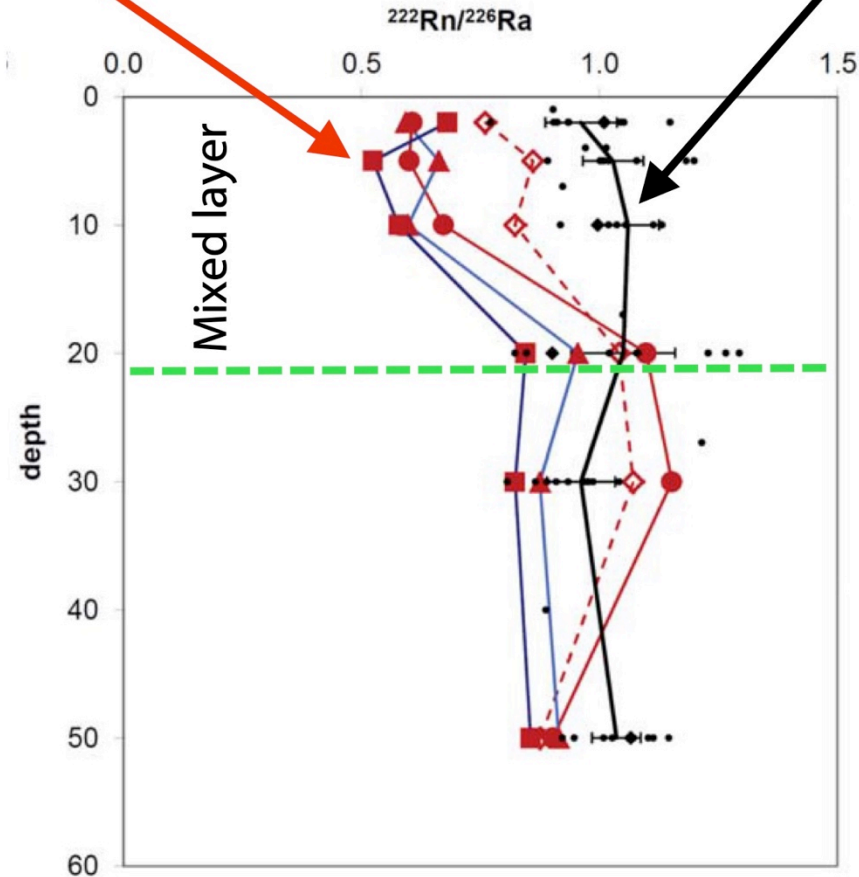
September 19, 2011

Rutgers van der Loeff et al. (2014b)

ARK XXIV/3 TransArc 2011

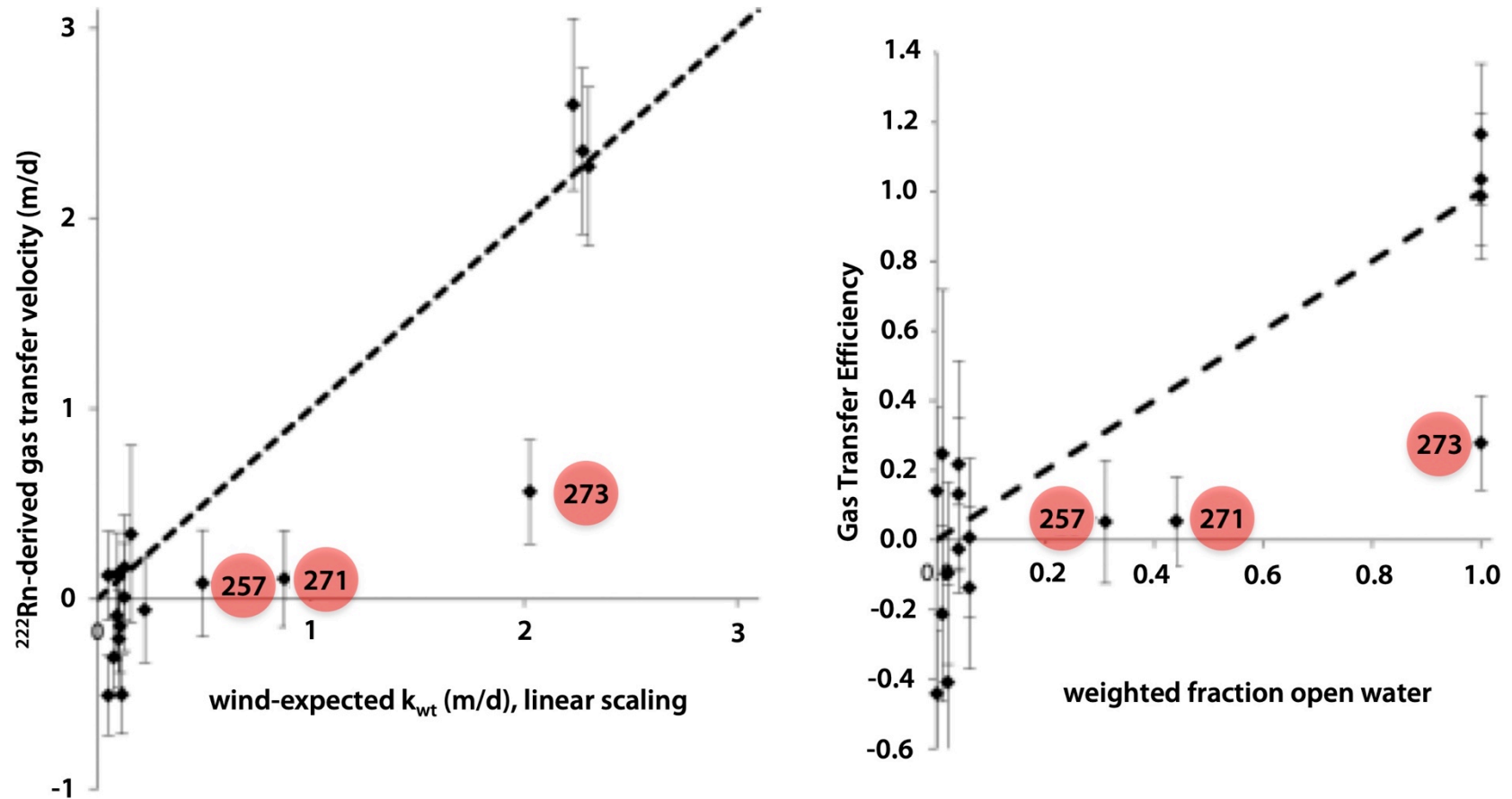
Open Water

Completely Ice Covered



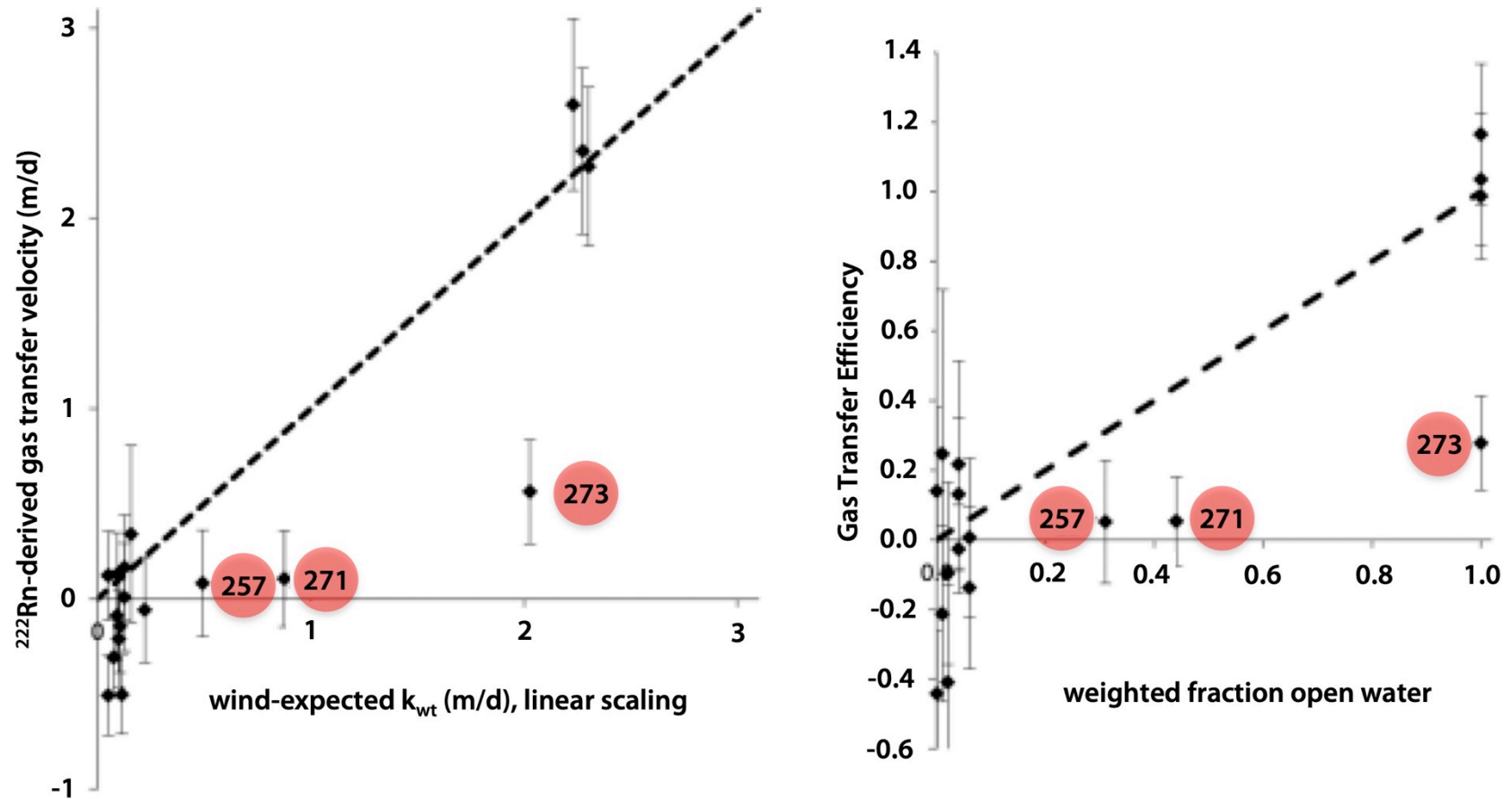
Rutgers van der Loeff et al. (2014b)

