Radioactivity in the Marine Environment

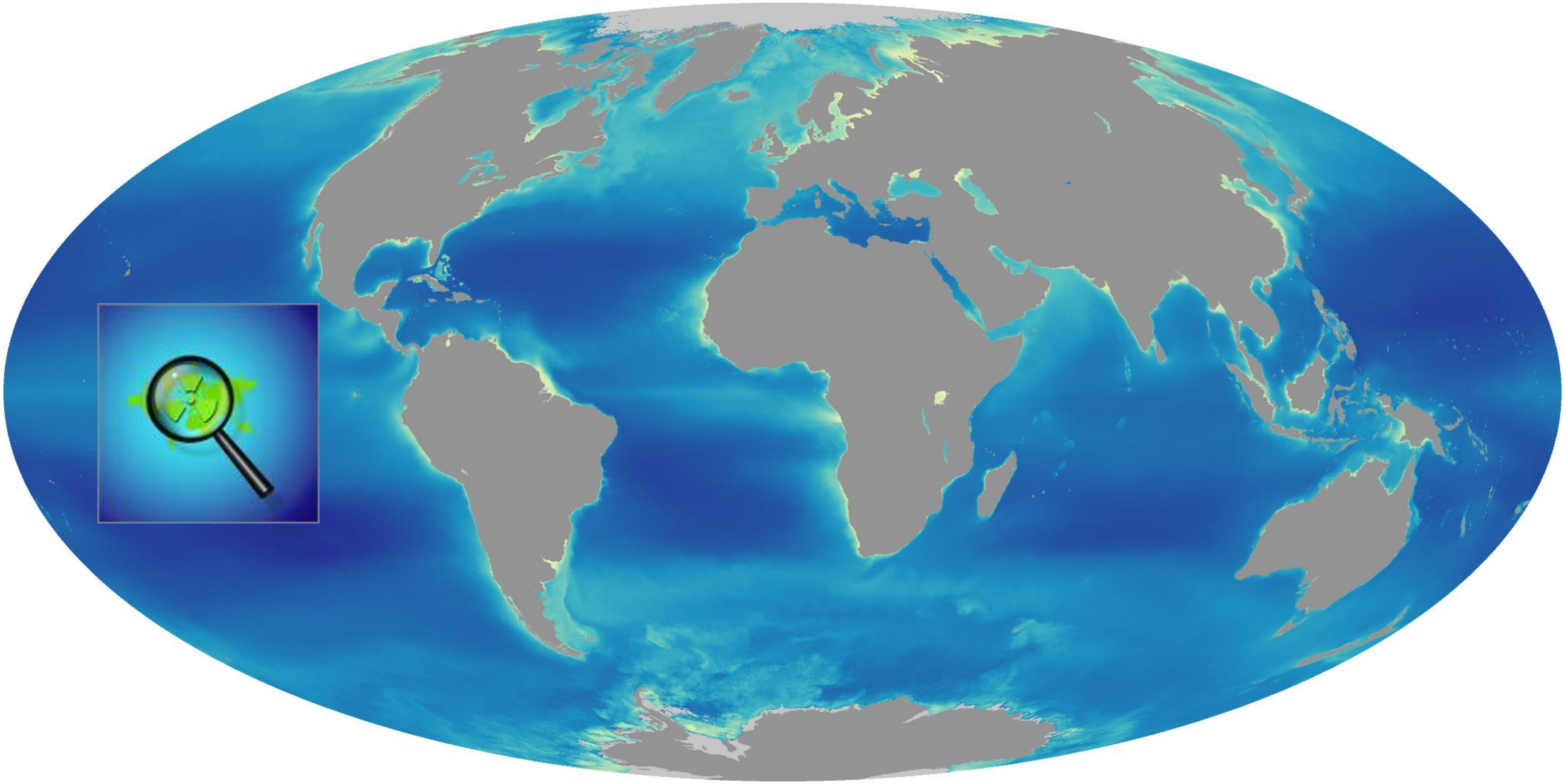
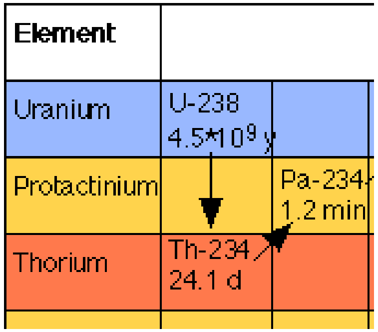


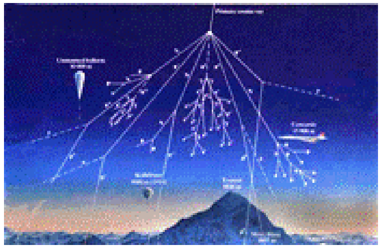
Image: http://earthobservatory.nasa.gov/IOTD/view.php?id=8129

Radionuclides in the Marine Environment

The focus of this lecture is to understand:

1) The 3 main sources of radionuclides to the marine environment





2) Key examples of how radionuclides can be used to trace a variety of ocean processes.

1988

Three main sources of radionuclides to Marine Environment:

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

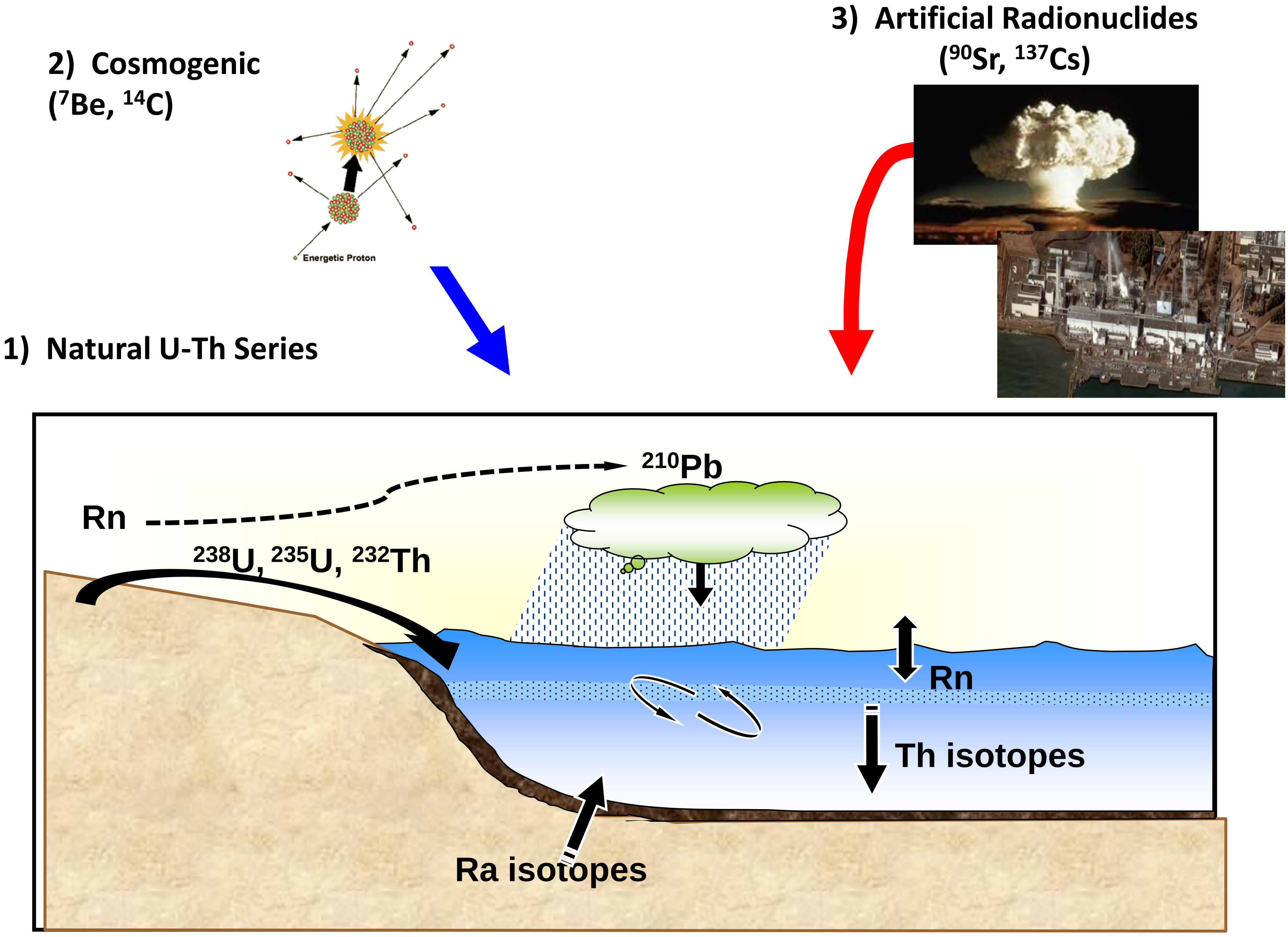
Examples: ^{238}U, ^{234}Th, ^{232}Th, ^{210}Pb and ^{222}Rn

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

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

3) Artificial radionuclides – continuously being produced by humans.

Examples: ^{90}Sr, ^{137}Cs, ^{239}Pu


2) Cosmogenic
(^{7}Be, ^{14}C)
Energetic Proton
3) Artificial Radionuclides
(^{90}Sr, ^{137}Cs)
1) Natural U-Th Series
^{210}Pb
Rn
^{238}U, ^{235}U, ^{232}Th
Rn
Th isotopes
Ra isotopes

Radionuclide distributions in the ocean are controlled by:

1) Geochemical and physical properties of elements

Particle reactive radionuclides (Th, Pb, Po) are tracers of:
- "scavenging" processes (Goldberg, 1954)
- "export"
- "particle flux"

Conservative tracers (U, Ra) are tracers of:
- physical processes,
- including horizontal and vertical mixing, dilution

Gas exchange (Rn)

2) Half lives for radioactive decay determine processes that can be traced

days- ^{224}Ra
months- ^{234}Th
years- ^{228}Th
decades- ^{210}Pb
millennia- ^{231}Pa

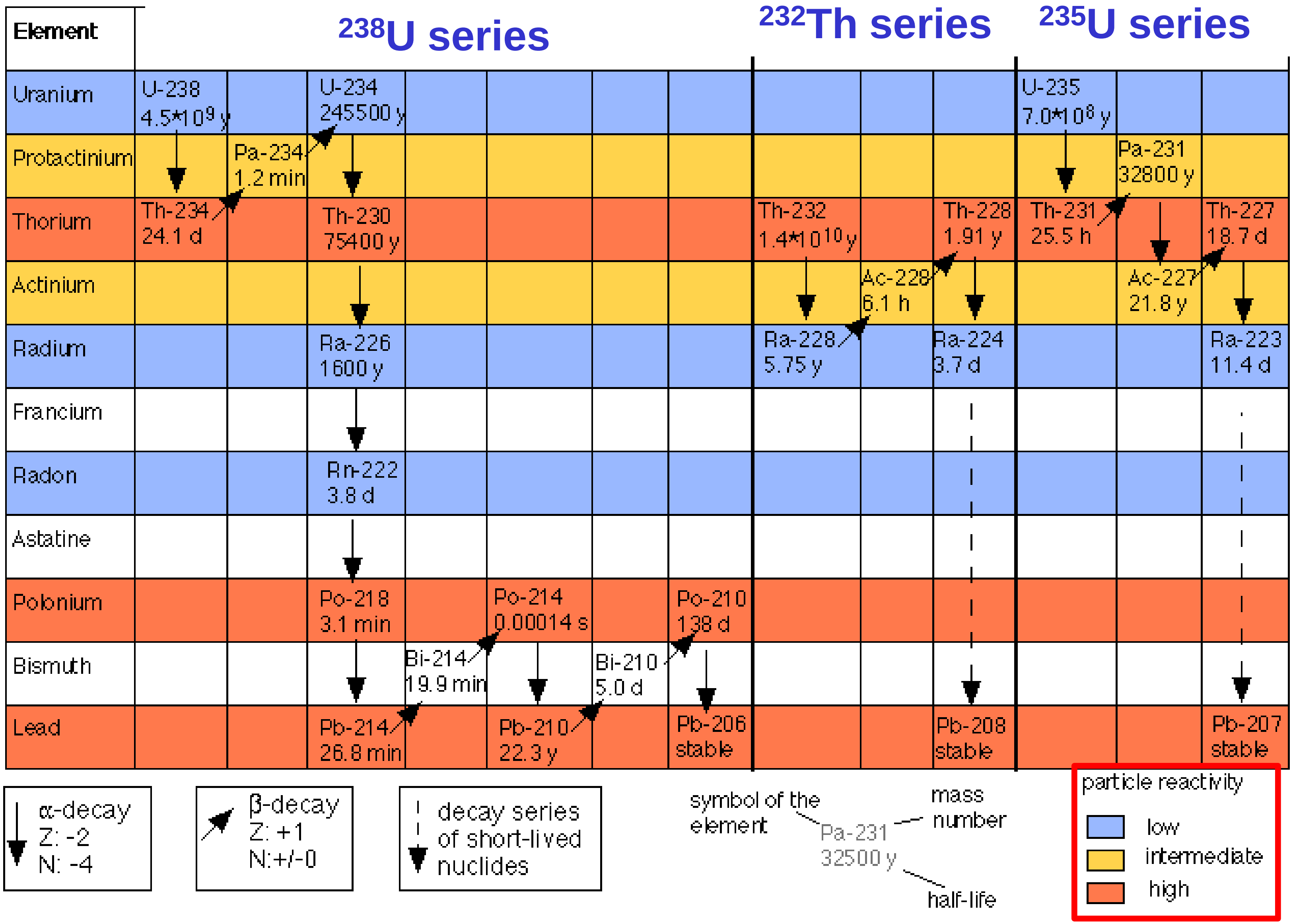
Reservoir	Process	Parameter (examples)	Tracer nuclides	Reference
Atmosphere	Aerosol Scavenging	Residence time	$^{7}Be/^{10}Be$ $^{222}Rn/^{210}Pb/^{210}Po$	Raisbeck *et al.*, (1981), Bleichrodt, (1978) Martell and Moore (1974), Robbins (1978)
	Dry and wet dep.	Rates	^{131}I, ^{137}Cs, . . .	Wiffen (1958), Stewart and Crooks (1958) Santschi *et al.* (1988a)
	Atmospheric circulation	Rates	^{14}C, ^{37}Ar,	Loosli *et al.* (1973), Feely *et al.* (1966)
Soil	Mechanical and chemical erosion	Residence time in top soil	^{210}Pb, $^{239,240}Pu$ ^{137}Cs, ^{134}Cs ^{10}Be	Lewis (1979), Simpson *et al.* (1976) Dominik *et al.* (1987) Monaghan *et al.* (1983)
	Water movement	Rates	^{3}H	Höhn and Santschi (1987), Santschi *et al.* (1987b)
Ocean and lake water	Horizontal	Diffusion	^{228}Ra, ^{222}Rn,	Sarmiento and Roth (1980), Sarmiento *et al.* (1982), Moore and Santschi (1986), Broecker and Peng (1982)
	or vertical	coefficient	^{3}He, ^{3}H	Imboden and Emerson (1978), Roether *et al.* (1970), Imboden and Joller (1984), Li *et al.* (1984), Broecker and Peng (1982)
	diffusion		^{39}Ar, ^{85}Kr	Loosli (1983), Schlitzer *et al.* (1985)
	Horizontal	Rates	^{231}Pa, ^{230}Th,	Anderson *et al.* (1983), Bacon and Rosholt (1982), Broecker and Peng (1982)
	or vertical	Mechanisms	^{210}Pb, $^{239,240}Pu$	Carpenter and Beasley (1981), Beasley *et al.* (1982), Moore *et al.* (1981), Santschi *et al.* (1980b), Scott *et al.* (1983), Nyffeler *et al.* (1986, 1984)
	scavenging		^{10}Be ^{234}Th, ^{228}Th	Mangini (1984) Santschi *et al.* (1979, 1980a, 1983a), Honeyman and Santschi (1989)
	Sediment	Rates	^{234}Th, ^{228}Th	Aller and Cochran (1976), Broecker and Peng (1982)
	resuspension	Mechanisms	$^{134,137}Cs$, ^{7}Be, ^{210}Pb	Robbins and Eadie (1982), Santschi (1987a), Santschi *et al.* (1987c)
	Gas exchange	Rates	^{222}Rn, ^{14}C, $^{3}H/^{3}He$	Broecker and Peng (1982), Torgerson *et al.* (1977, 1982)
	Evaporation	Rates	^{3}H	Herczeg and Imboden (1988)
Sediments	Bioturbation	Rates	^{234}Th, ^{210}Pb, ^{14}C	Aller and Cochran (1976), Broecker and Peng (1982), Krishnaswami *et al.* (1984)
			$^{239,240}Pu$, ^{137}Cs	Santschi *et al.* (1980b, 1983b)
	Sedimentation	Rates	^{230}Th, ^{231}Pa, ^{10}Be	Armi *et al.* (1975), Broecker and Peng (1982)
			^{137}Cs, ^{134}Cs, ^{210}Pb	Wan *et al.* (1987), Robbin and Edington (1975), Robbins (1978)
	Diagenetic remobilization	Rates	^{234}U, ^{238}U, ^{230}Th, ^{234}Th	Osmond and Cowart (1982). Cochran *et al.* (1986), Cochran and Krishnaswami (1980), Colley *et al.* (1984), Colley and Thompson (1985)
	or fixation		$^{239,240}Pu$	Santschi *et al.* (1983b)
Groundwater	Sorption	Residence times	^{234}Th, ^{224}Ra, ^{226}Ra, ^{222}Rn, ^{210}Pb	Krishnaswami *et al* (1982) Hussain and Krishnaswami (1980) Rama and Moore (1984)
	Rock–water interaction	Rates	^{14}C	Mazor *et al.* (1986)
	Mixing	Rates	^{36}Cl, ^{81}Kr	Fabryka-Martin *et al.* (1987), Lehmann *et al.* (1986)

Many options

Table of radionuclides used to quantify geochemical processes (Santschi and Honeyman, 1989)

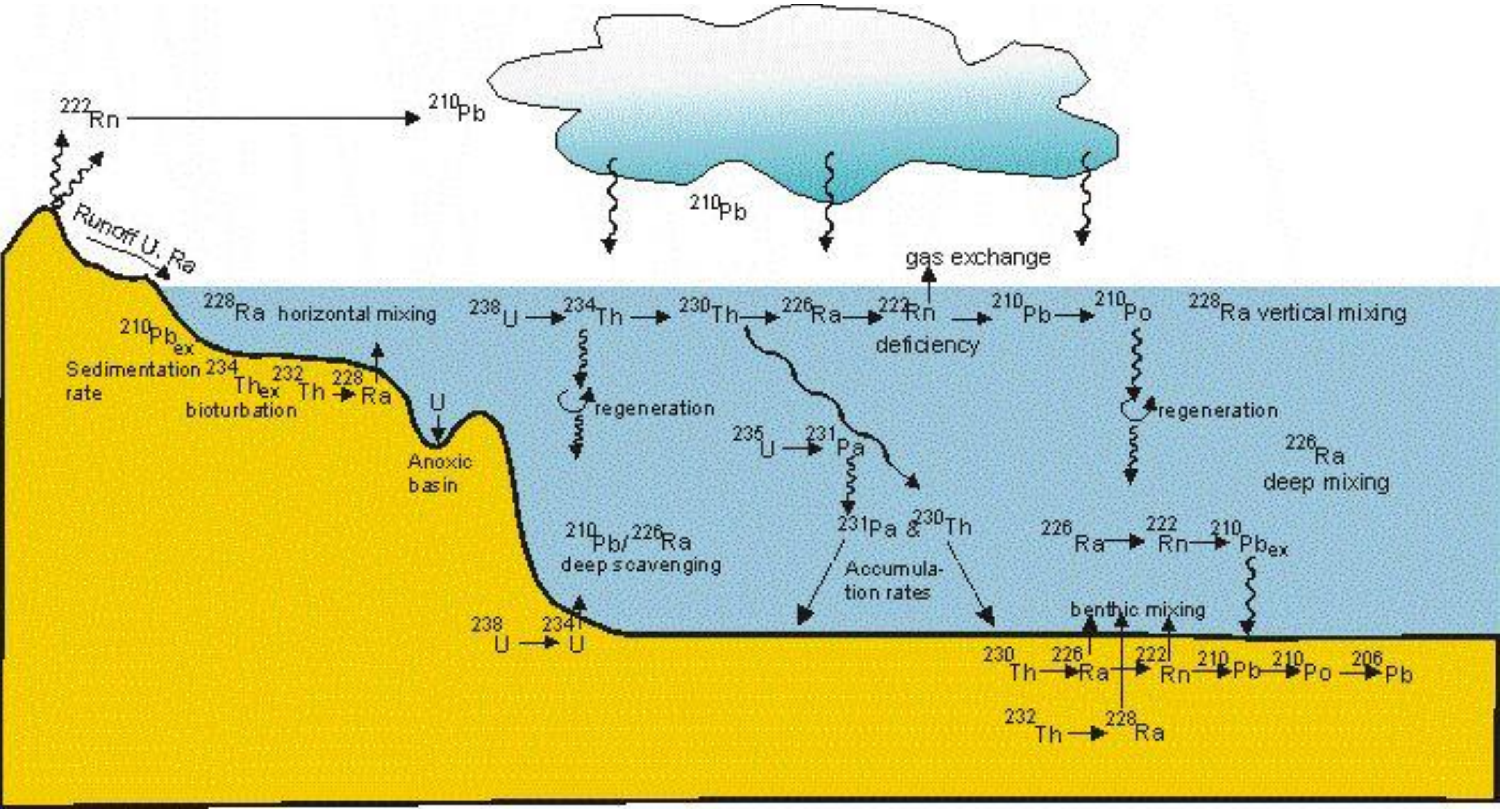
& more waiting to be discovered!

U-Th series decay series and ocean properties



Activities in the marine environment depend upon:
1) source, 2) chemistry of element, 3) decay

U-Th series decay chains



http://www.awi.de/en/research/research_divisions/geosciences/marine_geochemistry/research_themes/natural_radionuclides_and_geotraces/tracers_for_transport_processes/

U-Th series decay chains

Brief Overview:

1) Uranium

- soluble in seawater - “conservative” tracers
- source from weathering
- decays to Th in oceans and marine sediments
- multiple isotopes of interest: ^{238}U, ^{235}U, ^{234}U
- half-life > 10^5 – 10^9 years

2) Thorium

- particle reactive in seawater- “scavenging” tracers
- multiple isotopes: ^{230}Th, ^{228}Th, ^{234}Th
- short/long time scales- days to 10^4 years

3) Radium/Radon

- soluble- “conservative” tracers
- multiple Ra isotopes- ^{226}Ra, ^{228}Ra, ^{223}Ra, ^{224}Ra
- Rn used for gas exchange (^{222}Rn)
- both enriched in sediments and groundwater
- short to long time scales- days to 10^4 years

U-Th series decay chains

4) Lead/Polonium

- particle reactive - scavenging tracers
- ^{210}Pb & ^{210}Po disequilibrium used for scavenging
- ^{210}Pb tracer of sediment mixing/accumulation
- Time scales of 100’s days to 100 years

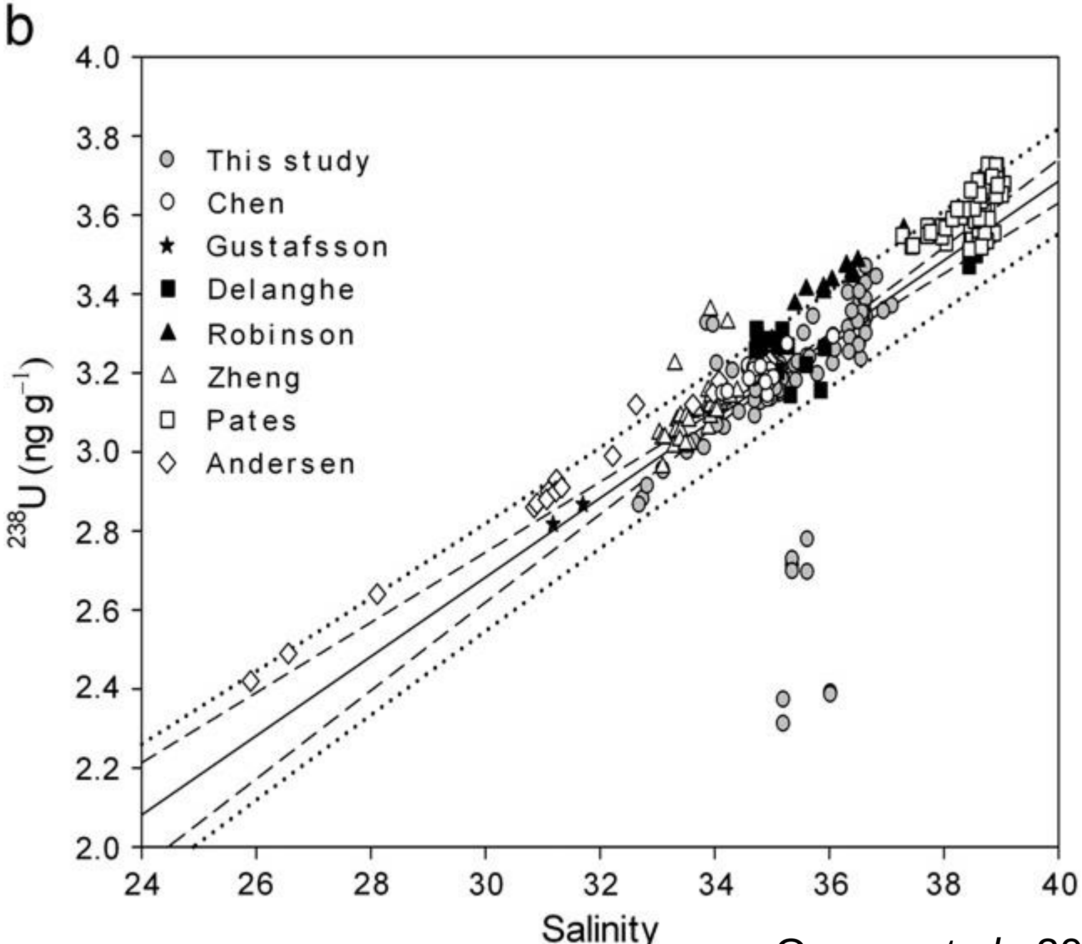
5) Protactinium

- scavenging tracer (^{231}Pa & ^{230}Th pair)
- long time scales- paleoflux
- ^{231}Pa: 10^4 year half life

6) Actinium

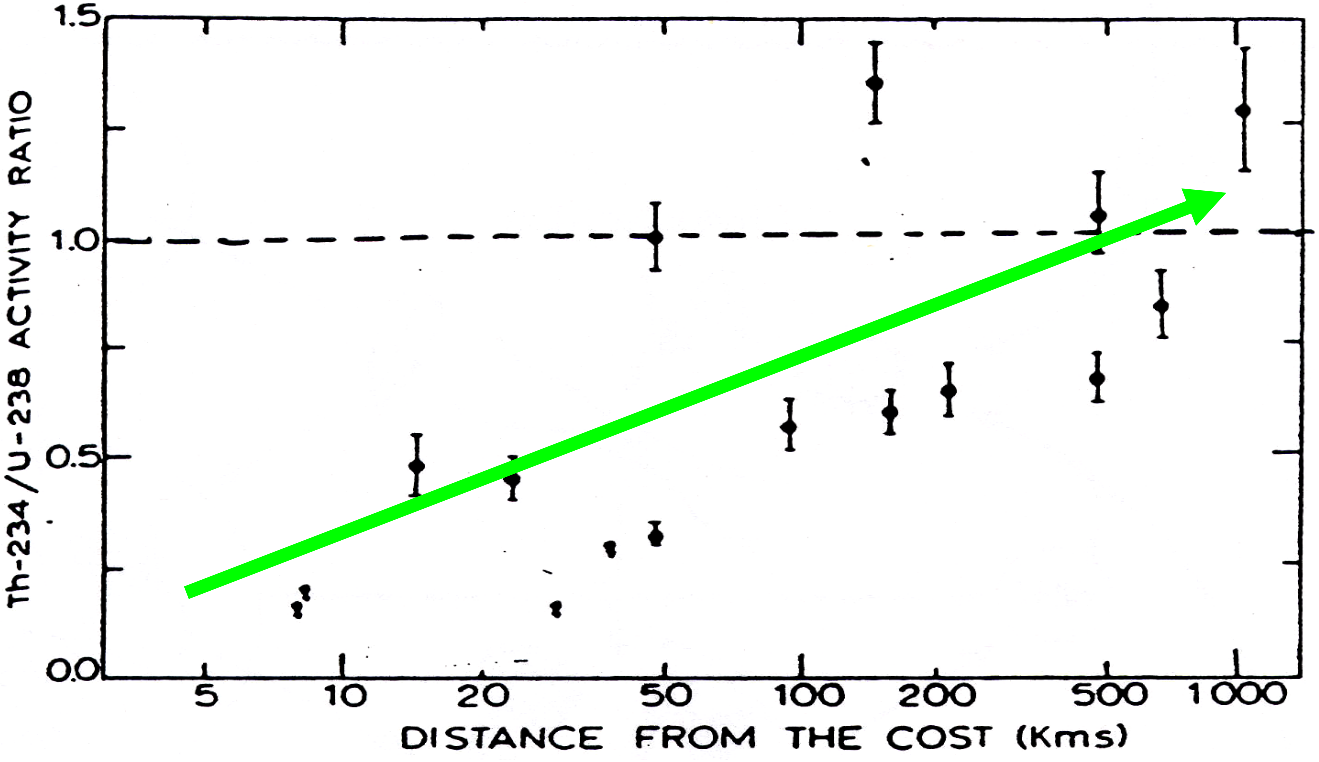
- conservative tracer
- relatively soluble in seawater
- less commonly used
- ^{227}Ac: 22 year half life

Key examples of U-Th series-
U is soluble, conservative in seawater



Owens *et al*., 2011

Key examples of U-Th series scavenging using ^{234}Th in the ocean



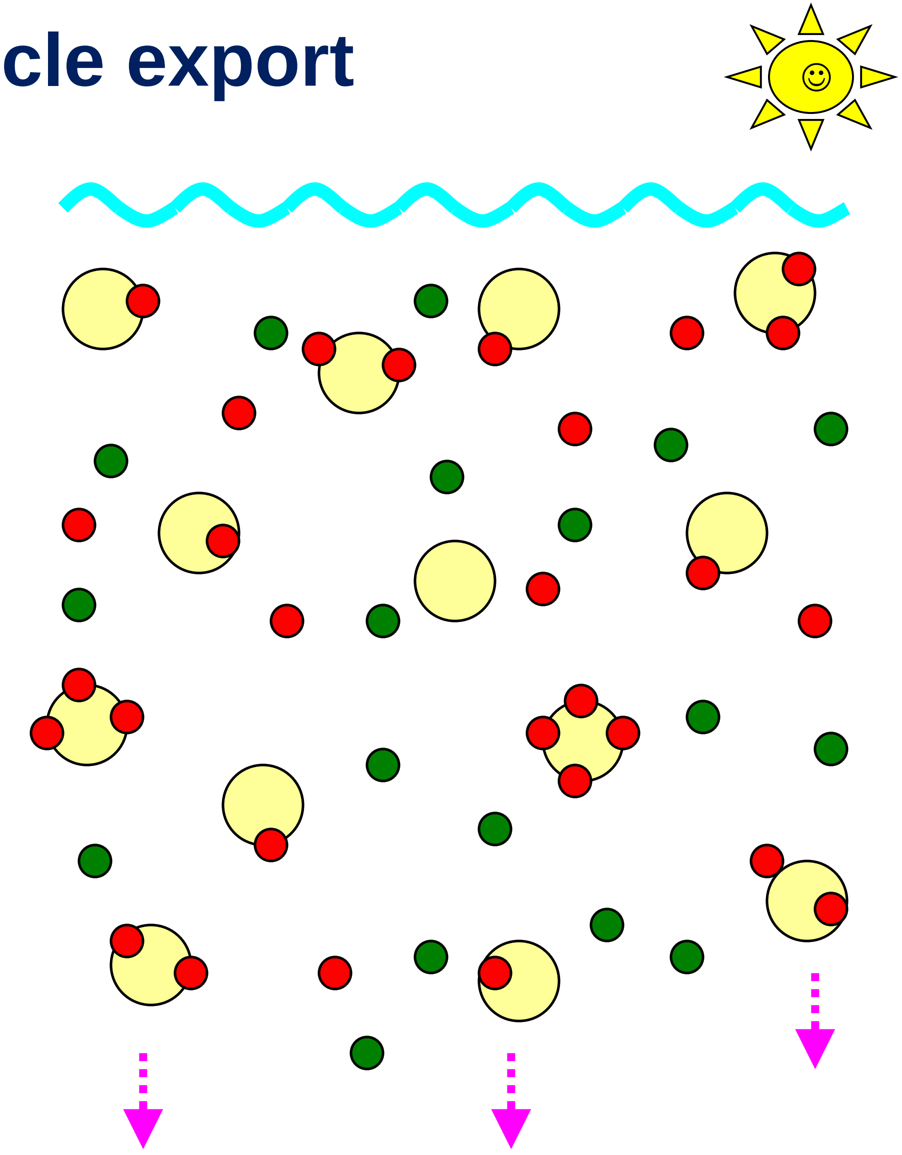
^{234}Th lower near coast due to higher particle flux

Bhat, Krishnaswami, Lal, Rama & Moore, 1969

How does it work?

^{234}Th as a tracer for particle export

^{238}U $\xrightarrow{\alpha}$ ^{234}Th ($t_{1/2}$ = 24.1 d)



^{238}U is conservative in seawater.