Ice covered regions had average gas transfer velocities $< 0.1 \text{ m d}^{-1}$.
 Open water gas transfer velocities were as expected
 and ranged from $0.6 - 2.6 \text{ m d}^{-1}$.



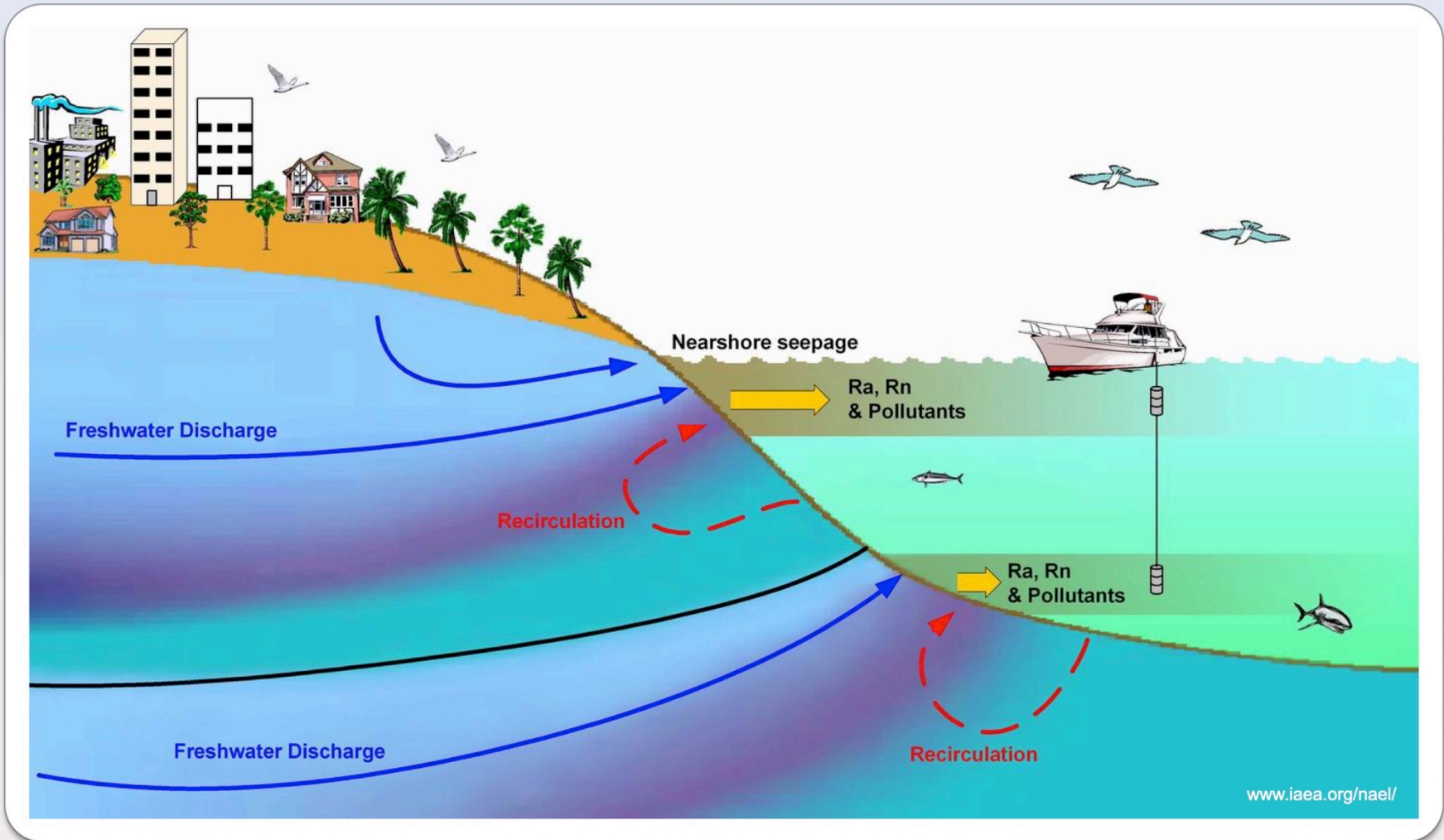
Rutgers van der Loeff et al. (2014b)

Partially ice-covered stations (red circles) exposed to wind had gas exchange rates significantly lower than expected.



Rutgers van der Loeff et al. (2014b)