^{234}Th is highly particle reactive

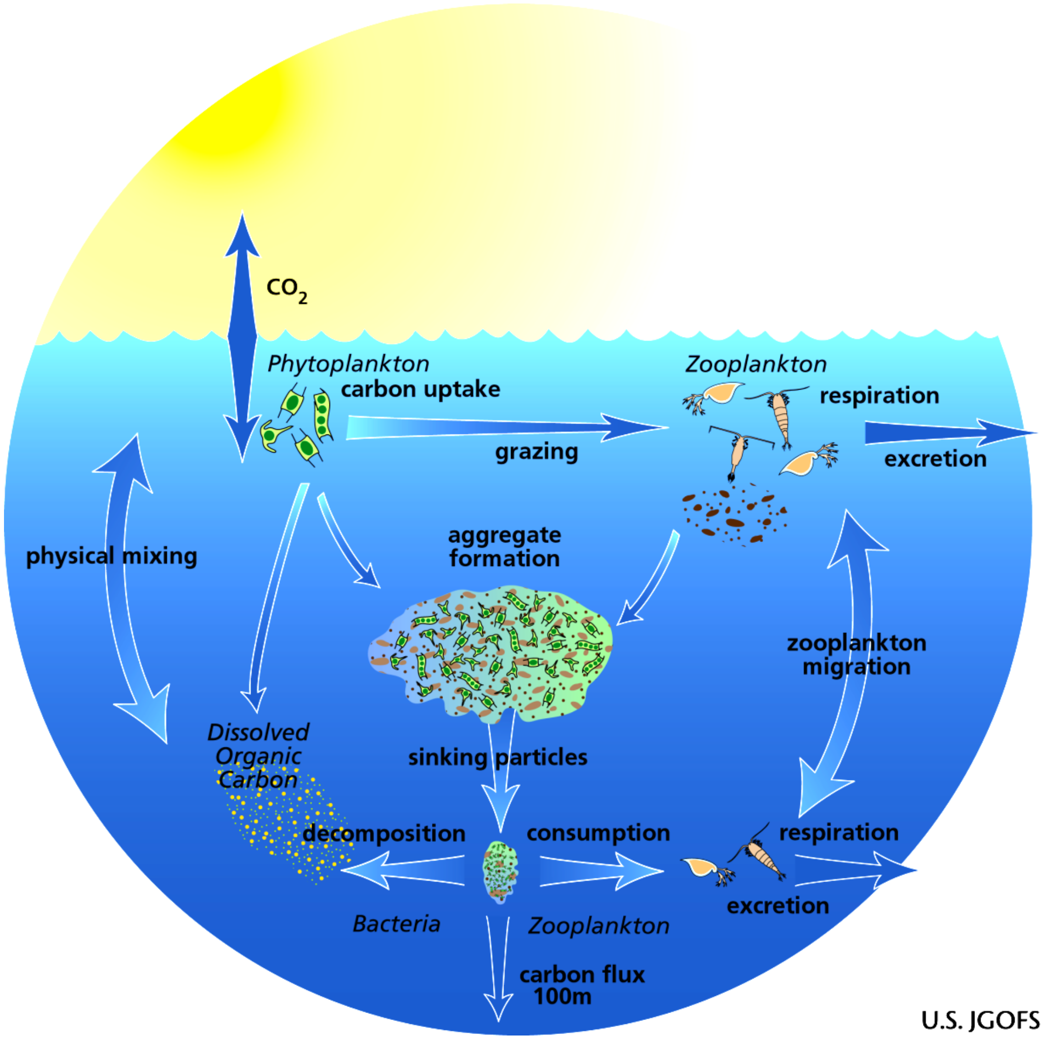
Expect secular equilibrium in an ocean without particles

Particulate matter *^{238}U* *^{234}Th*

Biological Pump

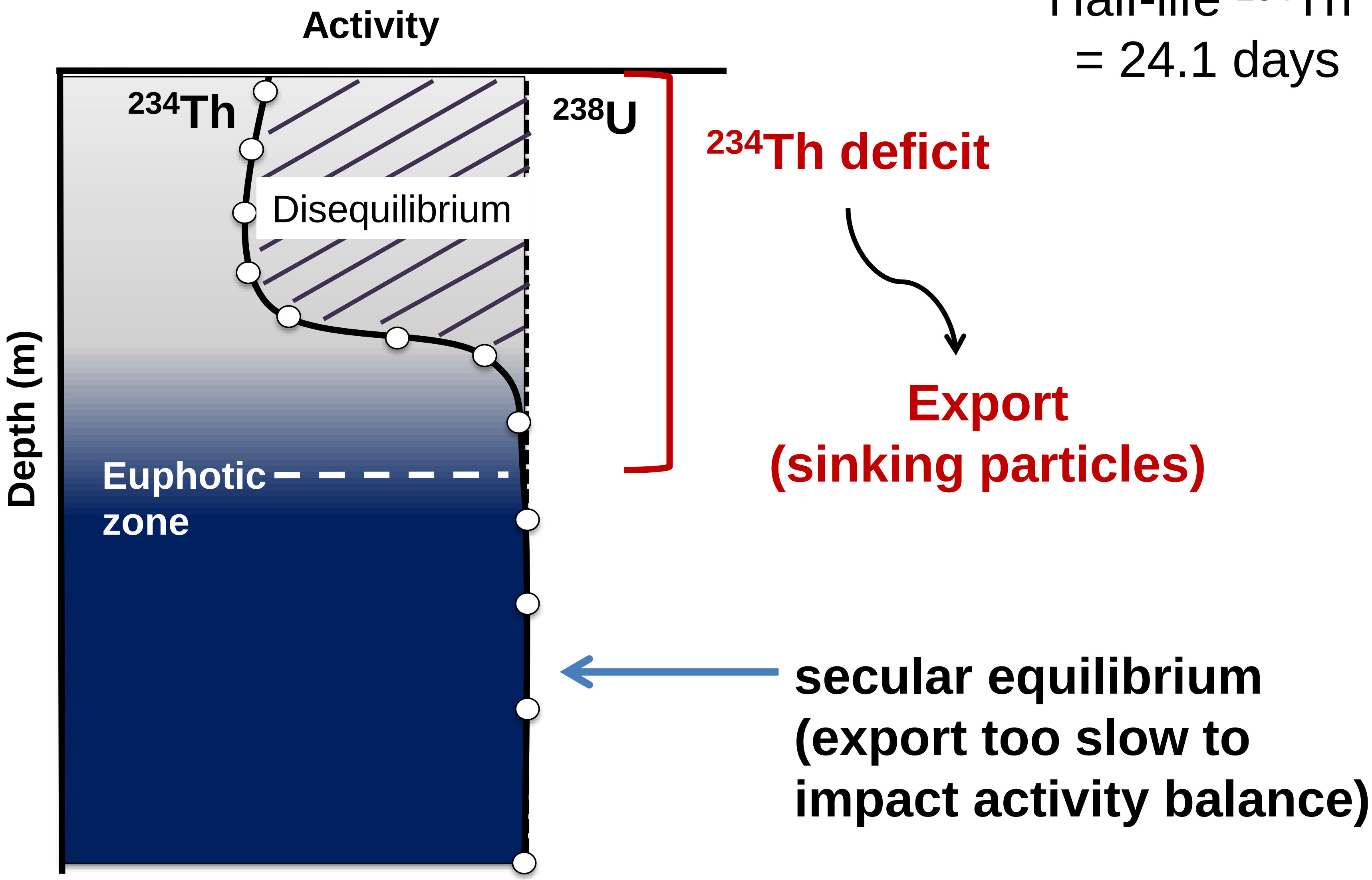
Combined processes that create sinking particles in the ocean

- scavenging tracers follow these routes



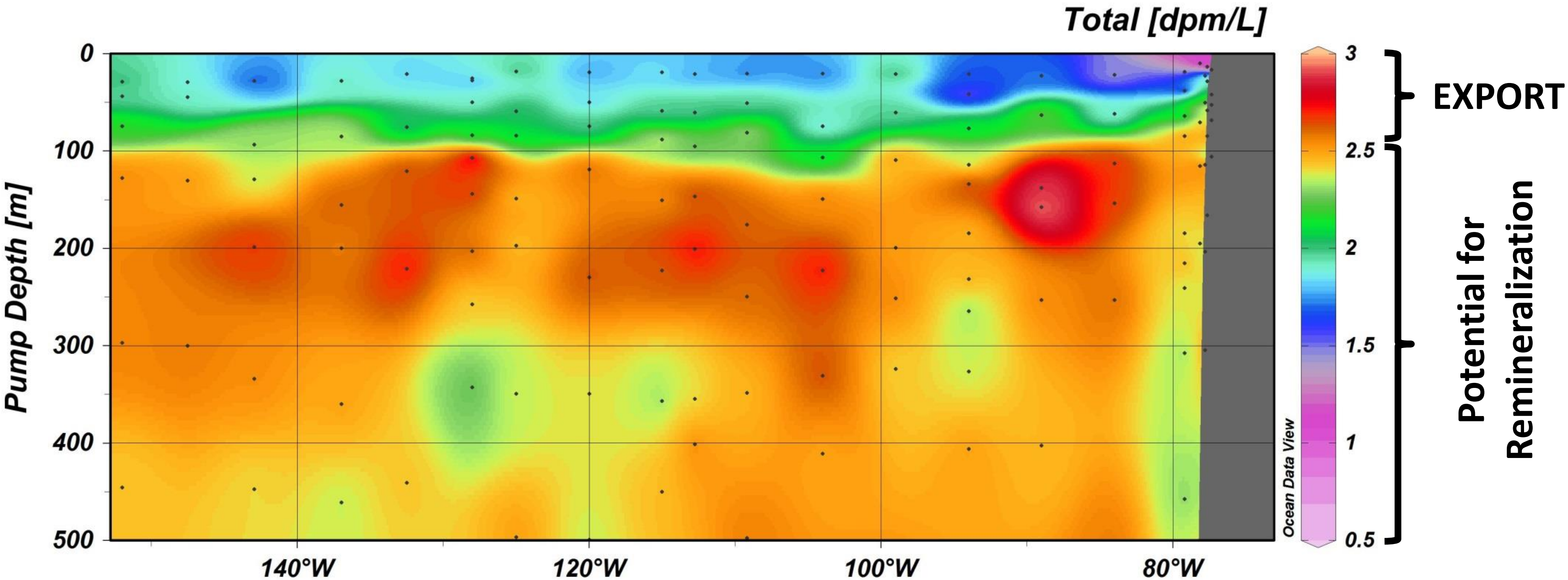
Expected ocean ^{234}Th distributions

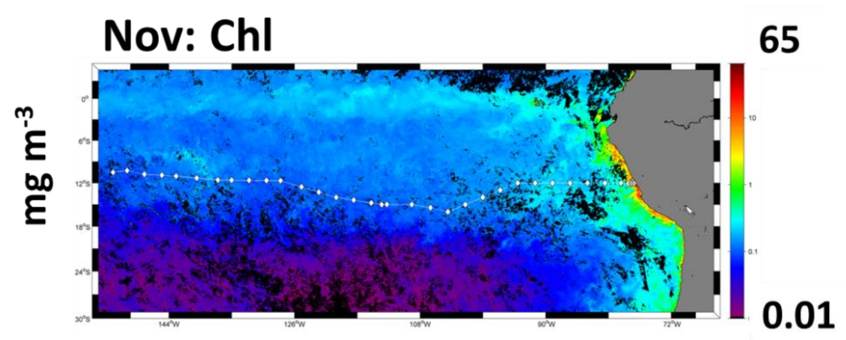
Half-life ^{234}Th = 24.1 days



Example-

Thorium-234 distribution top 500m off west coast South America

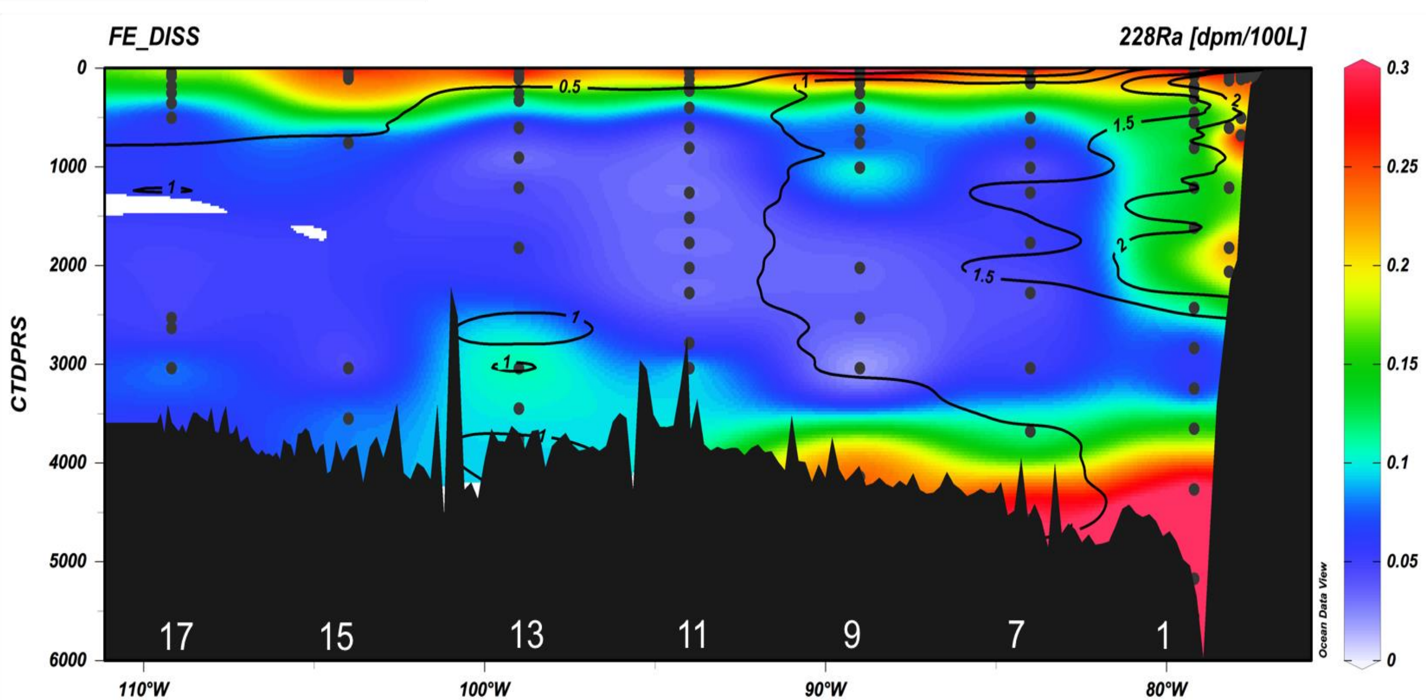


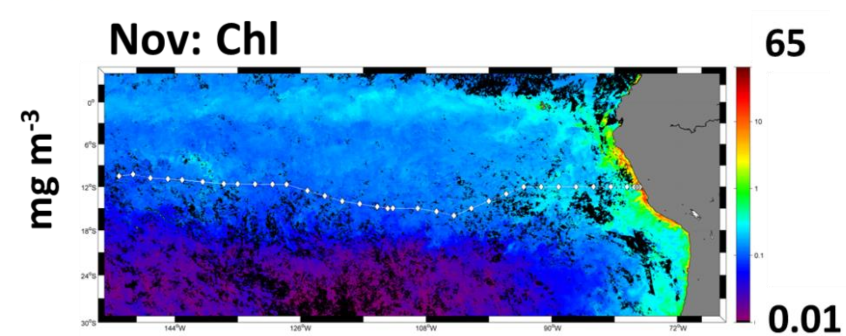


GEOTRACES program
Intermediate data product
Erin Black PhD thesis
in progress, WHOI

Example of conservative tracer- ^{228}Ra
half-life = 5.75 years

horizontal transport from margin source





GEOTRACES program
Intermediate data product
Lauren Kipp PhD thesis
V. Sanial, post doc
In progress, WHOI

Cosmogenic Radionuclides- many produced

Table 1. *Isotopes ($\tau_{\frac{1}{2}}$ >1 day) produced by cosmic rays in the atmosphere.*