3. Tracing Groundwater Discharge

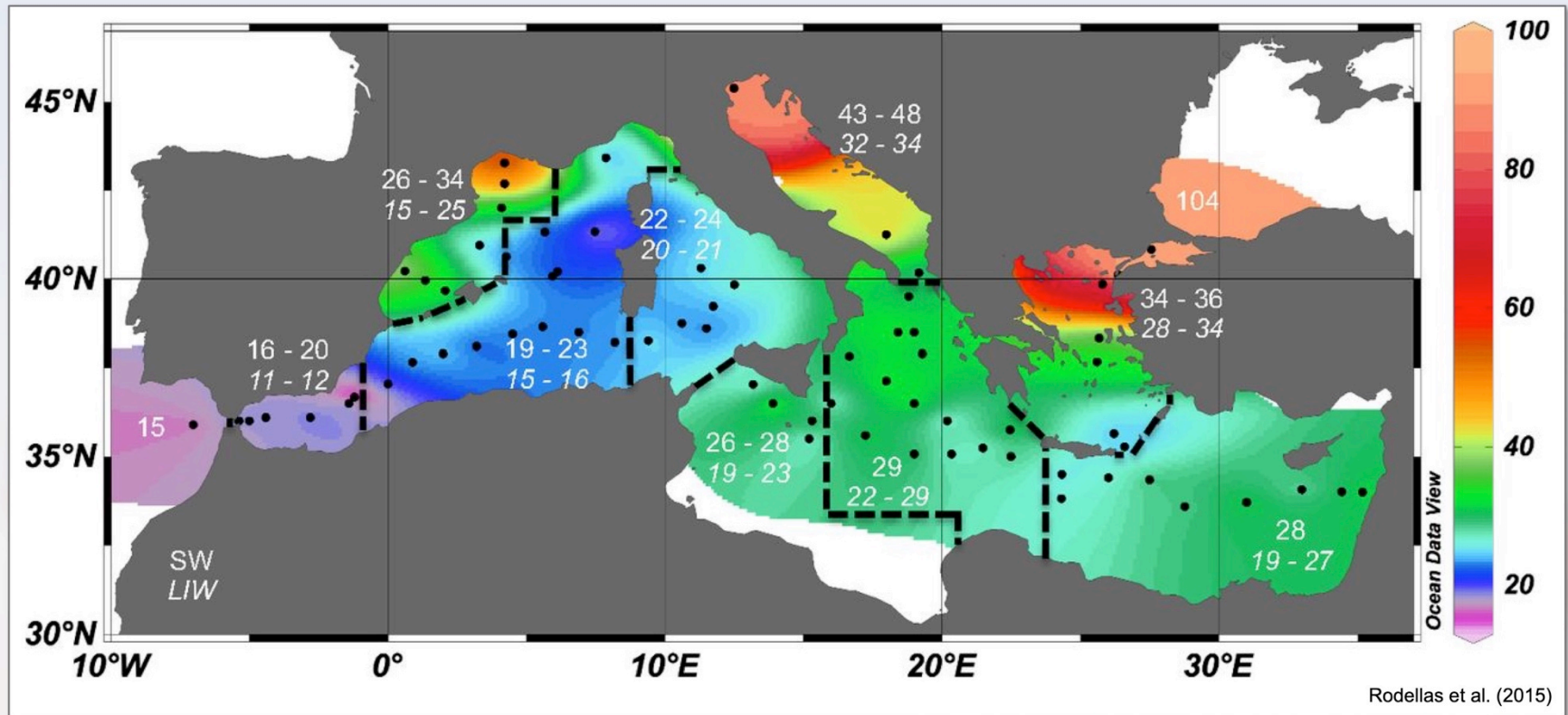


The Radium Quartet

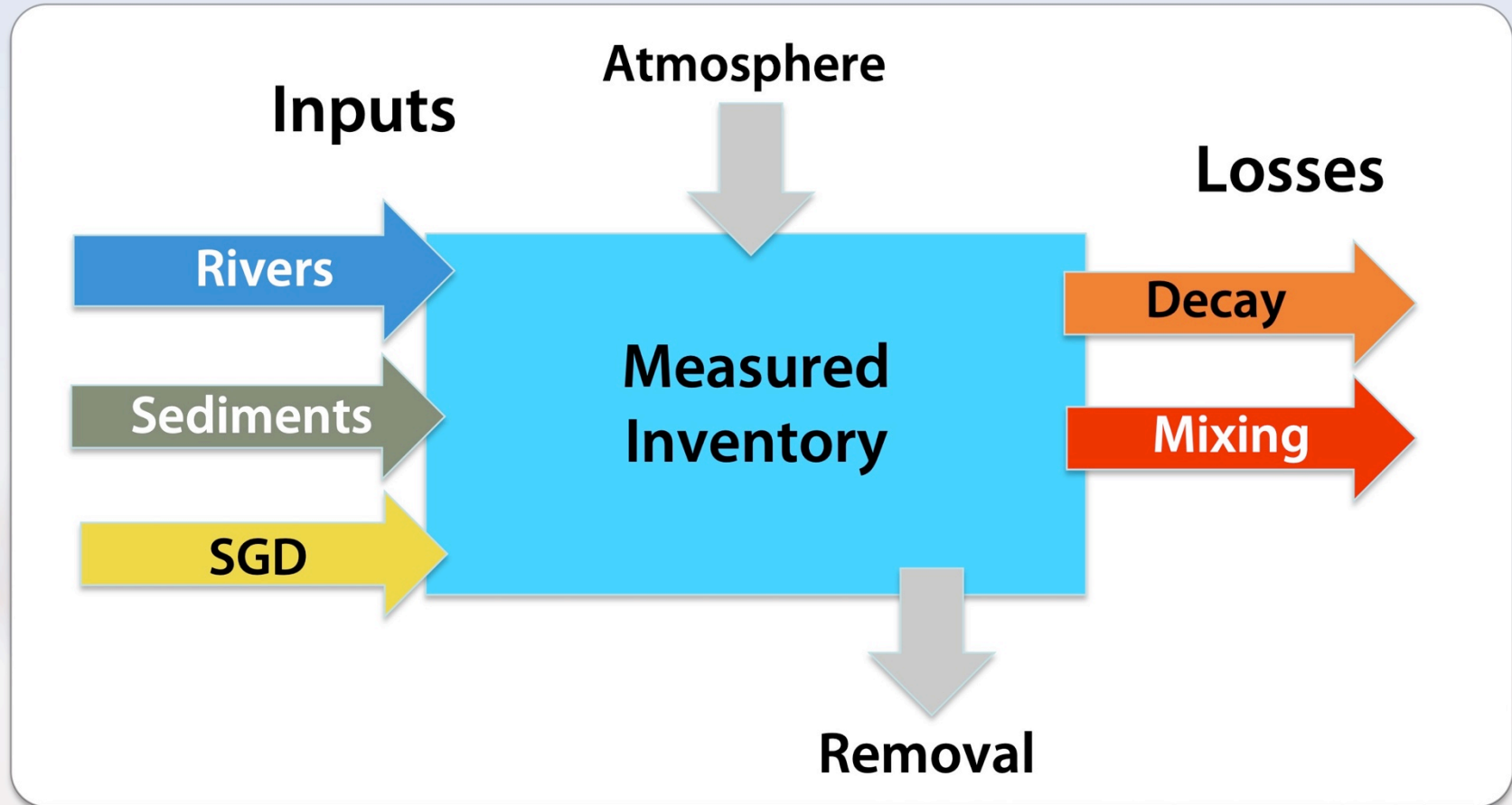


- Radioactive radium is derived from the decay of thorium isotopes.
- Ra adsorbs to particles in fresh water, but is mobile in salt water.
- Ra is not reactive in coastal waters.
- Ra concentrations are usually high in salty submarine groundwater (SGD) and low in ocean water.

Distribution of ^{228}Ra concentrations (dpm m $^{-3}$) in surface waters of the Mediterranean Sea.

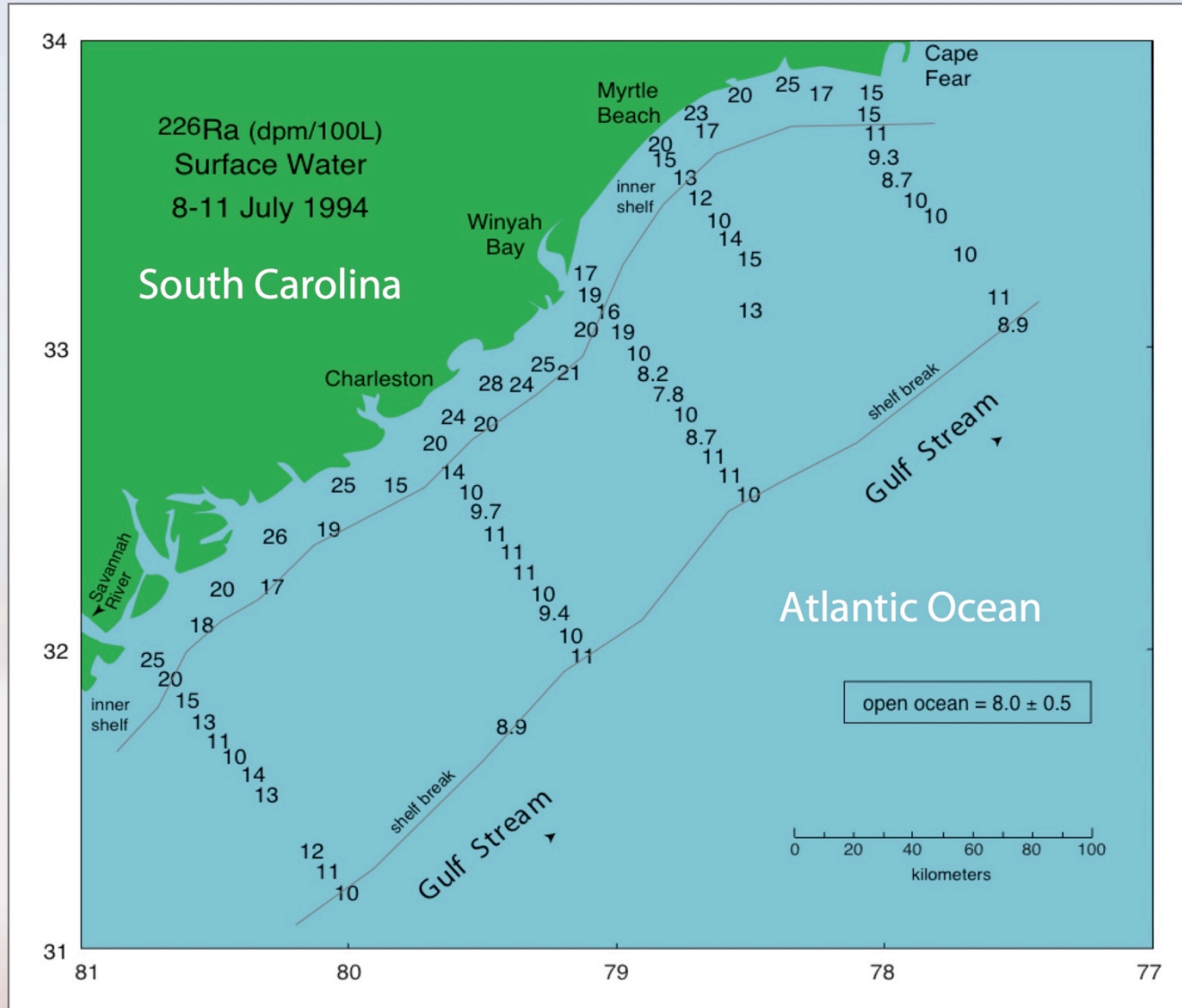


General Model for Quantifying SGD Using Radium Isotopes



Depending on the radium isotope used, various aspects of this model (e.g., decay for ^{226}Ra) can be assumed minimal.

Offshore Mixing and SGD using Radium isotopes



Moore (2000)

Offshore Mixing using ^{223}Ra & ^{224}Ra (short half-lives)

The change in concentration or activity (A) with time (t) as a function of distance offshore (x) for a radioactive tracer with decay constant (λ) may be expressed as a balance of advection (ω), dispersion (K_h), and radioactive decay.

$$\frac{dA}{dt} = K_h \left[\frac{d^2 A}{dx^2} \right] - \omega \left[\frac{dA}{dx} \right] - \lambda A$$

If $\omega = 0$:

$$\frac{dA}{dt} = K_h \left[\frac{d^2 A}{dx^2} \right] - \lambda A$$

If K_h is constant and the system is in steady state

$$A_x = A_0 \exp\left[-x \sqrt{\frac{\lambda}{K_h}}\right]$$

Moore (2000)

SGD to the southeastern N. Atlantic Ocean using ^{226}Ra

Flux of $^{226}\text{Ra} = K_h \times \text{offshore gradient}$

Flux $^{226}\text{Ra} = 400 \text{ m}^2 \text{ s}^{-1} \times 2.2 \text{ dpm m}^{-3} \text{ km}^{-1}$

$F = 7.6 \times 10^7 \text{ dpm d}^{-1} \text{ m}^{-1} \text{ km}^{-1}$ (convert seconds to days)

$F = 7.6 \times 10^{10} \text{ dpm d}^{-1} \text{ km}^{-2}$ (convert m to km)

The Ra is transported in a 10 m thick surface layer (0.01 km).