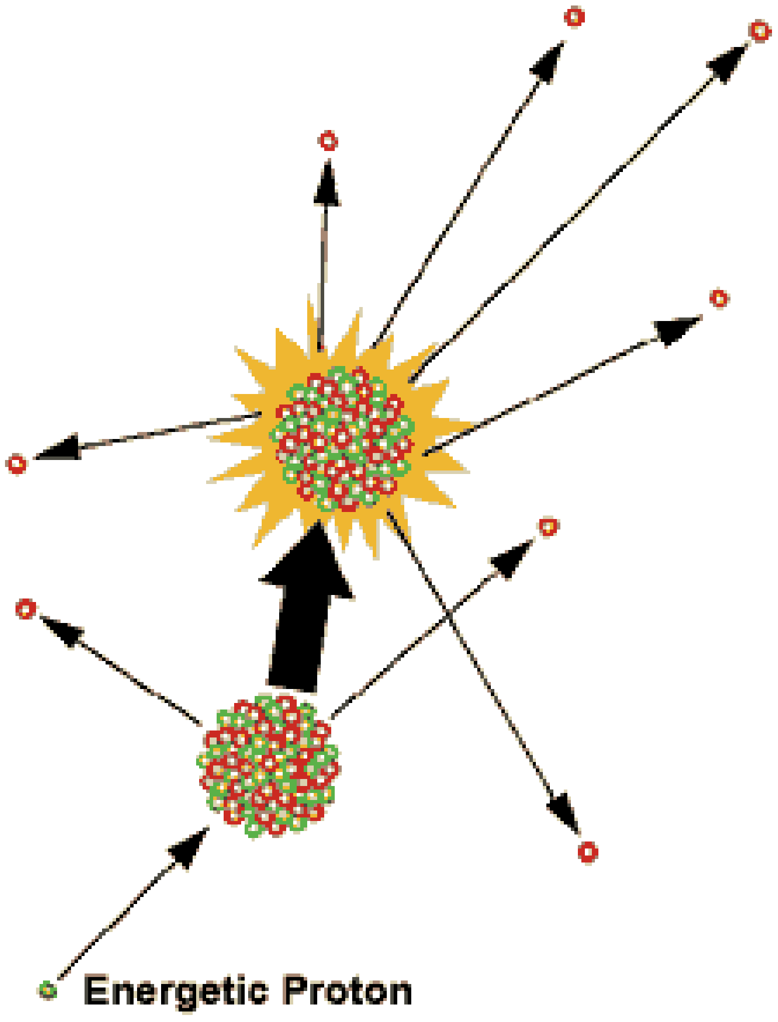
Isotope	Half-life	Main radiation	Main target nu-clide(s)	Reference
He^3	Stable	—	N, O	[*F 1*]
Be^{10}	2.5×10^6 y	β^-—550 KeV	N, O	[*P 1*]
Al^{26}	7.4×10^5 y	β^+—1.17 MeV	Ar	Not detected
Cl^{36}	3.1×10^5 y	β^-—714 KeV	Ar	[*D 2*]
Kr^{81}	2.1×10^5 y	K—X ray	Kr	[*H 3*]
C^{14}	5730 y	β^-—156 KeV	N, O	[*L 16*]
Si^{32}	500 y*	β^-—100 KeV	Ar	[*L 14*]
Ar^{39}	270 y	β^-—565 KeV	Ar	Not detected
H^3	12.3 y	β^-—18 KeV	N, O	[*F 1*]
Na^{22}	2.6 y	β^+—540 KeV γ—1.3 MeV	Ar	[*M 9*]
S^{35}	87 d	β^-—167 KeV	Ar	[*G 5*]
Be^7	53 d	γ—480 KeV	N, O	[*A 6*]
Ar^{37}	35 d	K—X ray	Ar	Not detected
P^{33}	25 d	β^-—250 KeV	Ar	[*L 12*]
P^{32}	14.3 d	β^-—1.7 MeV	Ar	[*M 8*]

* Based on cross section measurements [*H 2*].

Lal and Peters, 1967

Cosmogenic Radionuclides - Sources

Spallation: A high energy nuclear reaction resulting in the emission of two or More fragments from the nucleus



Cosmogenic Radionuclides - Sources

Spallation: how do we know?

Photographic Emulsion of a spallation reaction involving silver

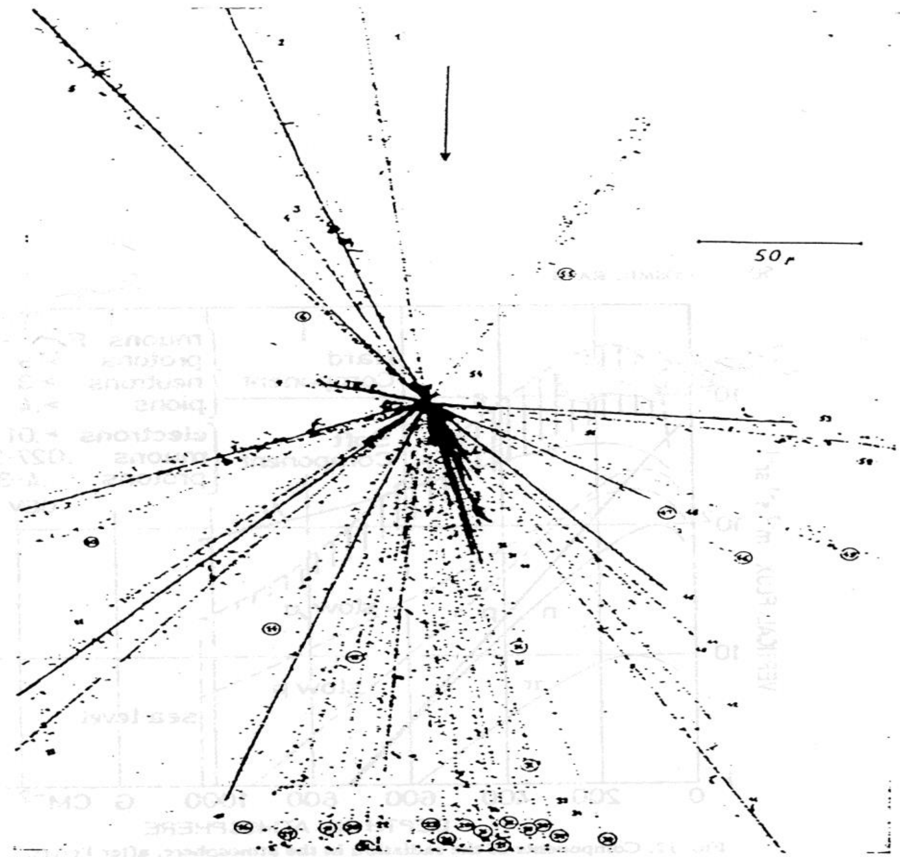
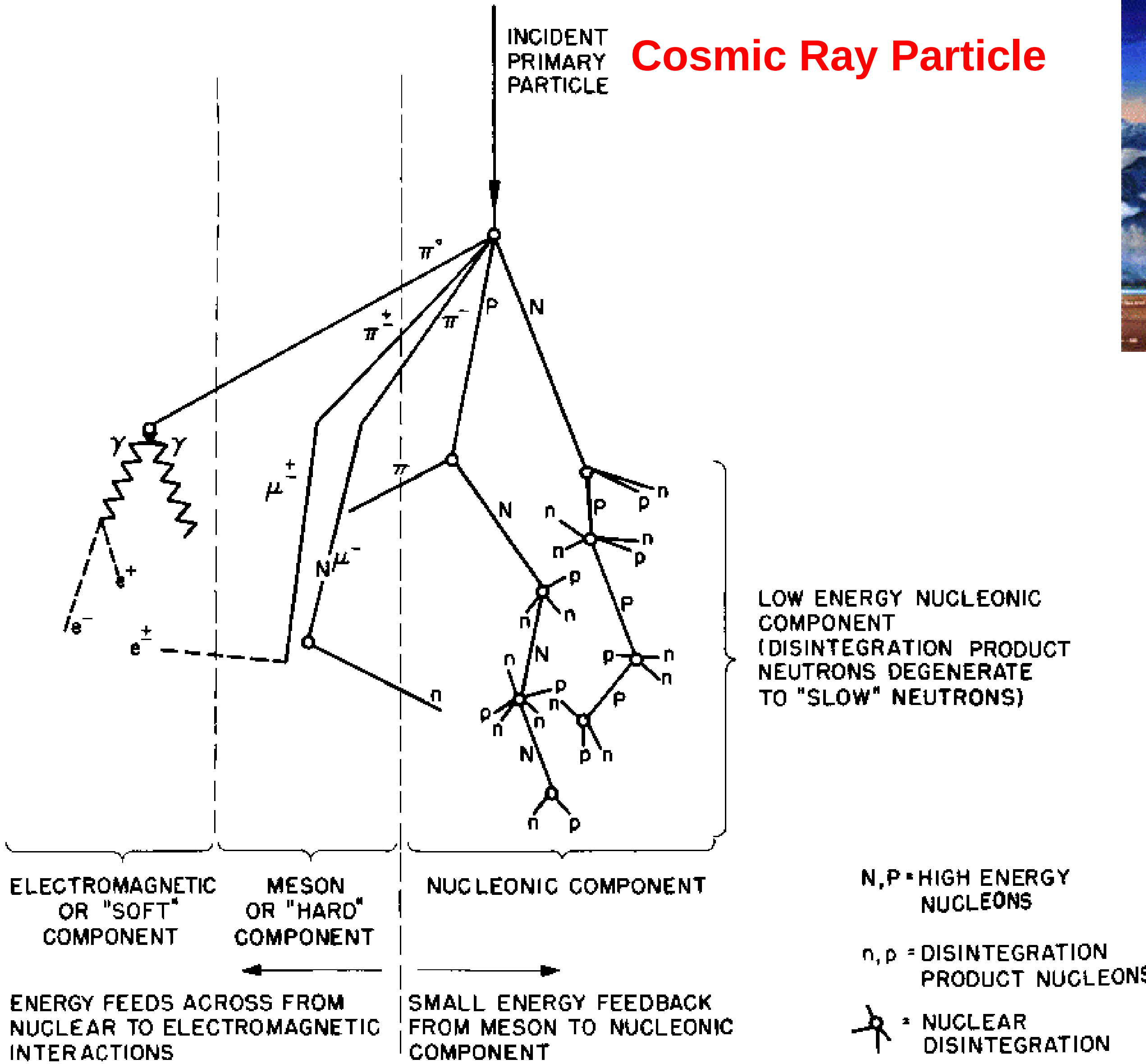


Fig. 8.1.11. Photomicrograph of a nuclear event observed by Leprince-Ringuet, Bousser, Hoang, Jauneau, and Morellet (LRL49.2; LRL49.3) in an Ilford G5 electron-sensitive emulsion flown at about 15 g cm^{-2}. The star appears to be produced by a neutral primary particle. It contains 27 secondary particles of greater than minimum ionization, which can be identified as protons, α-particles, and heavier nuclear fragments. It also contains 28 secondary particles of minimum ionization, emitted within a fairly narrow cone. Some of these particles must be created during the interaction because the total charge carried by the star particles is greater than the total charge of the heaviest nucleus present in the emulsion. According to the authors, some of the secondary particles of minimum ionization are mesons, some are protons.

402

“Cosmic Ray Shower”

Cosmic Ray Particle



Schematic Diagram of Cosmic Ray Shower

http://www.physics.purdue.edu/primelab/rosetest/plresearch.php

As such…. cosmogenic nuclide production varies with height in the atmosphere!

Most cosmic rays are "galactic" and originate from supernovas, but ***our sun*** is also a significant source.

89% are hydrogen
10% helium, and
1% heavier elements.

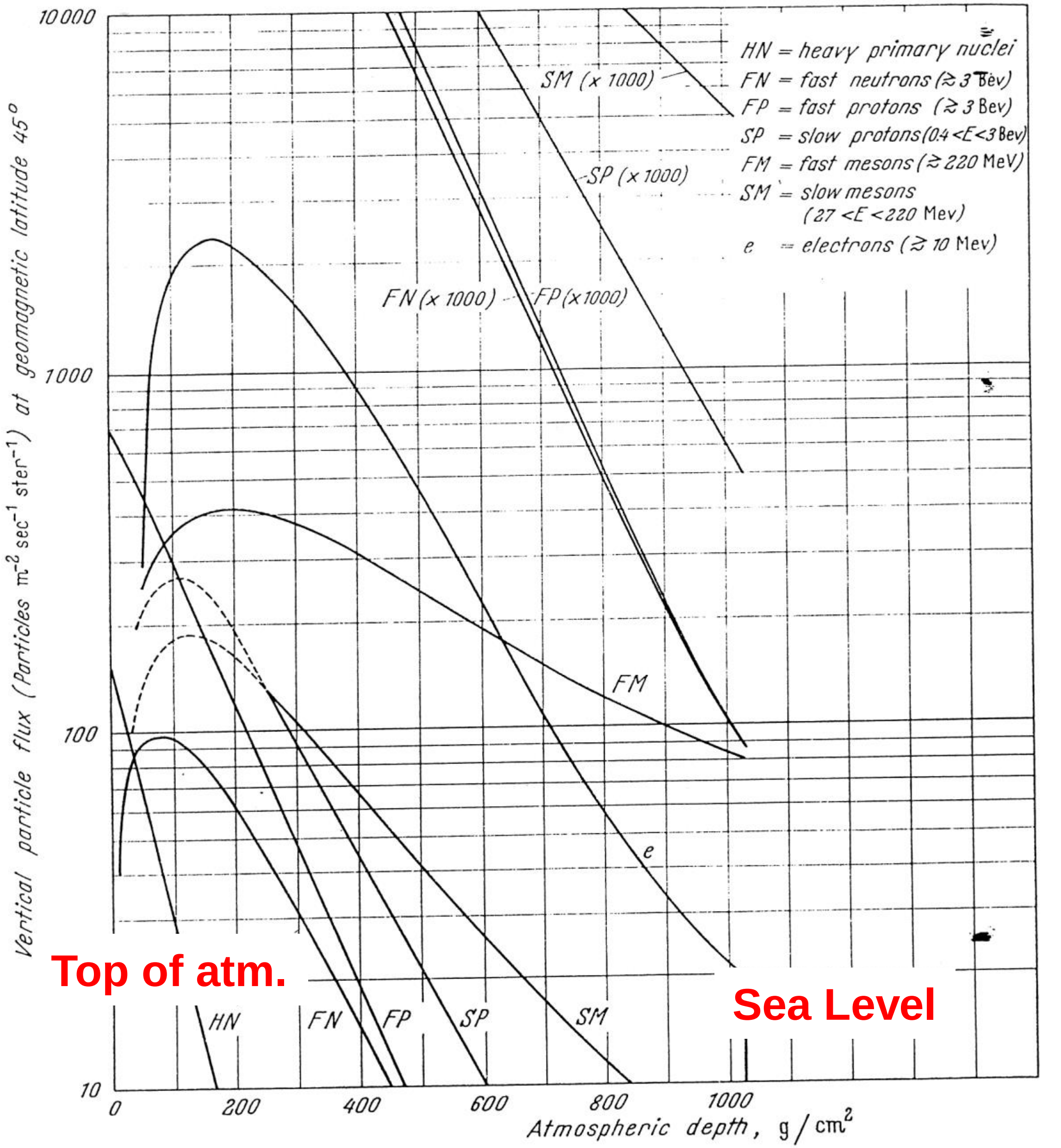
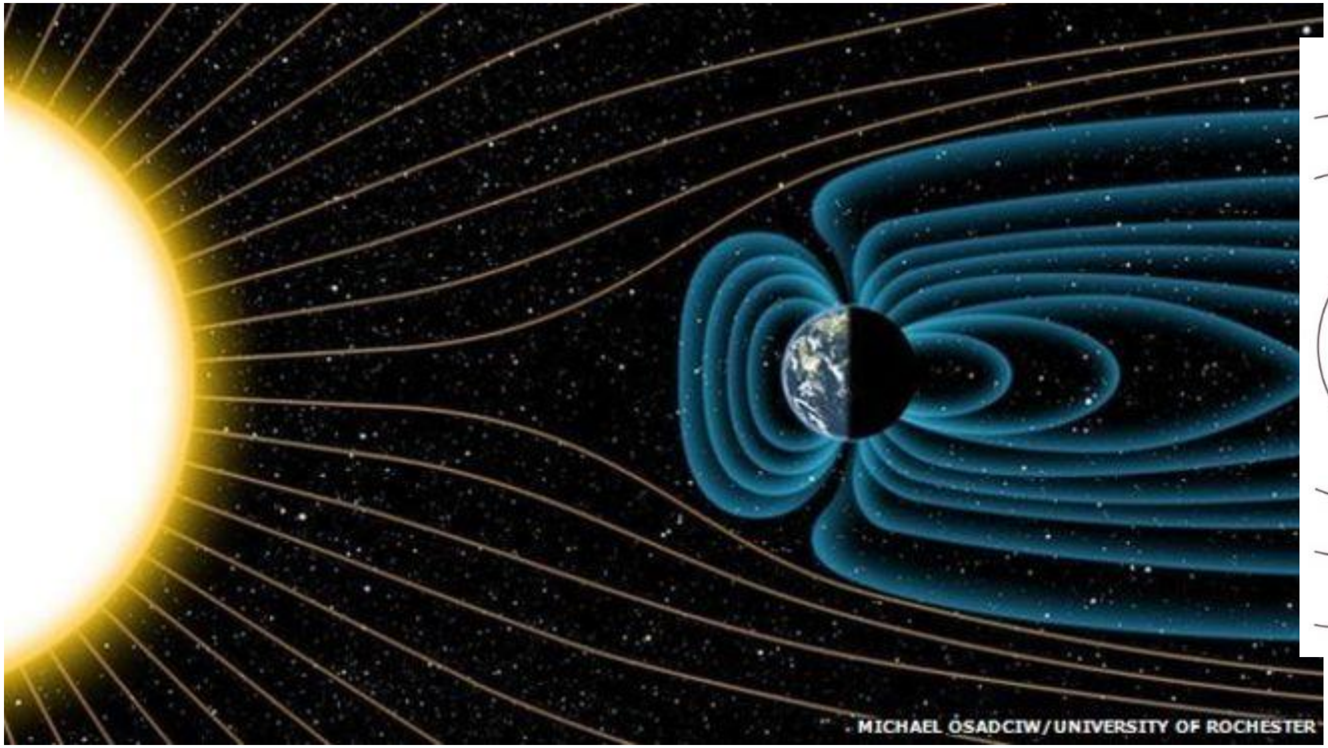


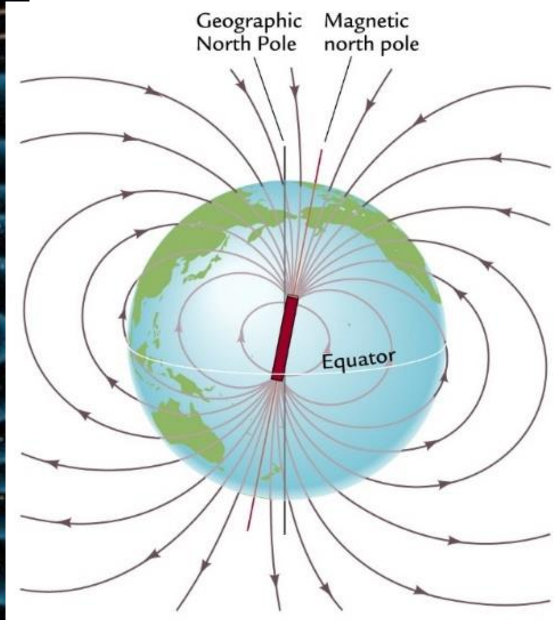
Fig. 3. Composition of cosmic radiation as a function of atmospheric depth. The flux values refer to geomagnetic latitude 45° and to longitude 80° West. (From PETERS [P3].)