Therefore: $F = 7.6 \times 10^8 \text{ dpm d}^{-1} \text{ km}^{-1}$

For a 320 km long coast, the flux is $2.43 \times 10^{11} \text{ dpm d}^{-1}$

Moore (2010)

SGD to the southeastern N. Atlantic Ocean using ^{226}Ra

To translate the ^{226}Ra flux to a flux of SGD, we must know the concentration of ^{226}Ra in SGD.

The average concentration of ^{226}Ra in 111 analyses of coastal groundwater from the region is 1.6 dpm L^{-1} .

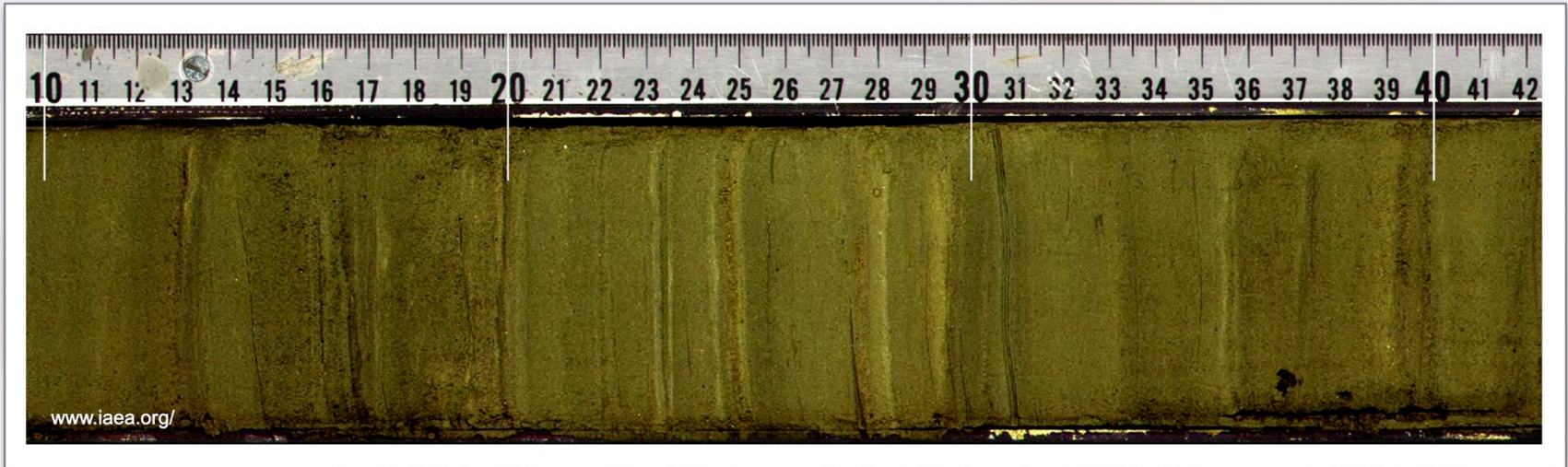
$$\text{SGD} = \frac{2.43 \times 10^{11} \text{ dpm d}^{-1}}{1.6 \text{ dpm L}^{-1}} = 15 \times 10^{10} \text{ L d}^{-1}$$

Moore (2010)

Average river flow is $7 \times 10^{10} \text{ L d}^{-1}$.

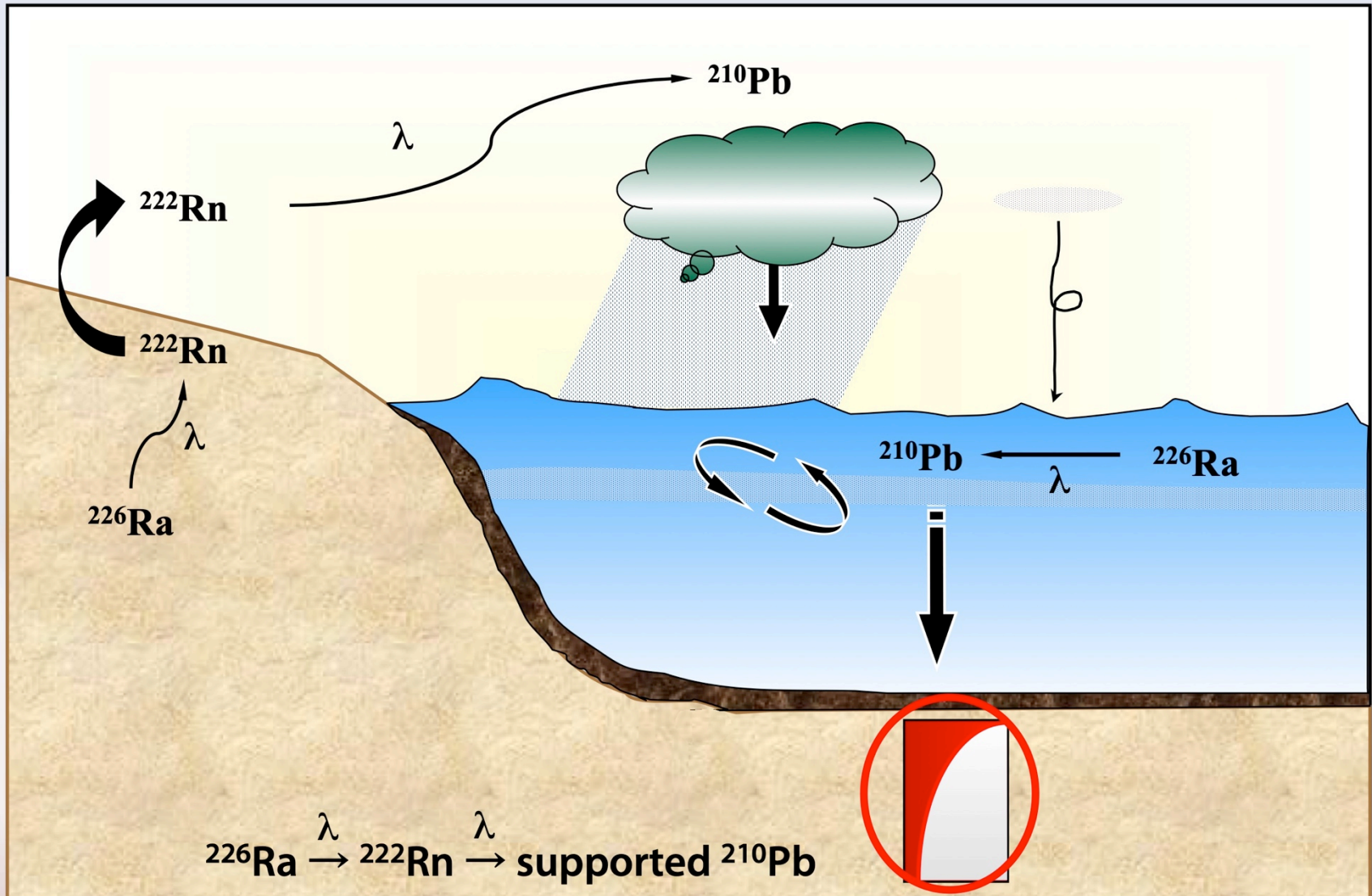
4. Sedimentation/Age Dating

Sediments provide records of processes that have occurred in the past. Sediments further serve as a storage reservoir for chemically and biologically reactive elements, such as carbon and heavy metals.