Production may vary with latitude due to:

Solar winds deflected by Earth's magnetic field



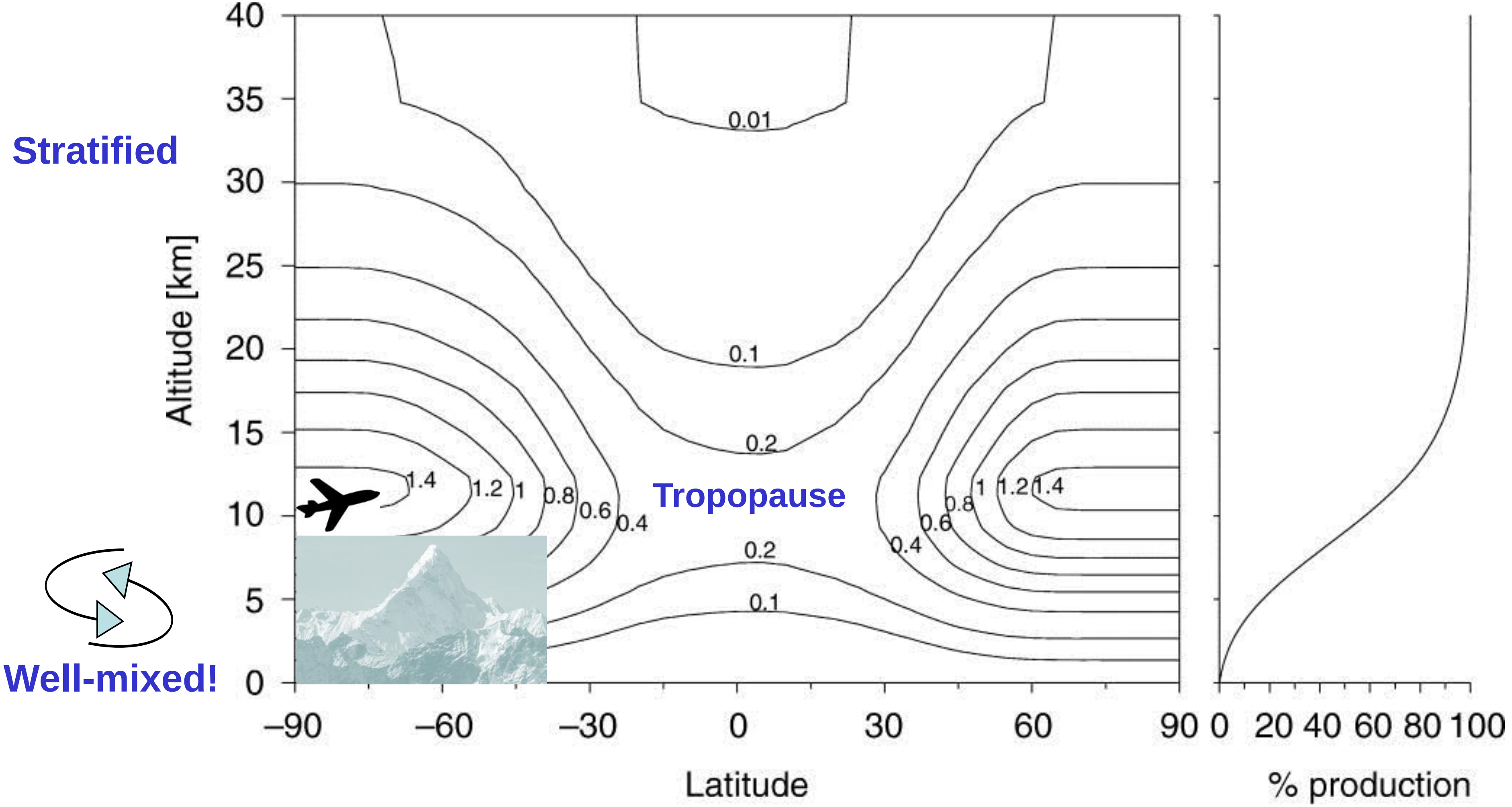
Earth's Magnetic field



https://www.youtube.com/watch?v=6hD52H7rQak

Example- ^{10}Be production in the Atmosphere

atoms/y/cm^3



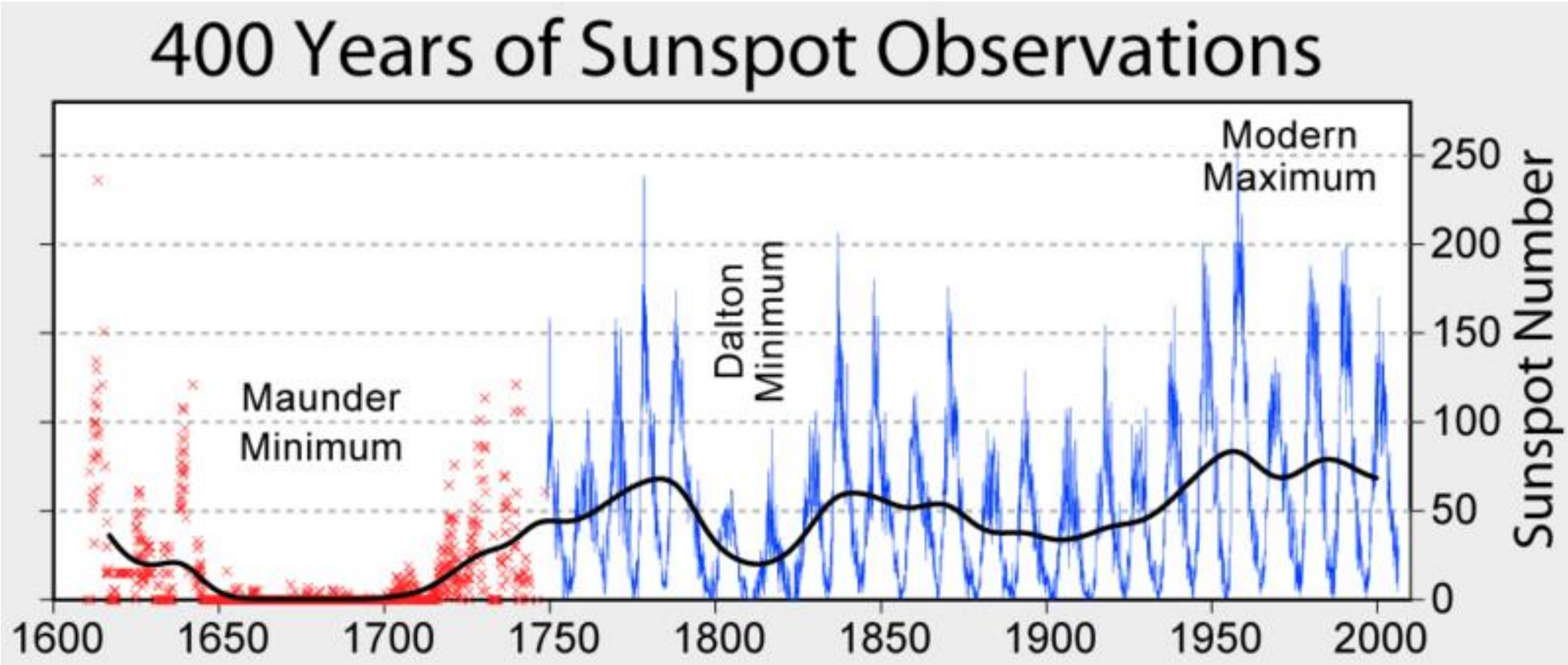
DELAYGUE, Gilles; BEKKI, Slimane; BARD, Edouard. Modelling the stratospheric budget of beryllium isotopes. Tellus B, [S.l.], v. 67, aug. 2015. ISSN 1600-0889.

Variations over short/seasonal time scales

Northern lights in County Donegal, Ireland January 24, 2012
(From a solar flare that erupted on Jan 22nd)

Photo taken by Twitter ser @andrew_chester via lockerz.com

Variations on 11 yr solar cycle and longer time scales due to “solar dynamo”



“Little Ice Age”

Source variability complicates use of 14C

http://oneminuteastronomer.com/1042/sunspots-cycles/

What are Cosmogenic Radionuclides Used for?

Mainly ^{14}C (complicated by “bomb” ^{14}C)
and ^{7}Be

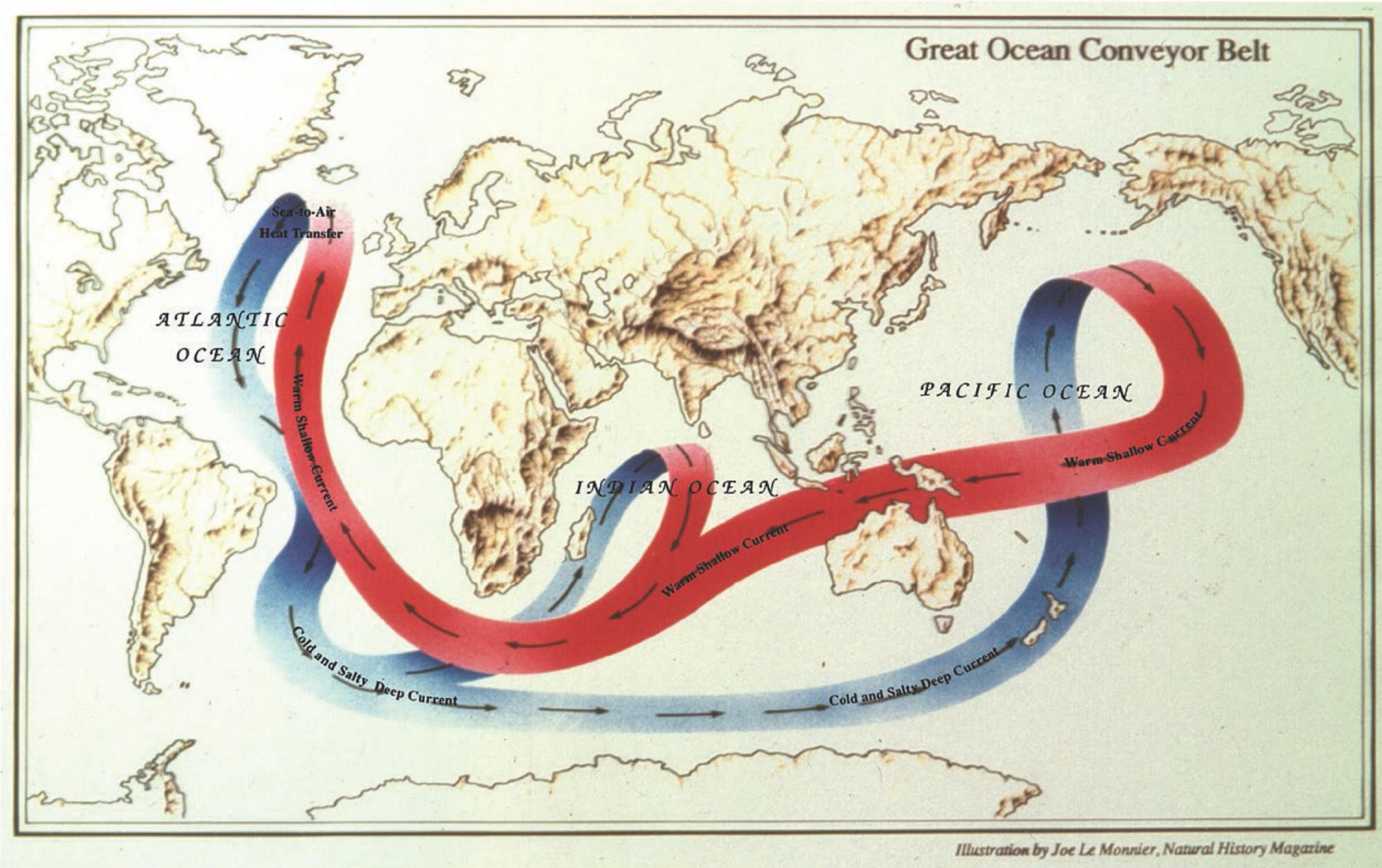
Since they are added to the surface ocean, can be tracers of:

1) Long time scale ocean circulation
   - ^{14}C ($t_{1/2}$ = 5,700 yr)

2) Short time scale upper ocean vertical mixing rates
   - ^{7}Be ($t_{1/2}$ = 53 d)

3) Sediment accumulation & mixing- ^{14}C

4) Ages/dating of organic matter (C cycle)- ^{14}C
   Dissolved organic carbon
   Particulate organic carbon
   Specific organic compounds

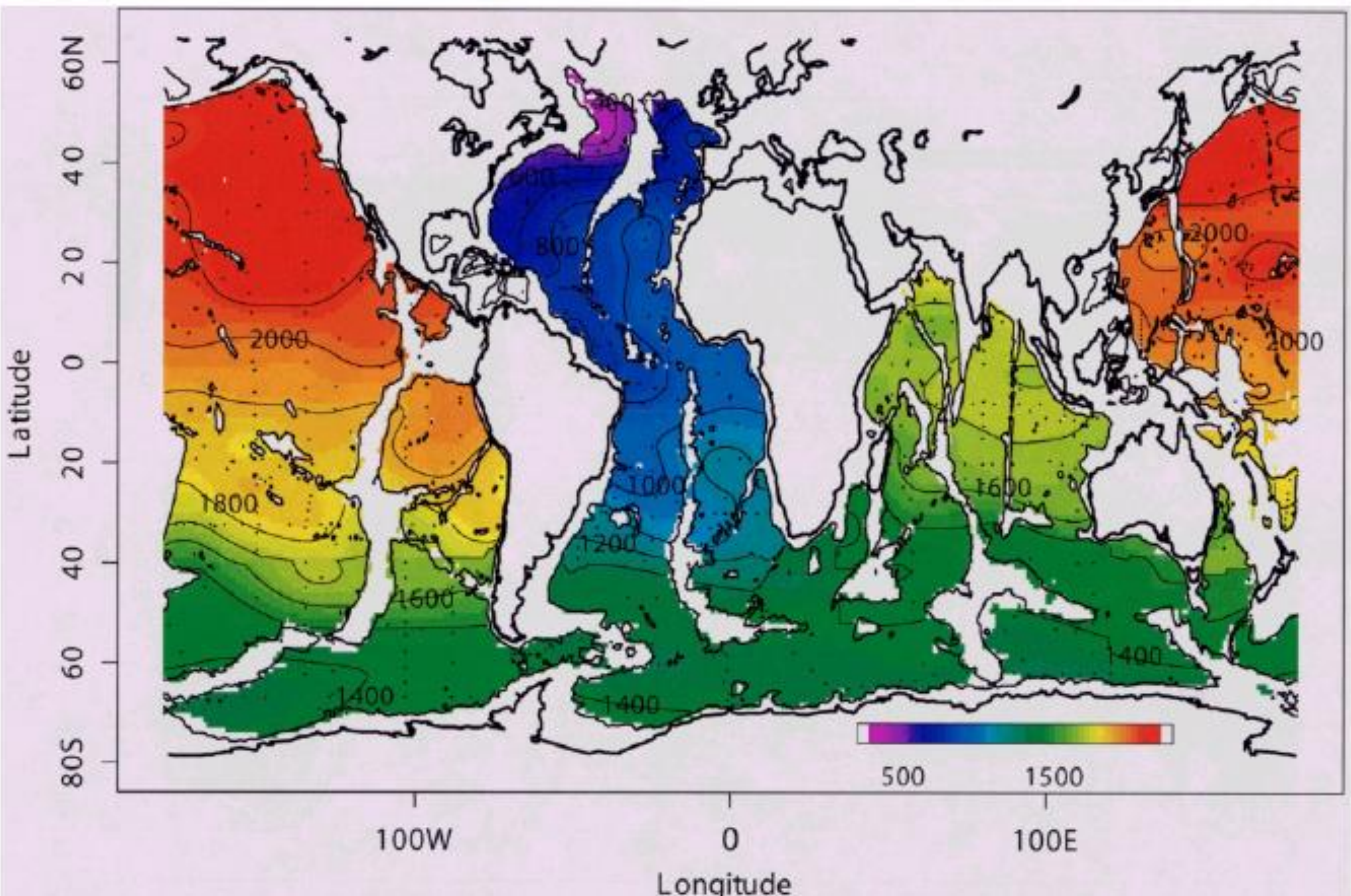


Credit: Joe Le Monnier (Natural History Magazine, 1987).

Figure 19b An idealised diagram depicting the global sweep of the Great Ocean Conveyor Belt. The blue depicts the path followed by the deep water formed in the North Atlantic; and the red, the path followed by the returning upper ocean flow. This diagram was prepared for an article published in the popular *Natural History Magazine*. I never dreamed that it would become a logo for global change.

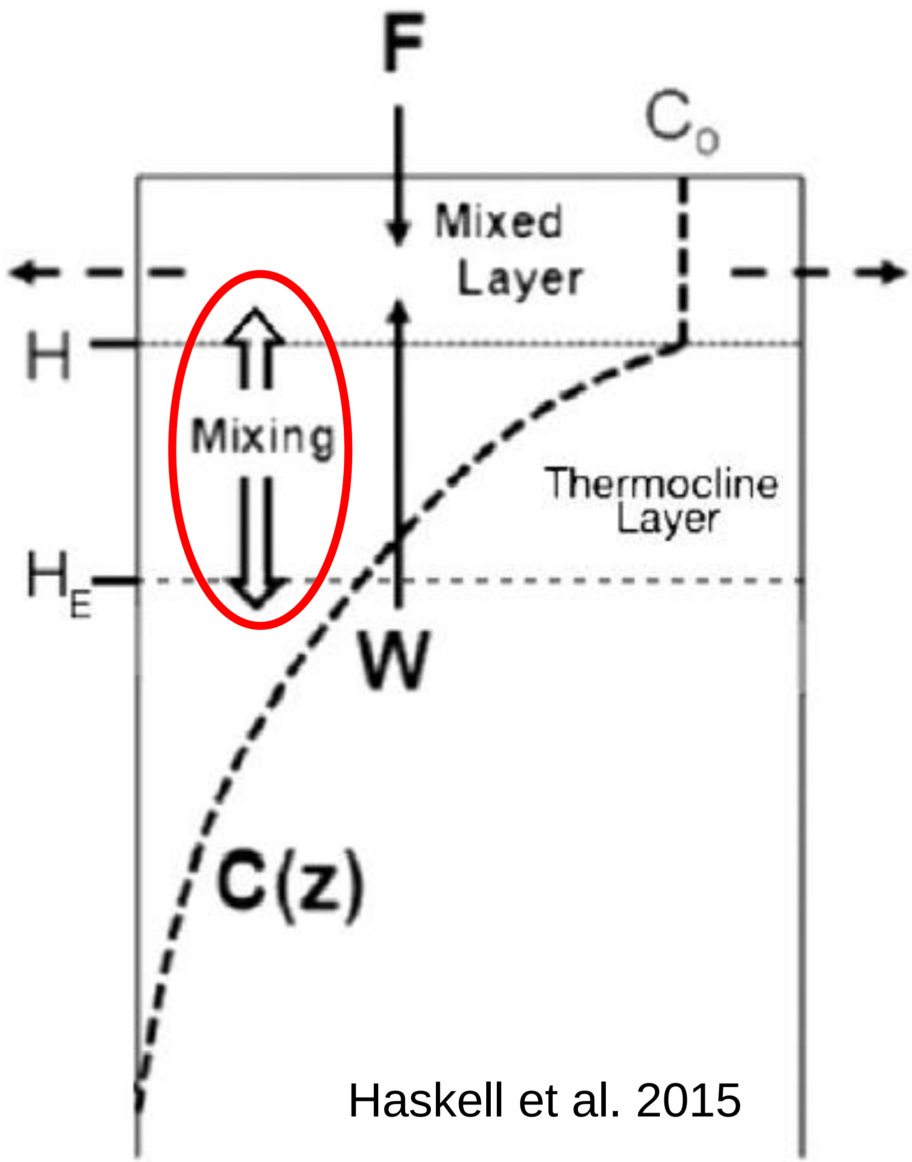
W. Broeker

Example- Age of ocean waters at 3500m determined from ^{14}C



Matsumoto and Key 2004

Example- ^{7}Be for upper ocean mixing conceptual model



Haskell et al. 2015

^{7}Be half-life = 53 d

C = change in ^{7}Be concentration along a mixing line (dashed)

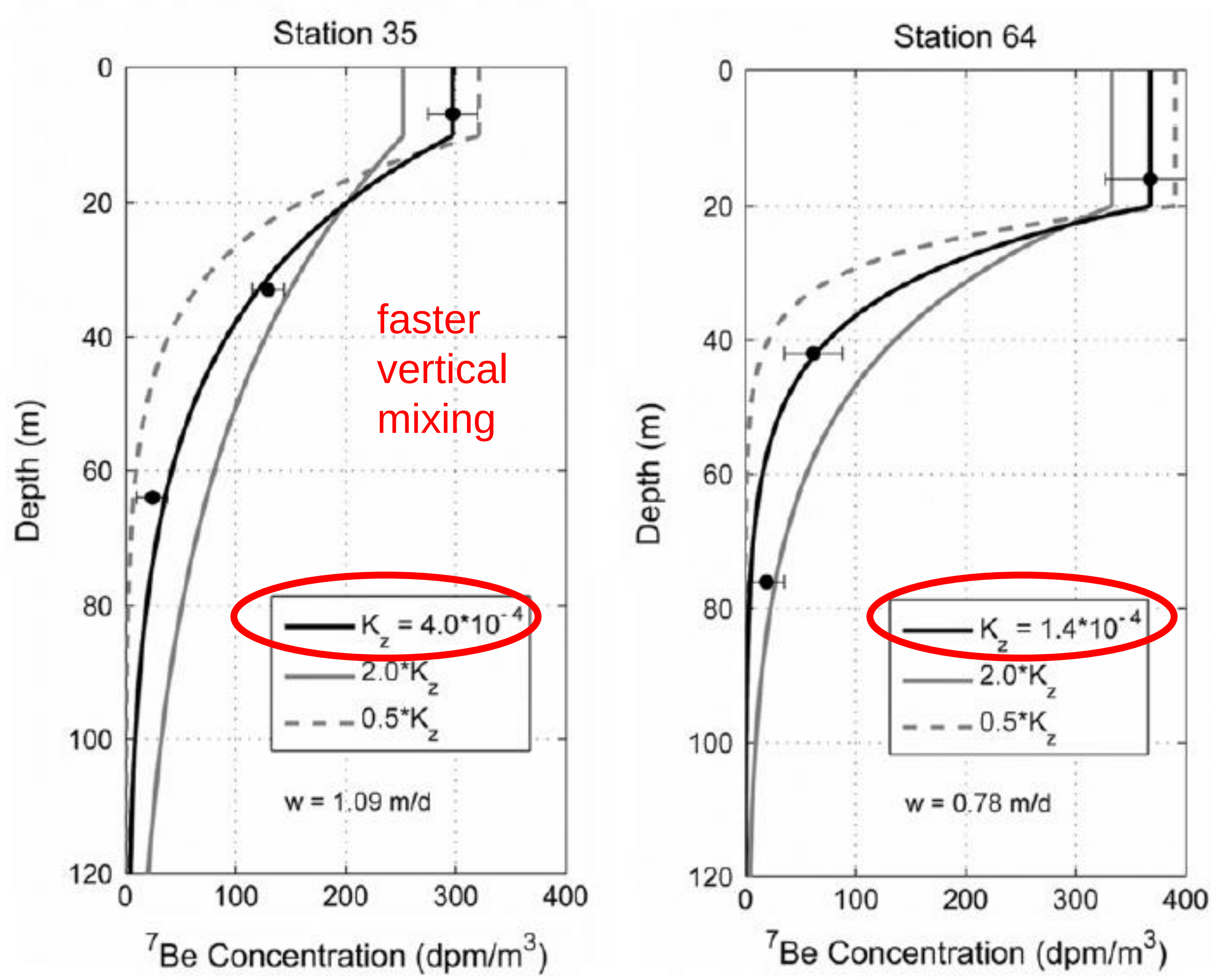
F = Atmospheric ^{7}Be input

H = mixed layer depth
H_E = depth of euphotic zone

W = upwelling velocity

Dashed arrows = horizontal flow

Vertical mixing (K_z) is derived from change in ^{7}Be concentration



Kadko and Johns, 2011

Artificial Radionuclides

Bikini island 1946

Artificial Radionuclides are those radioactive nuclides that do not naturally occur on Earth, i.e. “man-made”

Different sources:

1) nuclear reactor operations

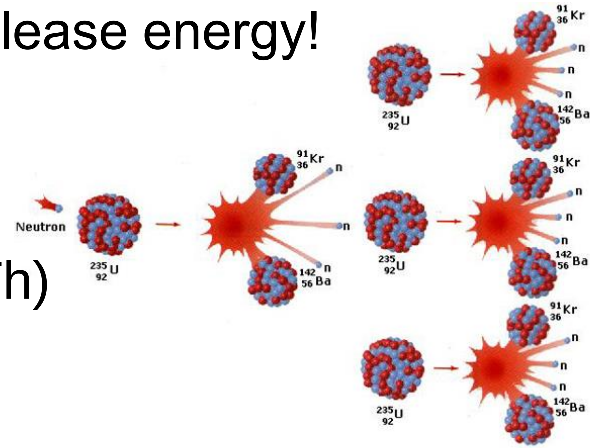
2) nuclear weapons

3) particle accelerators

Why make artificial radionuclides?

Nuclear power

It's all about using a fission chain reaction to release energy!

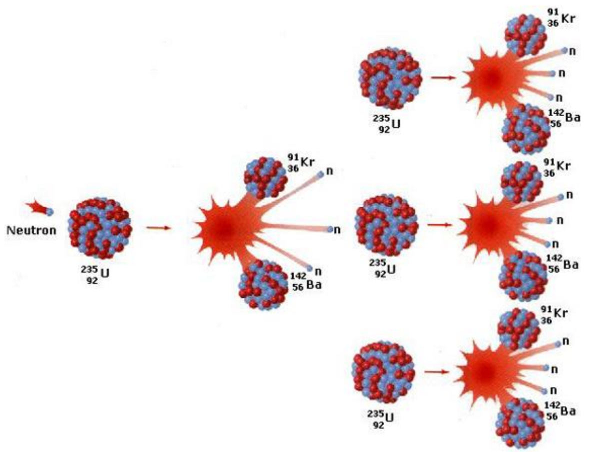


The main fissionable radionuclides are:

1) ^{233}U (produced by neutron irradiation of ^{232}Th)
2) **^{235}U (naturally occurring ~0.72% of U)**
3) ^{239}Pu (produced by neutron irradiation ^{238}U)

Most nuclear reactors use Low Enriched Uranium (LEU) that is about 3-5% uranium-235 for **controlled nuclear fission** to generate heat

Why make artificial radionuclides?



& Nuclear weapons

"uncontrolled" nuclear fission

High Enriched Uranium (HEU) is enriched to more than 20% uranium-235 and is weapons-usable, but the lower the enrichment level the greater the amount of material required to achieve a critical mass— i.e. mass to build a bomb.

Weapons-grade HEU, is typically defined as 90% HEU or above, to minimize weapons' size. Smaller and lighter nuclear weapons are much easier to deliver

Can also use Pu for nuclear weapons- fission bombs

Why make artificial radionuclides? continued……

Medical

Treatments- cancer

& tracers- blood tracking

Industrial applications

Sensors (thickness), detectors, sterilization

Consumer products

Smoke detectors

Ceramic glazes

Major sources of artificial radionuclides to the environment

1) Nuclear weapons - atmospheric testing,

1950's-1960's most above ground tests in US and USSR

≈ 200,000 PBq total; ≈ 1,000 PBq ^{137}Cs

2) Planned releases from nuclear fuel reprocessing cycle

Windscale/Sellafield (UK); Cap de la Hague (France)

≈45 PBq total; ≈ 40 PBq ^{137}Cs

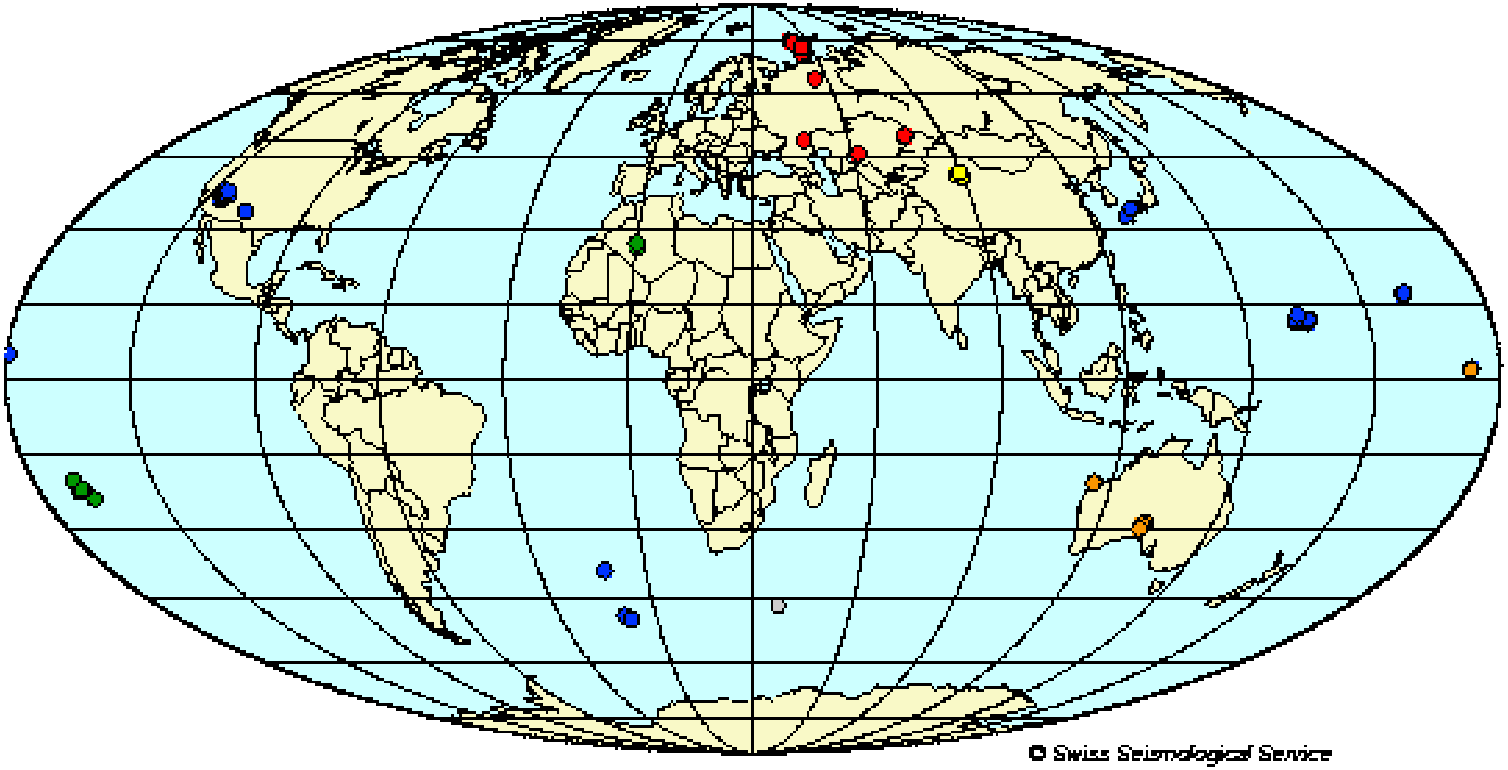
3) Accidents related to nuclear power/production

Chernobyl (April 26, 1986) ≈ 5000 PBq total; ≈ 100 PBq ^{137}Cs

Three Mile Island ≈ 0.00004 PBq ^{137}Cs

Fukushima (March 11, 2011) ≈ 500 PBq total; ≈ 20-40 PBq ^{137}Cs

Since 1945 there have been 517 known atmospheric nuclear weapon tests conducted worldwide.