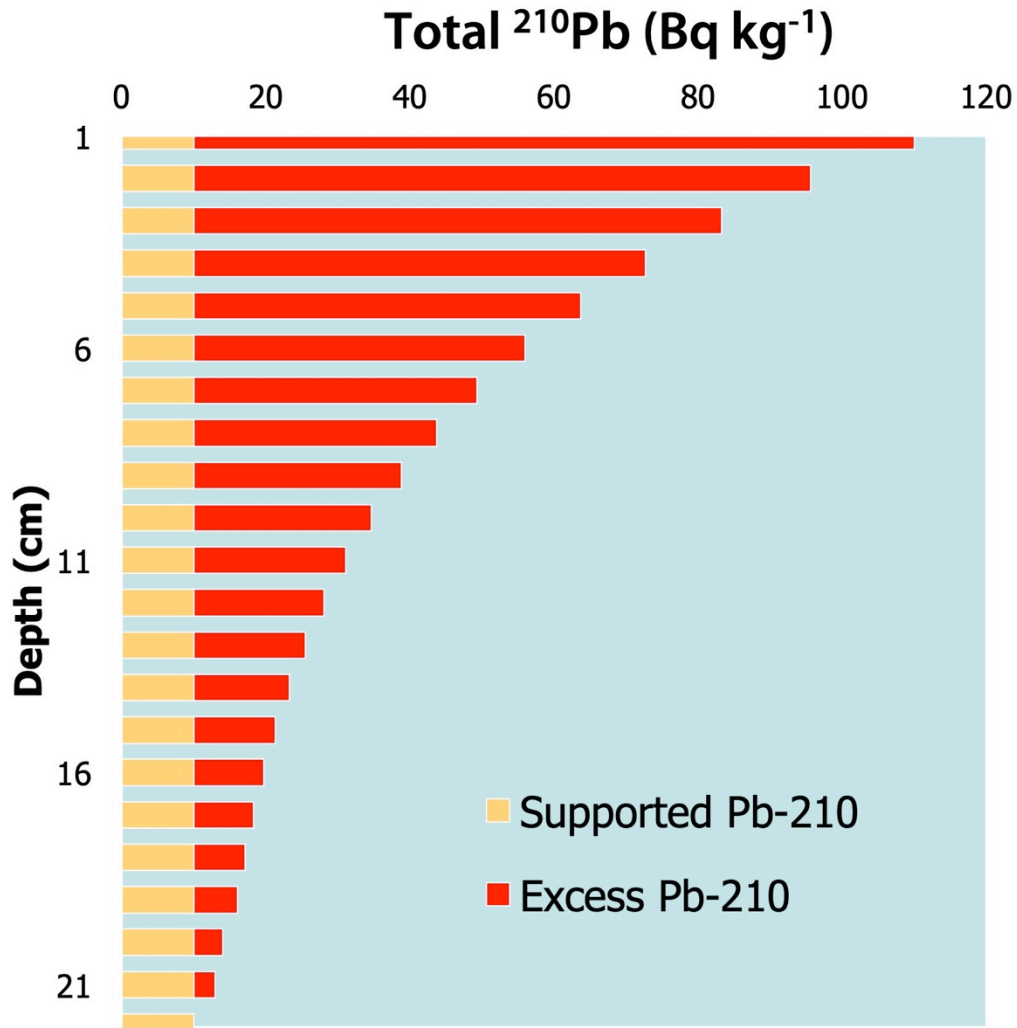


As such, knowledge of the age of sediments and rate at which sediments accumulate is critical information for answering a wide variety of questions.

The ^{210}Pb cycle in the ocean



Age dating and sedimentation/mass accumulation rates using "excess" ^{210}Pb : **The Basics**



The decay of excess ^{210}Pb can be described by the classic radioactive decay equation:

$$A = A_0 e^{-\lambda t}$$

Supported ^{210}Pb is derived from in situ decay from ^{226}Ra via ^{222}Rn

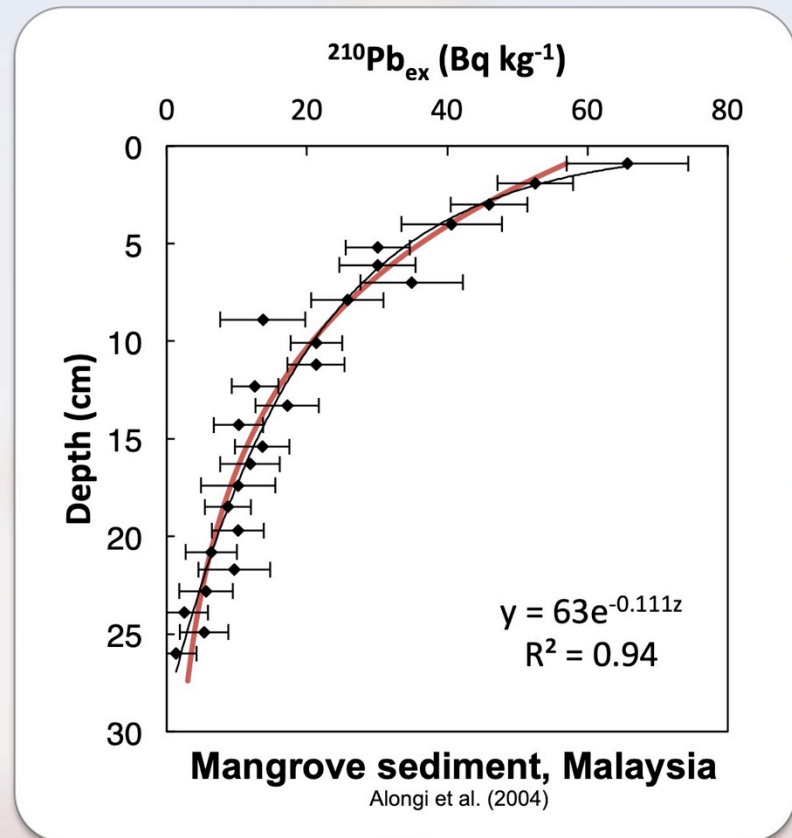
Excess ^{210}Pb is from overlying water column

^{210}Pb : Constant flux-constant sedimentation rate (CF:CS) Model

$$A_z = A_0 e^{-\lambda t} \rightarrow A_0 e^{-\left(\frac{\lambda}{S}\right)z} \rightarrow \ln A_z = \ln A_0 - \left(\frac{\lambda}{S}\right)z$$

Where:

- λ is the radioactive decay constant for ^{210}Pb (in y^{-1})
- A_0 is the excess ^{210}Pb activity at the initial depth
- A_z is the excess ^{210}Pb activity at depth, z
- S is the average sedimentation rate over the depth interval of interest.



²¹⁰Pb: Constant Initial Concentration (CIC) Model

$$A_z = A_0 e^{-\lambda t} \quad \longrightarrow \quad t = \frac{1}{\lambda} * \ln \frac{A_0}{A_z}$$

Where:

- λ is the radioactive decay constant for ²¹⁰Pb (in y⁻¹)
- A_0 is the excess ²¹⁰Pb activity at the initial depth
- A_z is the excess ²¹⁰Pb activity at depth, z

²¹⁰Pb: The constant rate of supply model (CRS)

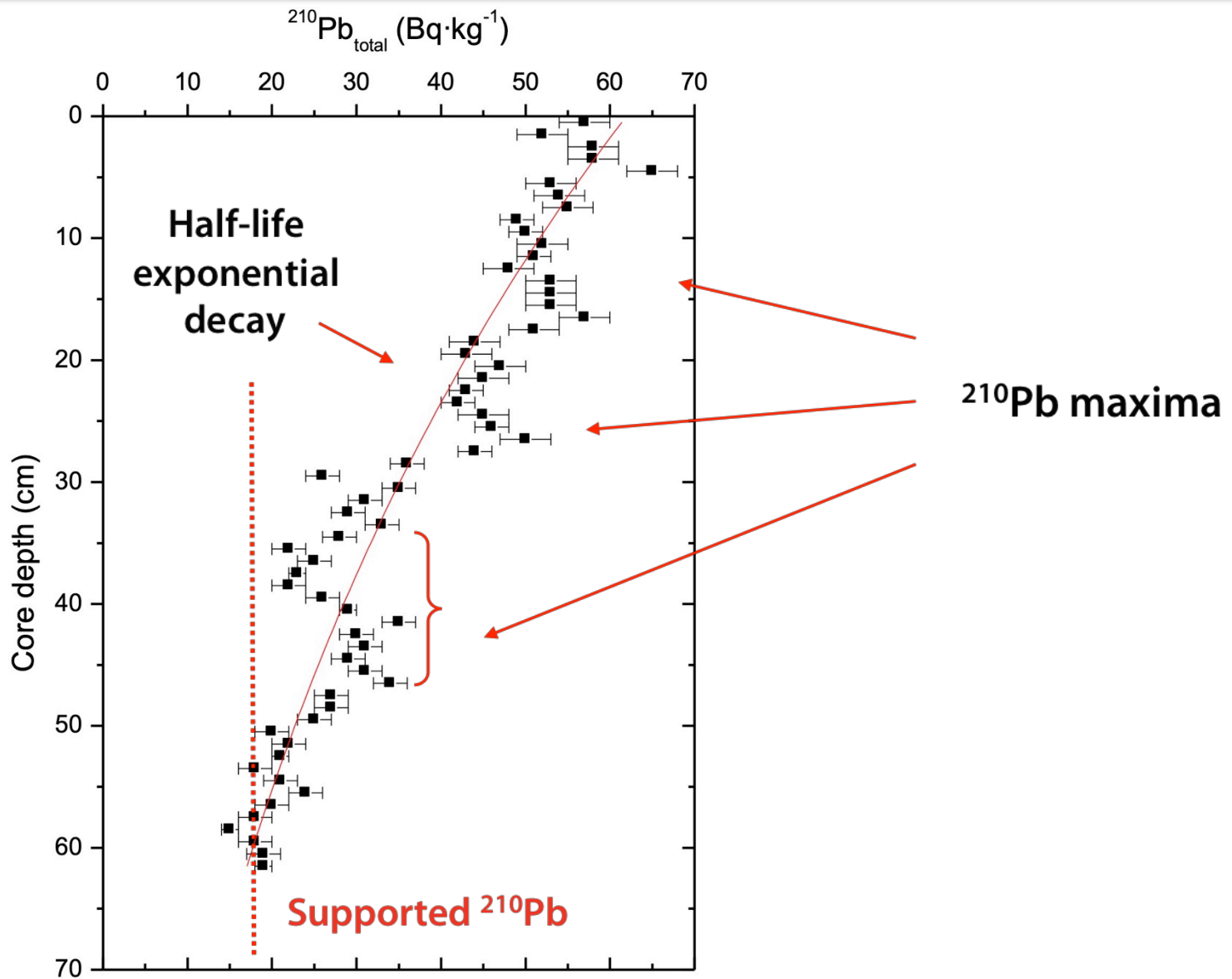
$$I_x = \int_t^{\infty} e^{-\lambda t} dt = I_0 e^{-\lambda \cdot t} \longrightarrow t_m = \frac{1}{\lambda} \left[\ln \left(\frac{I_0}{I_x} \right) \right]$$