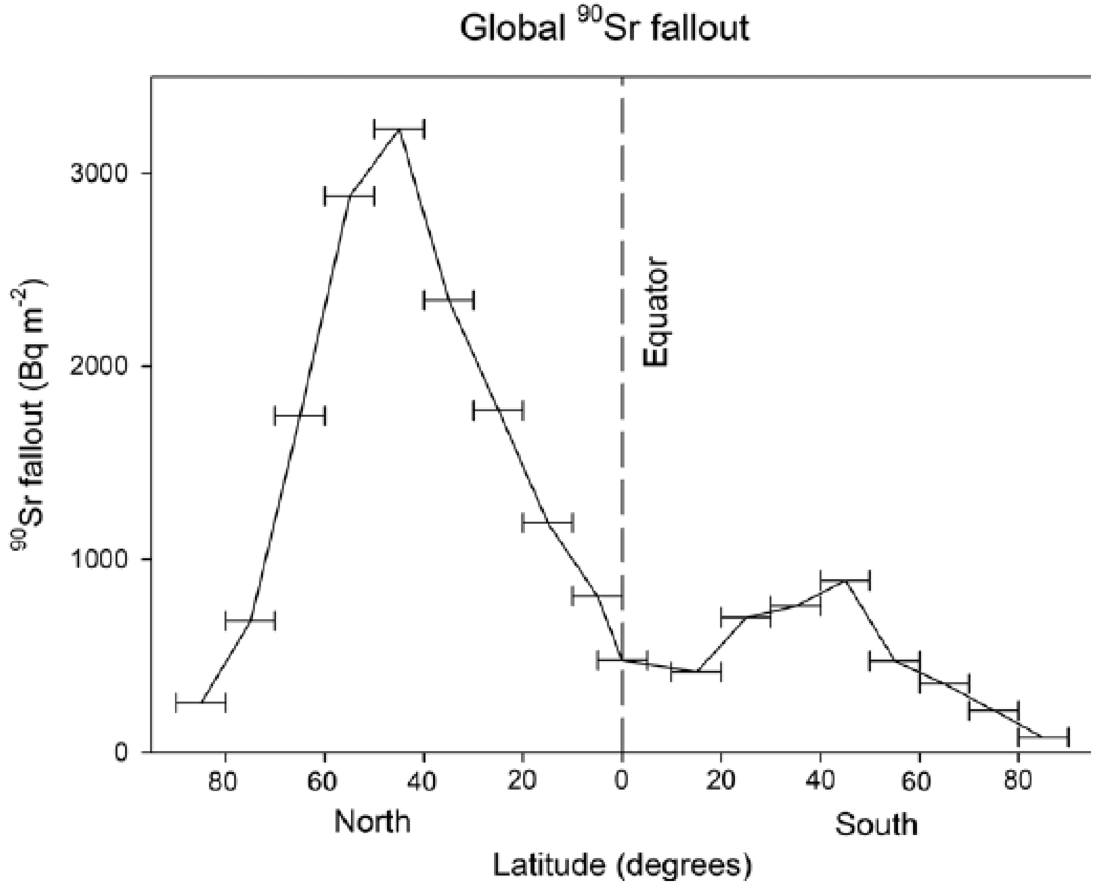


USA (216 Events)
Soviet Union (213 Events)
France (45 Events)
China (21 Events)
Great Britain (21 Events)

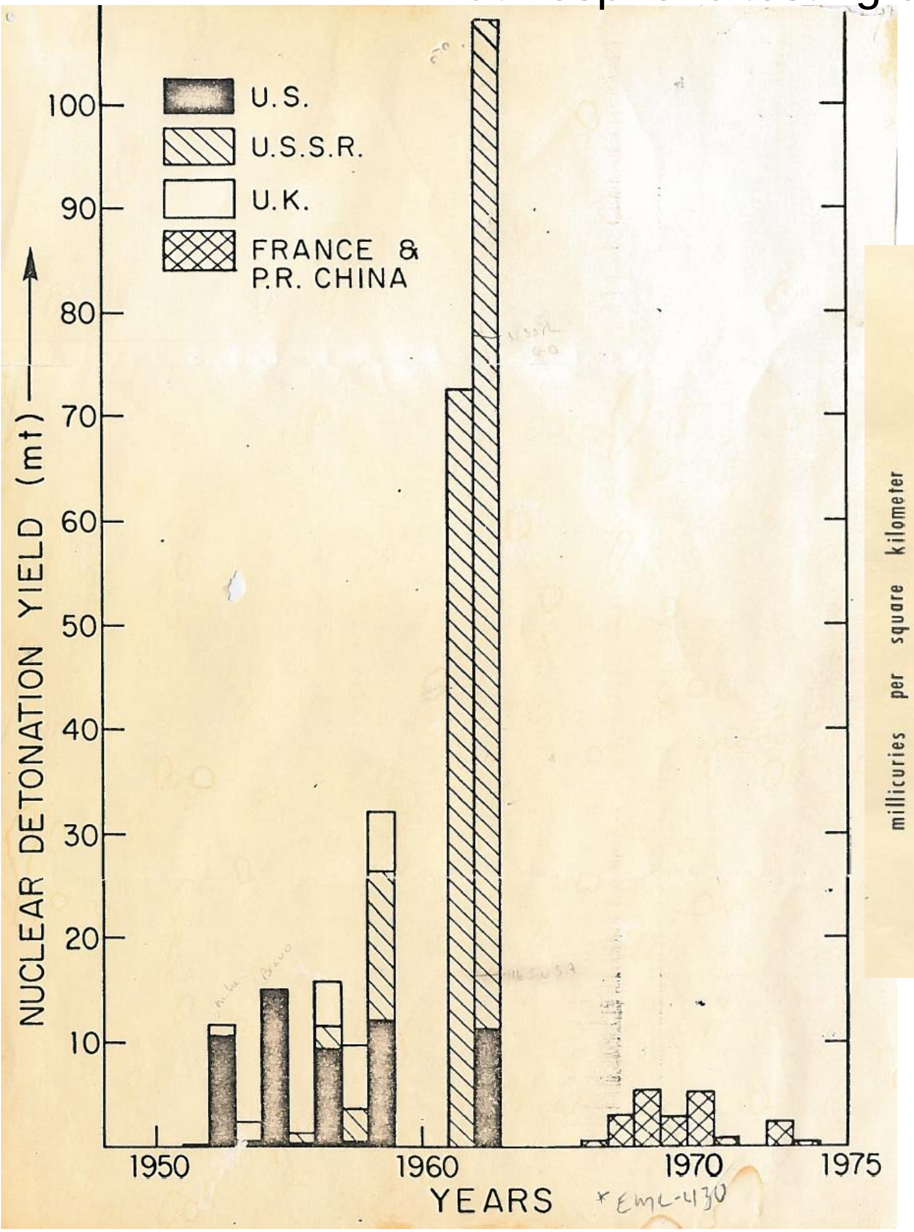
http://www.seismo.ethz.ch/research/groups/veri/projects/nu_atmospheric
https://www.youtube.com/watch?v=310-GYiitpM

Where?
Atmospheric nuclear weapons testing leads to global fallout

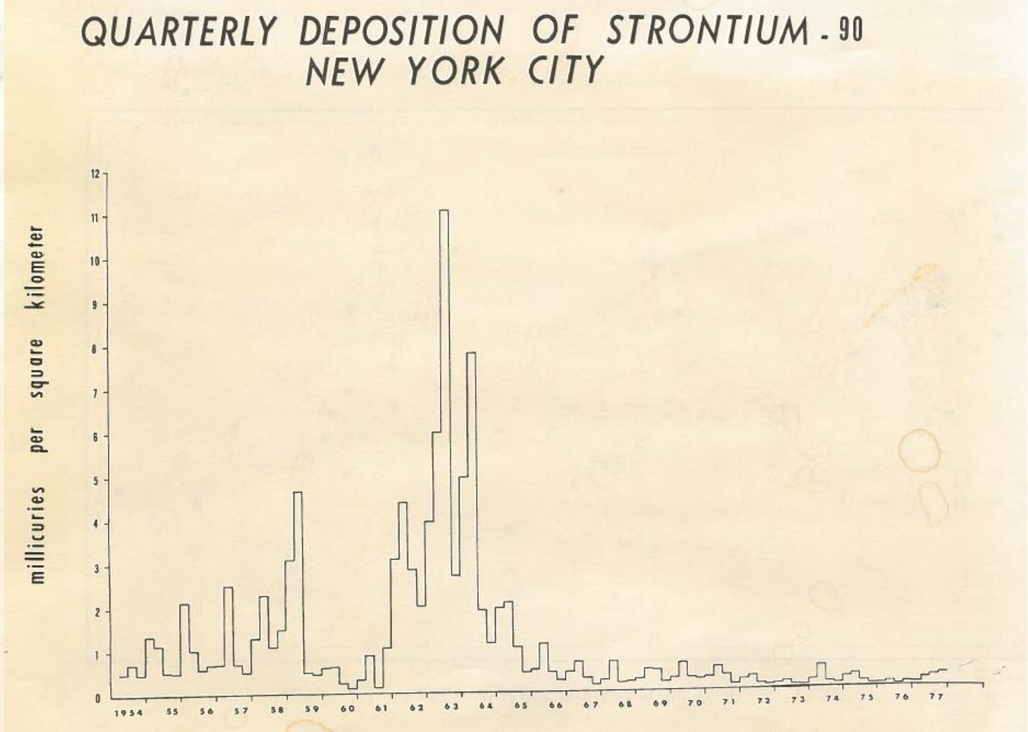
12% of the fallout deposited as “close-in” fallout near the test site
10% percent has ended up in a band around the same latitude as the test site.
78% percent is “global” fallout, most of which has ended up in the same hemisphere



When? testing in/after WWII and Cold War era
testing ban around 1960 followed by peak in 1962
atmospheric testing ceased entirely in 1980.



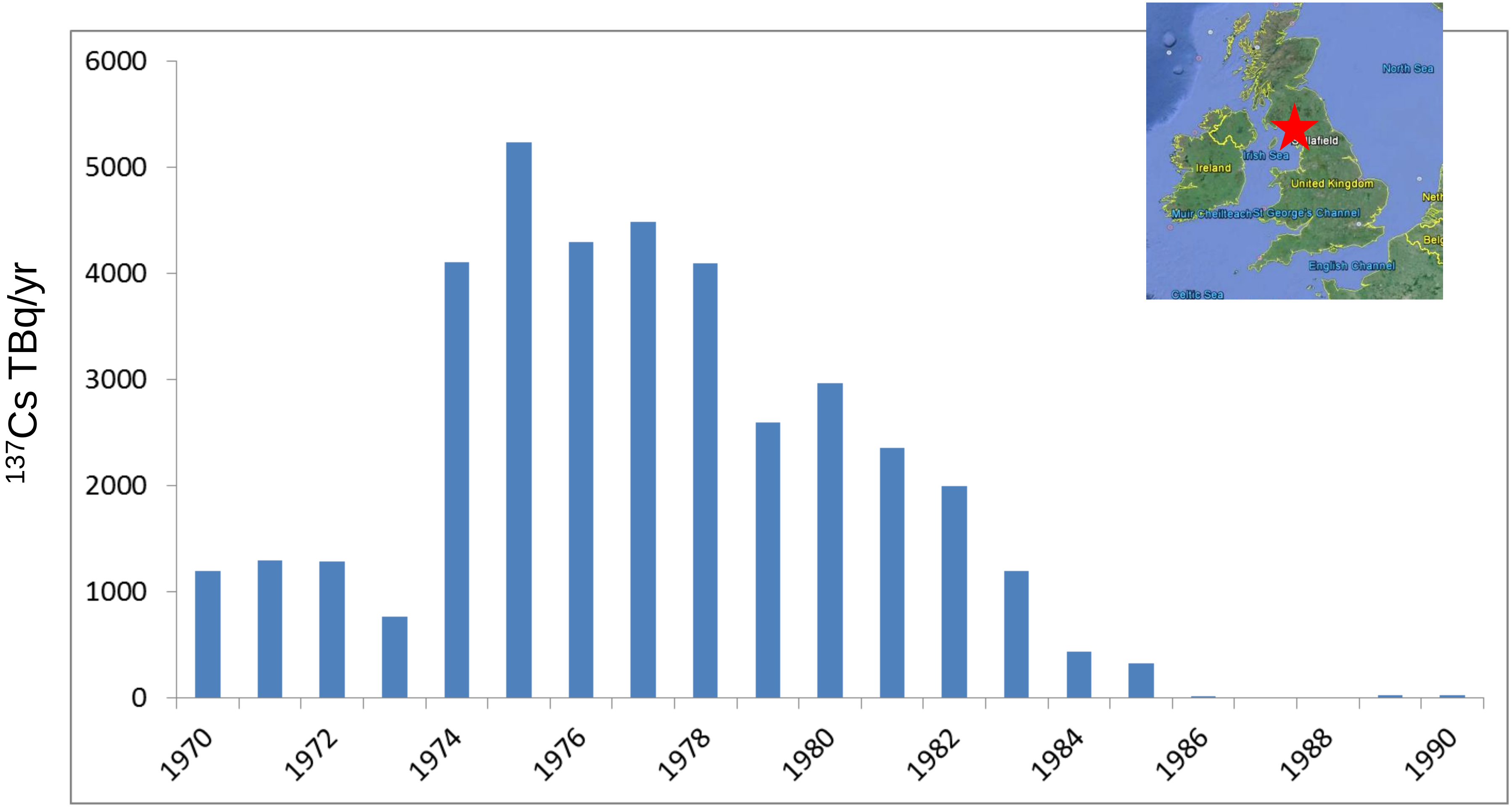
Global monitoring networks established



1954-

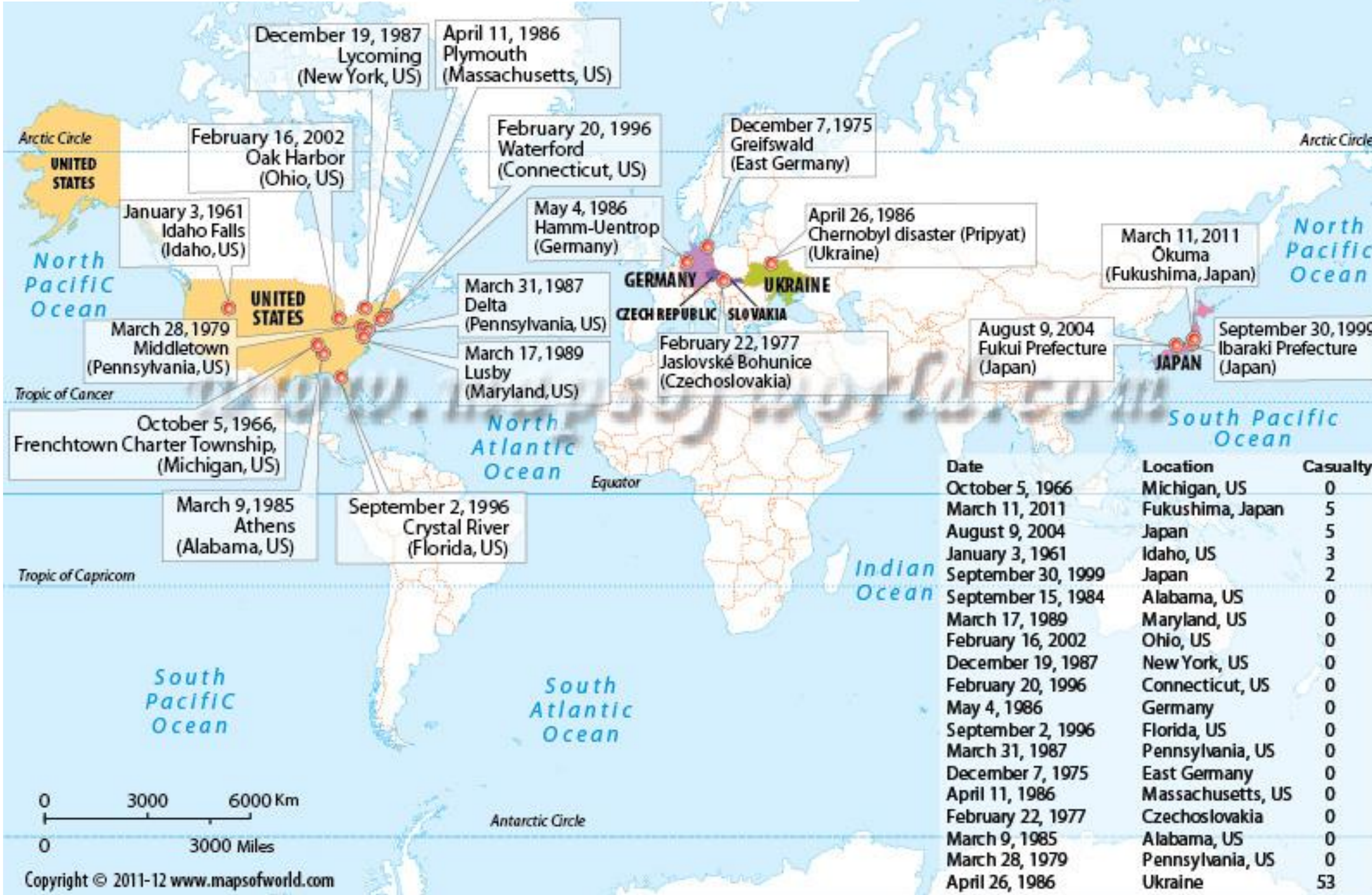
1977-

Source from nuclear fuel reprocessing discharges to Irish sea
Sellafield- peak inputs in mid-1970's