Where:

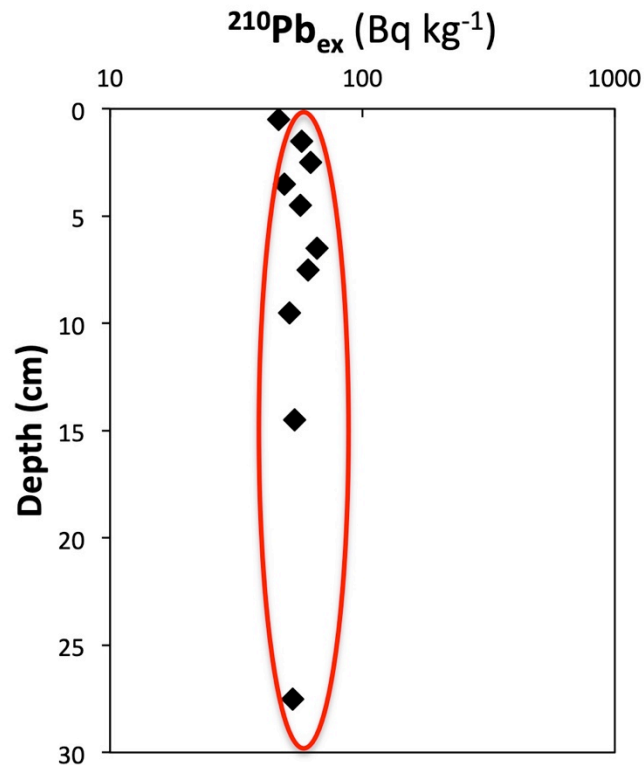
- λ is the radioactive decay constant for ²¹⁰Pb (in y⁻¹)
- A_x is the excess ²¹⁰Pb activity at depth, x (per cm⁻³)
- I_0 is the excess ²¹⁰Pb inventory of the entire core (per cm⁻²)
- I_x is the excess ²¹⁰Pb inventory beneath depth, x, of sediments (per cm⁻²)
- t_m is the sediment age at depth x

$$\text{Sedimentation rate (cm y}^{-1}\text{)} = S = \frac{\lambda I_x}{A_x}$$

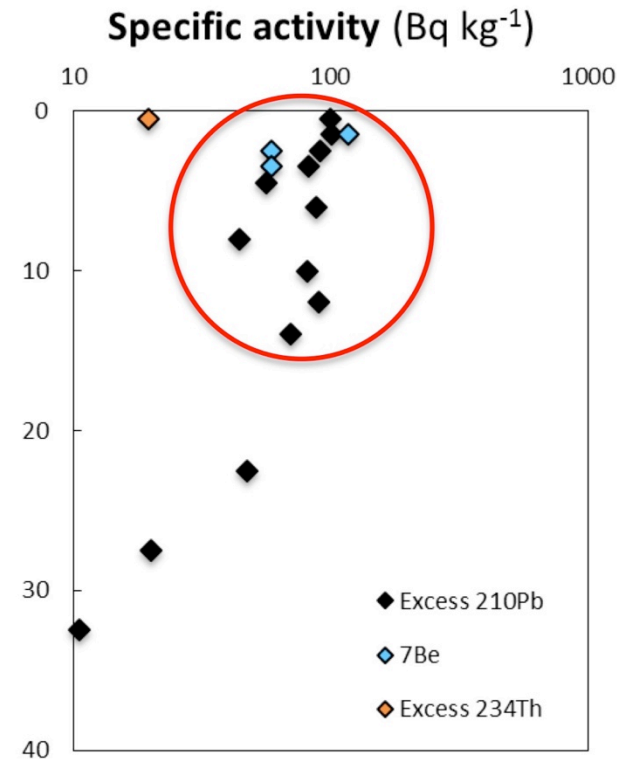
^{210}Pb : Example of a more complicated core



^{210}Pb : Example of a more complicated core



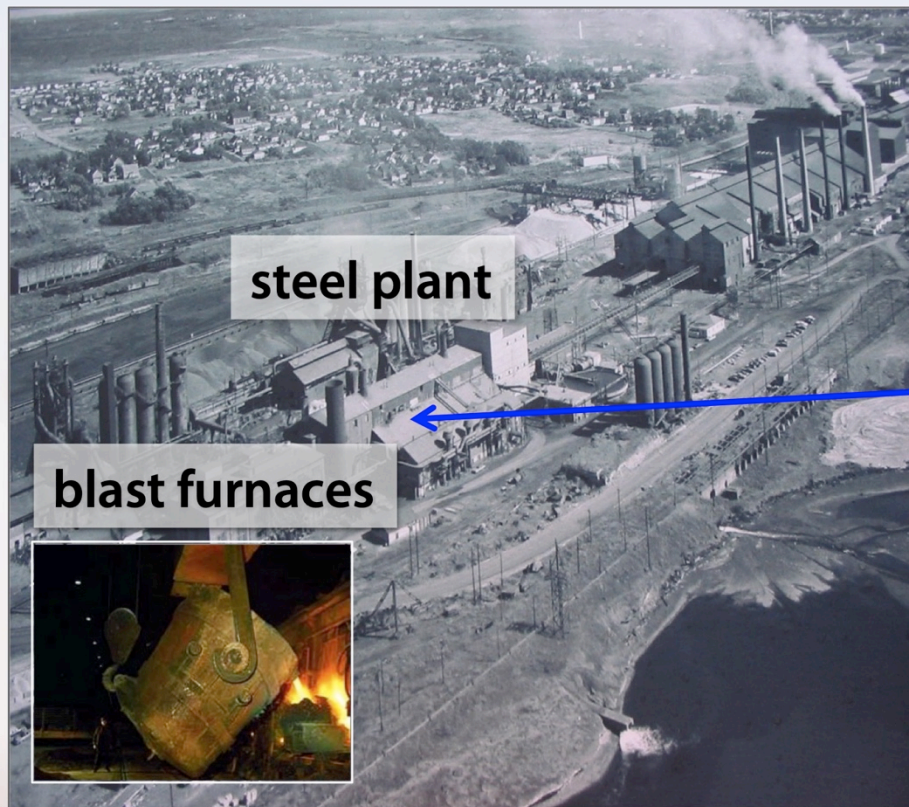
No benthic fauna
= **Physical mixing**



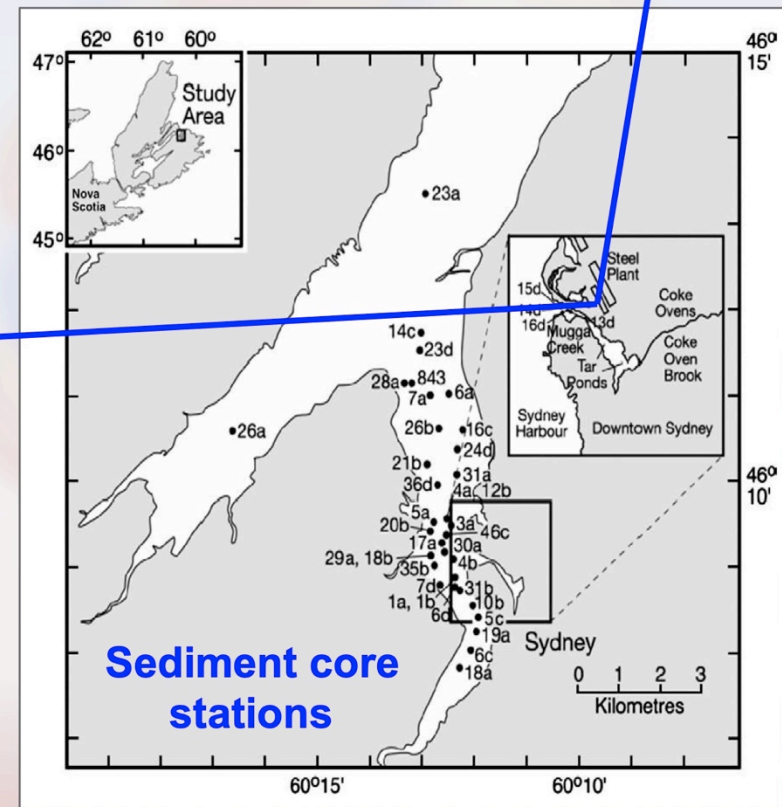
Many species of benthic fauna
 ^7Be and ^{234}Th in subsurface
layers suggest recent deposition
= **Bioturbation**

Smoak and Patchineelam (1999)

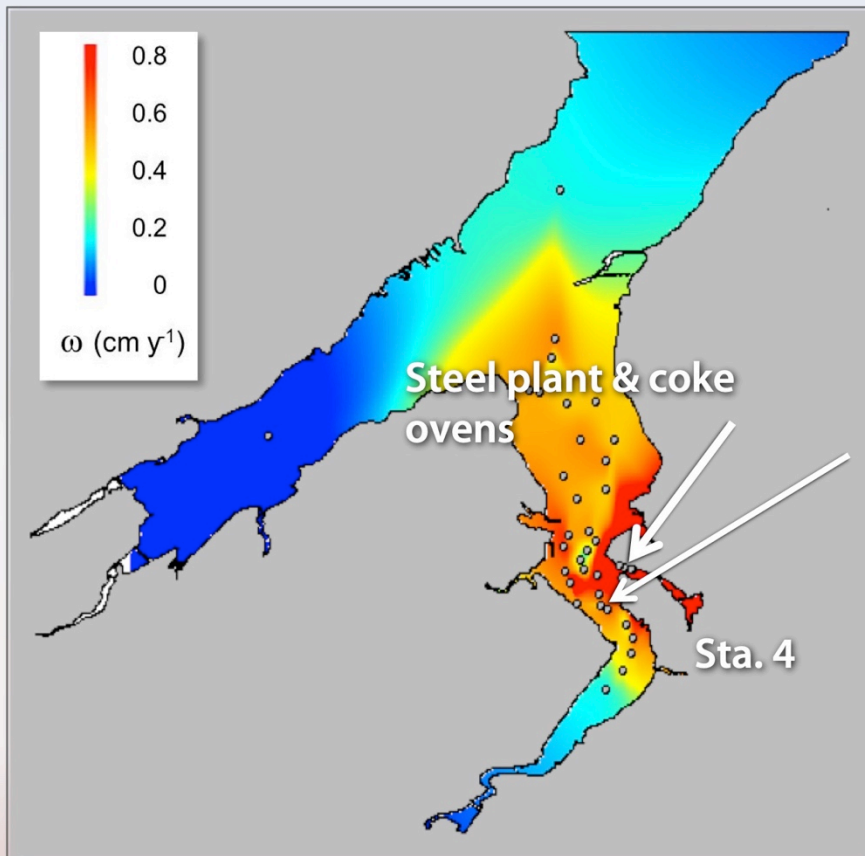
^{210}Pb applied to history of PAH contamination from steel plant/coke ovens in sediment cores from Sydney Harbour, Nova Scotia, Canada.



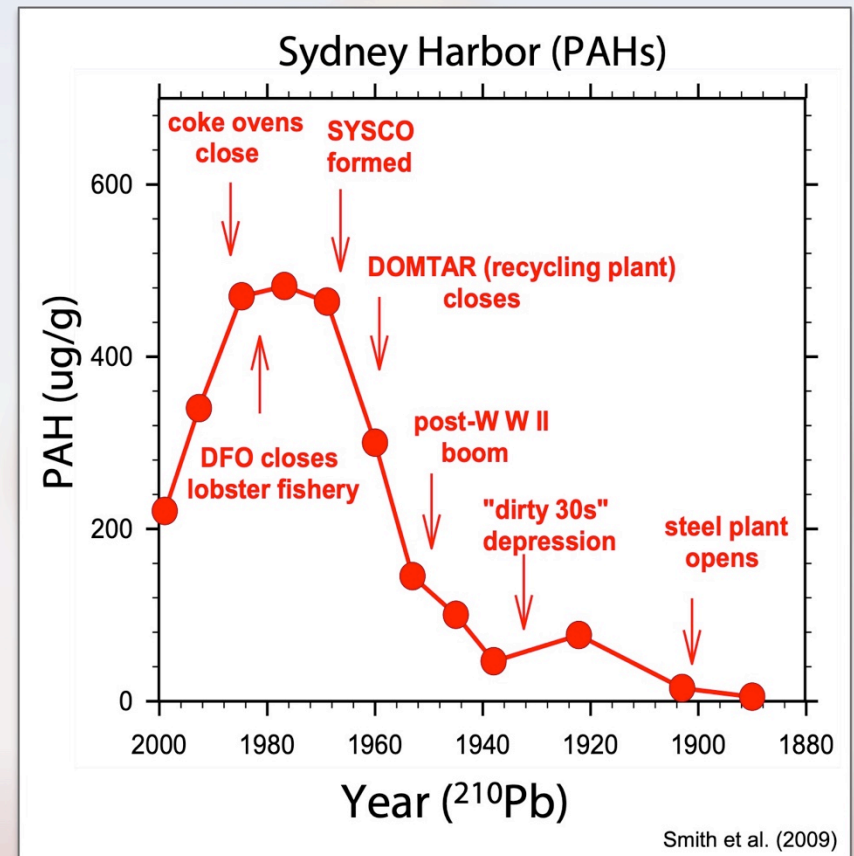
Smith et al. (2009)

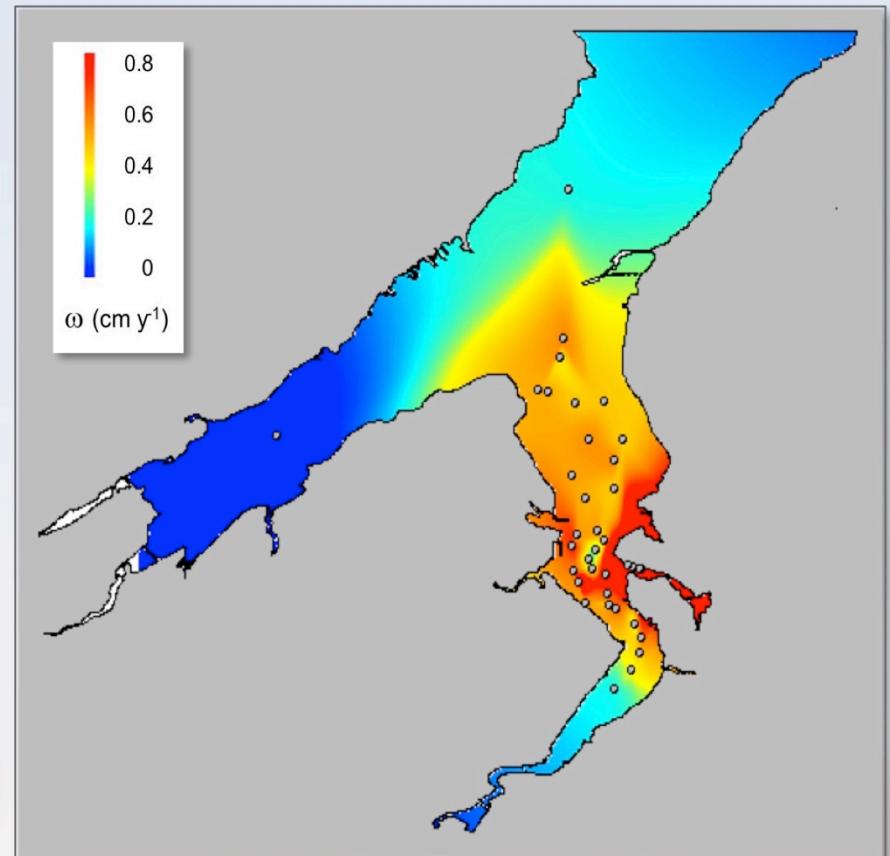
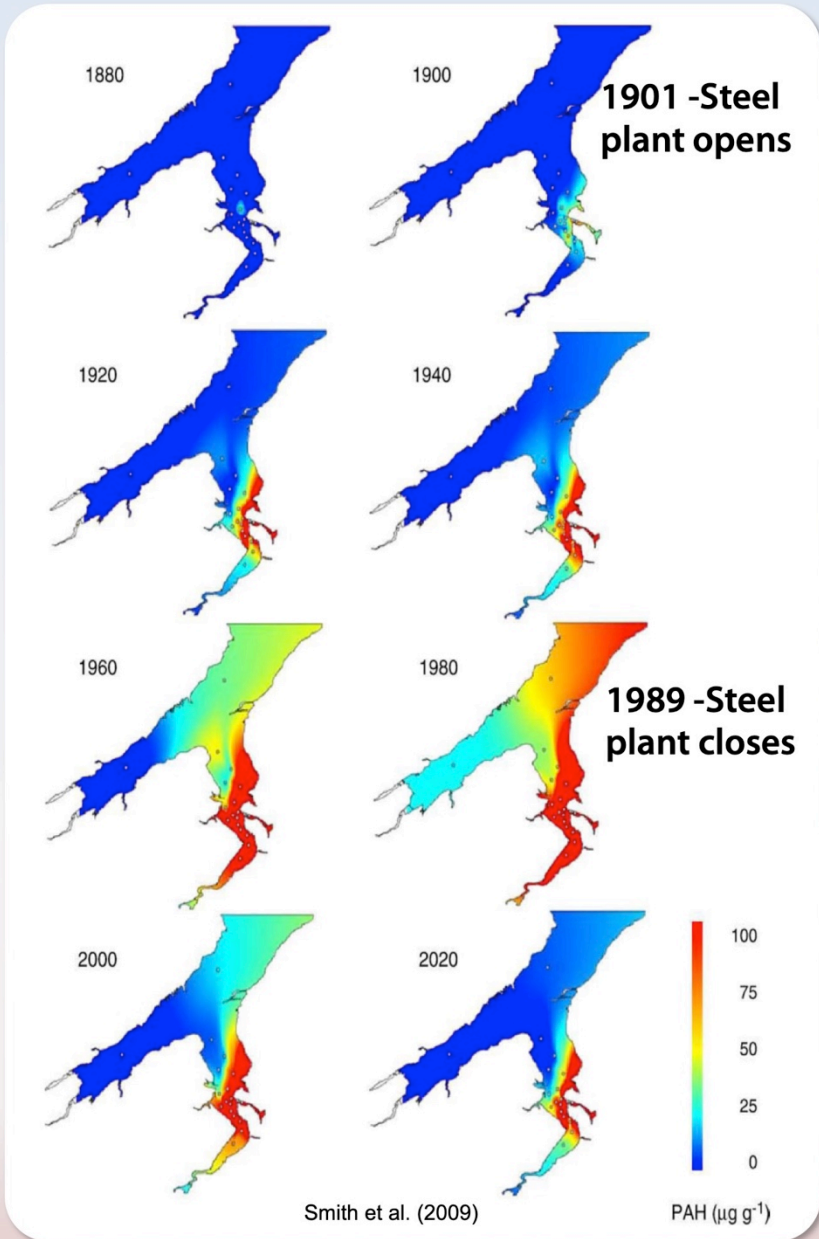


^{210}Pb derived sedimentation rates ω (cm y^{-1}) for 41 cores in Sydney Harbour. Below right is PAH history for Sta. 4. PAH decrease near core top reflects closure of steel mill and coke ovens in 1989.



PAH (Sta 4) plotted vs. ^{210}Pb date gives core PAH geochronology. Time slices through data set of 41 cores provide surface concentration maps for entire history of harbour.





Historical PAH surface concentration maps based on ^{210}Pb core dates. Extrapolation to 2020 is based on transport model using watershed residence time for PAHs of 10 years.

In summary

U-Th series radionuclides are very useful tools as **tracers** to study a **large variety of processes** in the oceans at various **time scales** from days to millions of years.

Examples include:

- Scavenging
- Air-Sea exchange
- Tracing groundwater discharge
- Sedimentation rates

Acknowledgements

This work would not have been possible without the generous contributions and thoughtful comments of Drs. Robert Anderson, Kirk Cochran, Peter Santschi and Alan Shiller. We wish to thank two anonymous reviewers who provided constructive comments that improved the presentation. Lectures would not have been possible without the outstanding assistance of graphic designer Jason Emmett.

This work was supported by the Scientific Committee on Ocean Research, SCOR Working Group 146: *Radioactivity in the Ocean, 5 Decades Later (RiO5)* and the Woods Hole Oceanographic Institution's *Center for Marine and Environmental Radioactivity*.

References

- Alongi, D.M., Sasekumar, A., Chong, V.C., Pfitzner, J., Trott, L.A., Tirendi, F., Dixon, P., Brunskill, G.J., (2004) Sediment accumulation and organic material flux in a managed mangrove ecosystem: estimates of land-ocean-atmosphere exchange in peninsular Malaysia. *Mar. Geol.* 208: 383–402.
- Bateman, H. (1910), The Transformation of the Electrodynamical Equations. *Proceedings of the London Mathematical Society*, s2-8: 223–264. doi:10.1112/plms/s2-8.1.223.
- Bhat, S. G., Krishnaswami, S., Lal, D., Rama, Moore, W.S. (1969) $^{234}\text{Th}/^{238}\text{U}$ ratios in the Ocean, *Earth and Planetary Science Letters*, 32: 277-296.
- Broecker, W.S. and Peng, T-H (1982) *Tracers in the sea*. New York: Lamont-Doherty Geological Observatory. Eldigio Press, Columbia University. Available online at: http://www.ldeo.columbia.edu/~broecker/Home_files/TracersInTheSea_searchable.pdf
- Buesseler, K. O., Benitez-Nelson, C.R., Moran, S.B., Burd, A., Charette, M., Cochran, J.K., Coppola, L., Fisher, N.S., Fowler, S.W., Gardner, W.D., Guo, L.D., Gustafsson, O., Lamborg, C., Masque, P., Miquel, J.C., Passow, U., Santschi, P.H., Savoye, N., Stewart, G., Trull, T. (2006) An assessment of particulate organic carbon to thorium-234 ratios in the ocean and their impact on the application of ^{234}Th as a POC flux proxy. *Marine Chemistry*, 100: 213-233.
- Buesseler, K.O., Pike, S., Maiti, K., Lamborg, C.H., Siegel, D.A., Trull, T.W. (2009) Thorium-234 as a tracer of spatial, temporal and vertical variability in particle flux in the North Pacific. *Deep Sea Research Part I: Oceanographic Research Papers* 56: 1143-1167.
- Coale, K.H., Bruland, K.W. (1985) $^{234}\text{Th}:$ ^{238}U disequilibria within the California Current. *Limnology and Oceanography* 30: 22-33.
- Coale, K.H., Bruland, K.W. (1987) Oceanic stratified euphotic zone as elucidated by $^{234}\text{Th}:$ ^{238}U disequilibria. *Limnology and Oceanography* 32: 189-200.
- Cochran, J.K., Masqué, P. (2003) Short-lived U/Th Series Radionuclides in the Ocean: Tracers for Scavenging Rates, Export Fluxes and Particle Dynamics. *Reviews in Mineralogy and Geochemistry* 52: 461-492.
- Goldberg, E.D. (1954) Marine geochemistry 1. Chemical scavengers of the sea. *J. Geol.* 62: 249–265.
- Loose, B., McGillis, W.R., Perovich, D., Zappa, C.J., Schlosser, P. (2014) A parameter model of gas exchange for the seasonal sea ice zone, *Ocean Sci.*, 10: 17–28.
- Moore, W.S. (2000) Determining coastal mixing rates using radium isotopes. *Continental Shelf Research* 20: 1993-2007.
- Moore, W.S. (2010) A reevaluation of submarine groundwater discharge along the southeastern coast of North America. *Global Biogeochemical Cycles*, 24: 10.1029/2009GB003747
- Nozaki, Y. (2001) Elemental distribution: Overview. In: *Encyclopedia of Ocean Sciences*, vol. 2. J.H. Steele, S.A. Thorpe, and K.K. Turekian, eds, Academic Press, London, Pp. 840–845 <http://dx.doi.org/10.1006/rwos.2001.0402>
- Owens S.A., Buesseler K.O., Sims K.W.W. (2011) Re-evaluating the ^{238}U -salinity relationship in seawater: Implications for the ^{238}U – ^{234}Th disequilibrium method. *Marine Chemistry* 127: 31-39.
- Peng, T.-H., Broecker, W.S., Mathieu, G.G., Li, Y.H., Bainbridge, A.E. (1979) Radon evasion rates in the Atlantic and Pacific oceans as determined during the GEOSECS program. *Journal of Geophysical Research* 84: 2,471–2,486

References

- Puigcorbé, V., Roca-Martí, M., Masqué, P., Benitez-Nelson, C., Rutgers van der Loeff, M., Bracher, A., Moreau, S. (2017). Latitudinal distributions of particulate carbon export across the North Western Atlantic Ocean. *Deep Sea Research Part I: Oceanographic Research Papers*, 129: 116-130. <https://doi.org/10.1016/j.dsr.2017.08.016>.
- Rodellas, V., Garcia-Orellana, J., Masqué, P., Feldman, M., Weinstein, Y. (2015) Submarine groundwater discharge as a major source of nutrients to the Mediterranean Sea. *Proceedings of the National Academy of Sciences* 112: 3926-3930.
- Rutgers van der Loeff, M.M. (2014a) Uranium-Thorium Decay Series in the Oceans: Overview. In Elias S (ed.) *Earth Systems and Environmental Sciences*, (Reference Module in Earth Systems and Environmental Sciences). Amsterdam: Elsevier 1-16.
- Rutgers van der Loeff, M.M. et al. (2014b): The influence of sea ice cover on air-sea gas exchange estimated with radon-222 profiles. *Journal of Geophysical Research-Oceans*, 119(5): 2735-2751
- Santschi, P.H., Honeyman, B.D. (1989) Radionuclides in aquatic environments, *Radiat. Phys. Chem.*, 34: 213–240
- Smith, J.N., Lee K., Gobeil, C., Macdonald, R.W. (2009) Natural rates of sediment containment of PAH, PCB and metal inventories in Sydney Harbour, Nova Scotia. *Science of The Total Environment* 407: 4858-4869.
- Smoak, J.M., Patchineelam, S.R. (1999) Sediment mixing and accumulation in a mangrove ecosystem: evidence from ^{210}Pb , ^{234}Th and ^7Be . *Mangroves and Salt Marshes* 3: 17-27.

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Paul J. Morris, Michiel Rutgers van der Loeff, John N. Smith**

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Uranium-Thorium Decay Series***

doi:10.1002/loe2.10009

e-Lecture received June 2016; accepted October 2018

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