Replotted from data in
IAEA Tech Doc 1429, 2005

Many accidental releases associated with nuclear power plants



Date	Location	Casualty
October 5, 1966	Michigan, US	0
March 11, 2011	Fukushima, Japan	5
August 9, 2004	Japan	5
January 3, 1961	Idaho, US	3
September 30, 1999	Japan	2
September 15, 1984	Alabama, US	0
March 17, 1989	Maryland, US	0
February 16, 2002	Ohio, US	0
December 19, 1987	New York, US	0
February 20, 1996	Connecticut, US	0
May 4, 1986	Germany	0
September 2, 1996	Florida, US	0
March 31, 1987	Pennsylvania, US	0
December 7, 1975	East Germany	0
April 11, 1986	Massachusetts, US	0
February 22, 1977	Czechoslovakia	0
March 9, 1985	Alabama, US	0
March 28, 1979	Pennsylvania, US	0
April 26, 1986	Ukraine	53

http://www.mapsofworld.com/world-maps/major-nuclear-disasters.html

Other sources to the marine environment

4) Satellite accidents

SNAP 9A, 1964 burn-up of ^{238}Pu powered satellite 0.004 PBq

5) Undetonated bomb accidents

Thule Greenland; Johnston atoll, Pacific
Palomares Spain

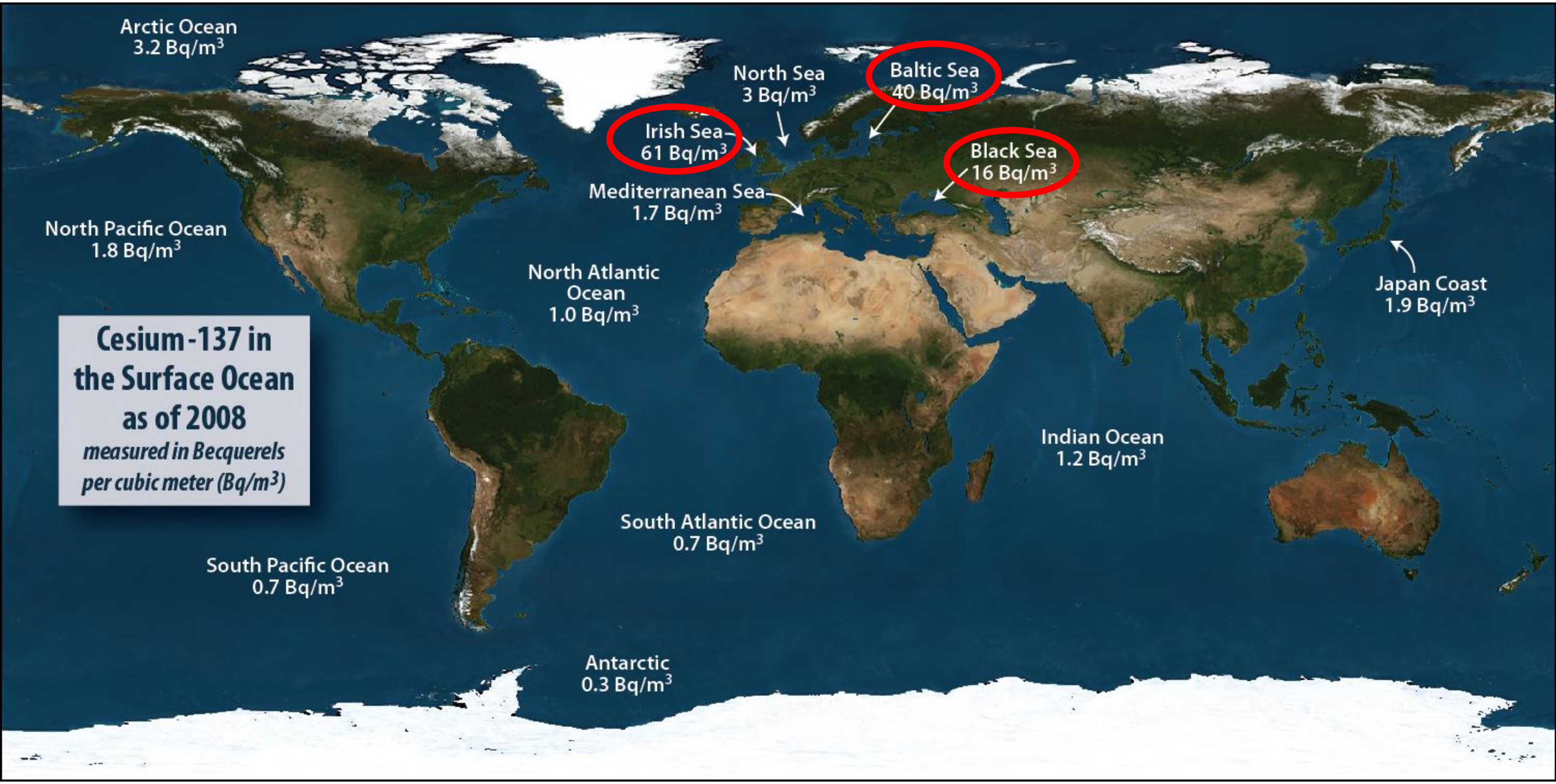
6) Waste dumping

At sea dumping of low level waste, now ended ~45 PBq
Medical wastes for hospitals
High level waste- reactors from subs, icebreakers in Arctic ~40 PBq

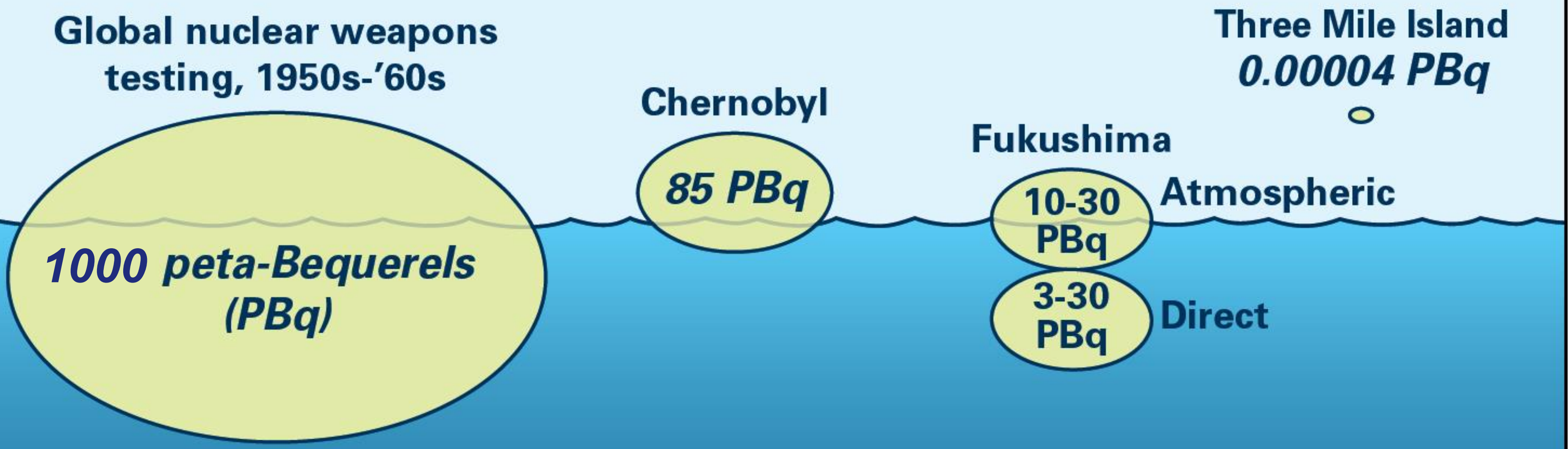
7) Nuclear Submarines

Operations (^{60}Co)
Accidents US, FSU

End result is uneven distribution in ocean

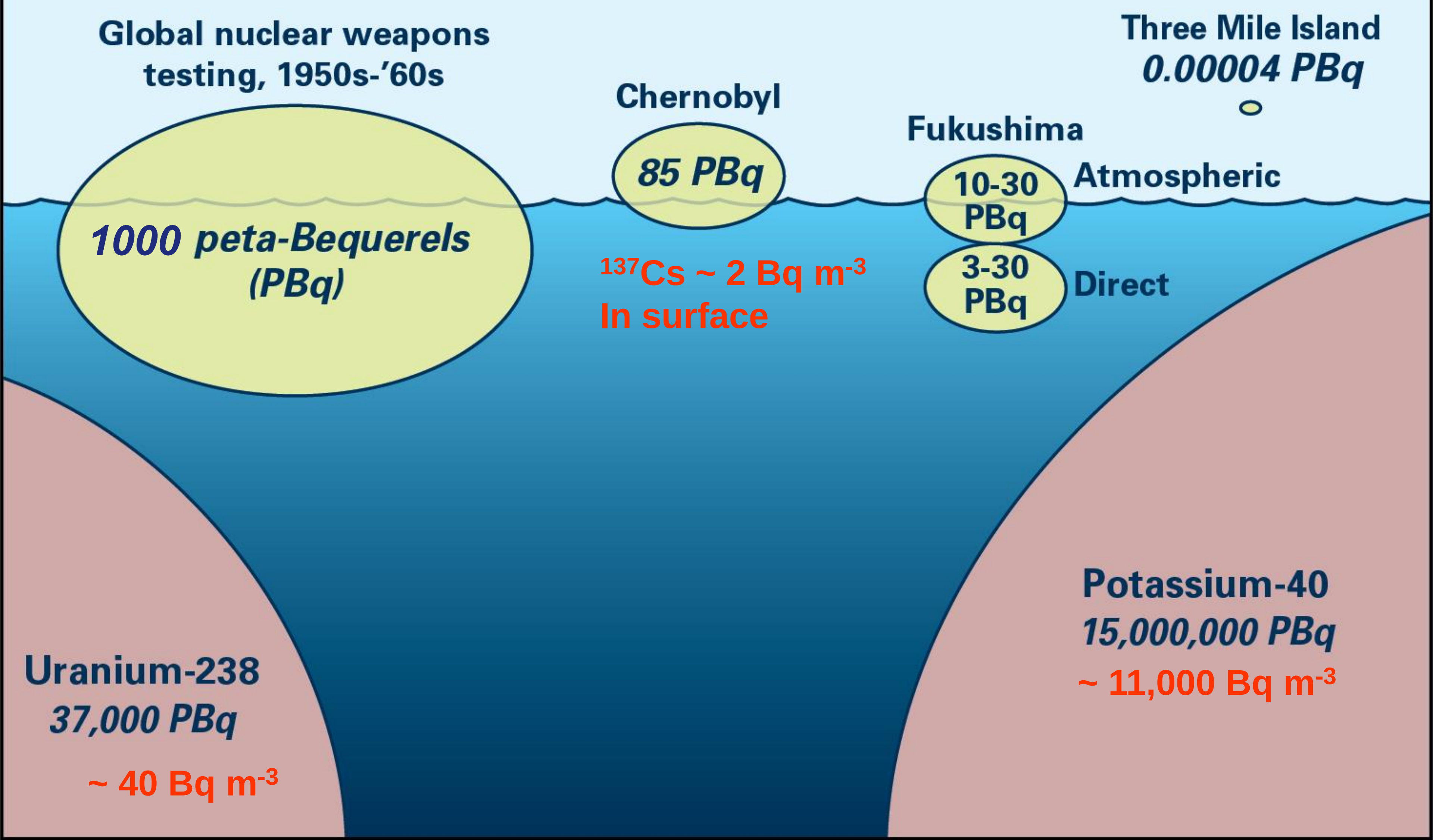


Total cesium-137 : what are largest sources?

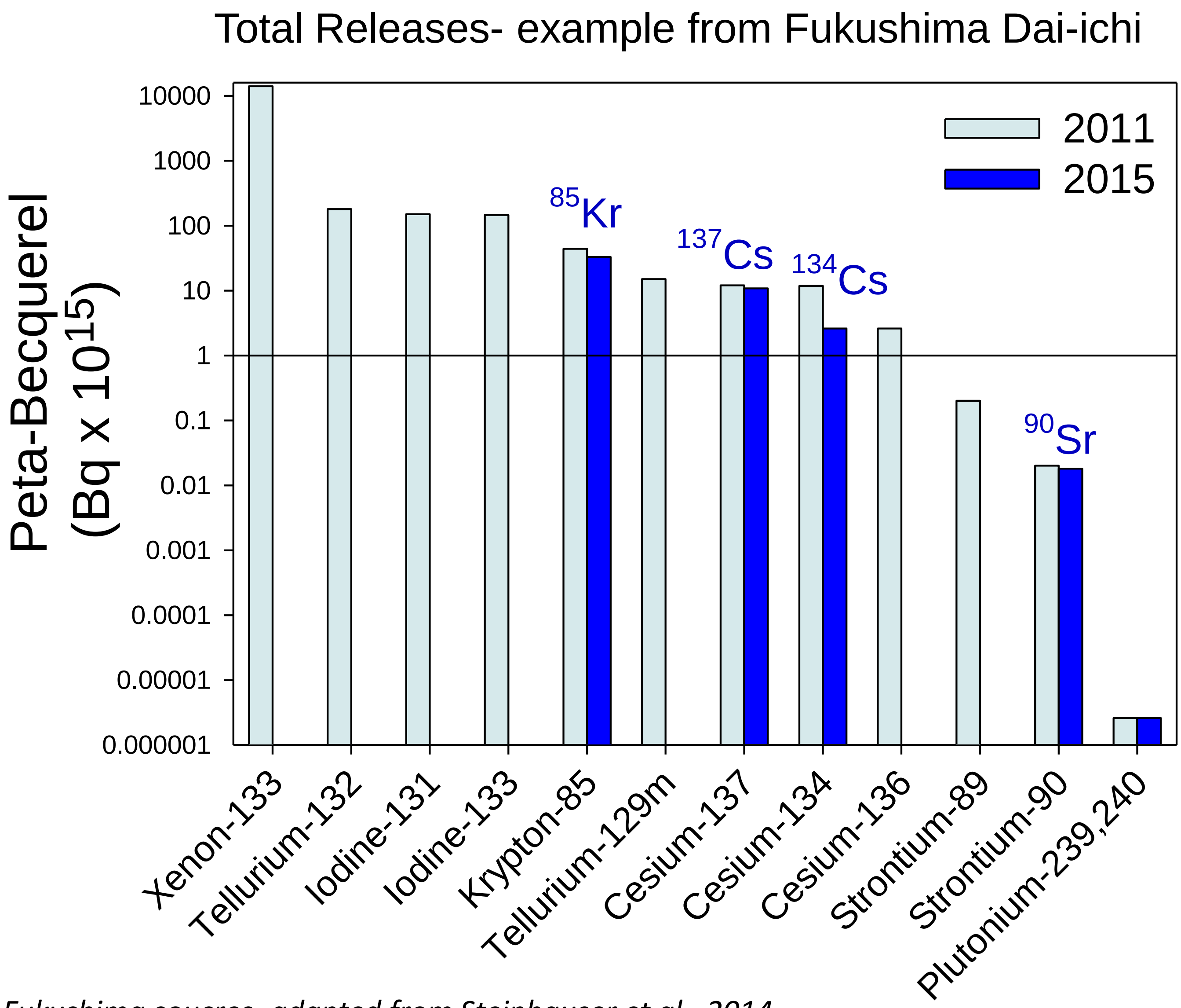


1 Bq = 1 Becquerel = one radioactive decay per second
1 PBq = peta-Becquerel = one million billion Bq
10^{15} Bq = 1,000,000,000,000,000 Bq

Total cesium-137 : how does it compare to natural radioactivity?



Why so much attention on cesium?



Fukushima soucres- adapted from Steinhauser et al., 2014

Applications of artificial radionuclides in ocean

1) Water/Atmospheric mixing rates (including groundwater and soil erosion)

^{3}H, ^{137}Cs, ^{14}C, ^{90}Sr

2) Particle scavenging & transport rates in water column

Pu, ^{241}Am, ^{106}Ru, ^{144}Ce

3) Sediment accumulation & mixing

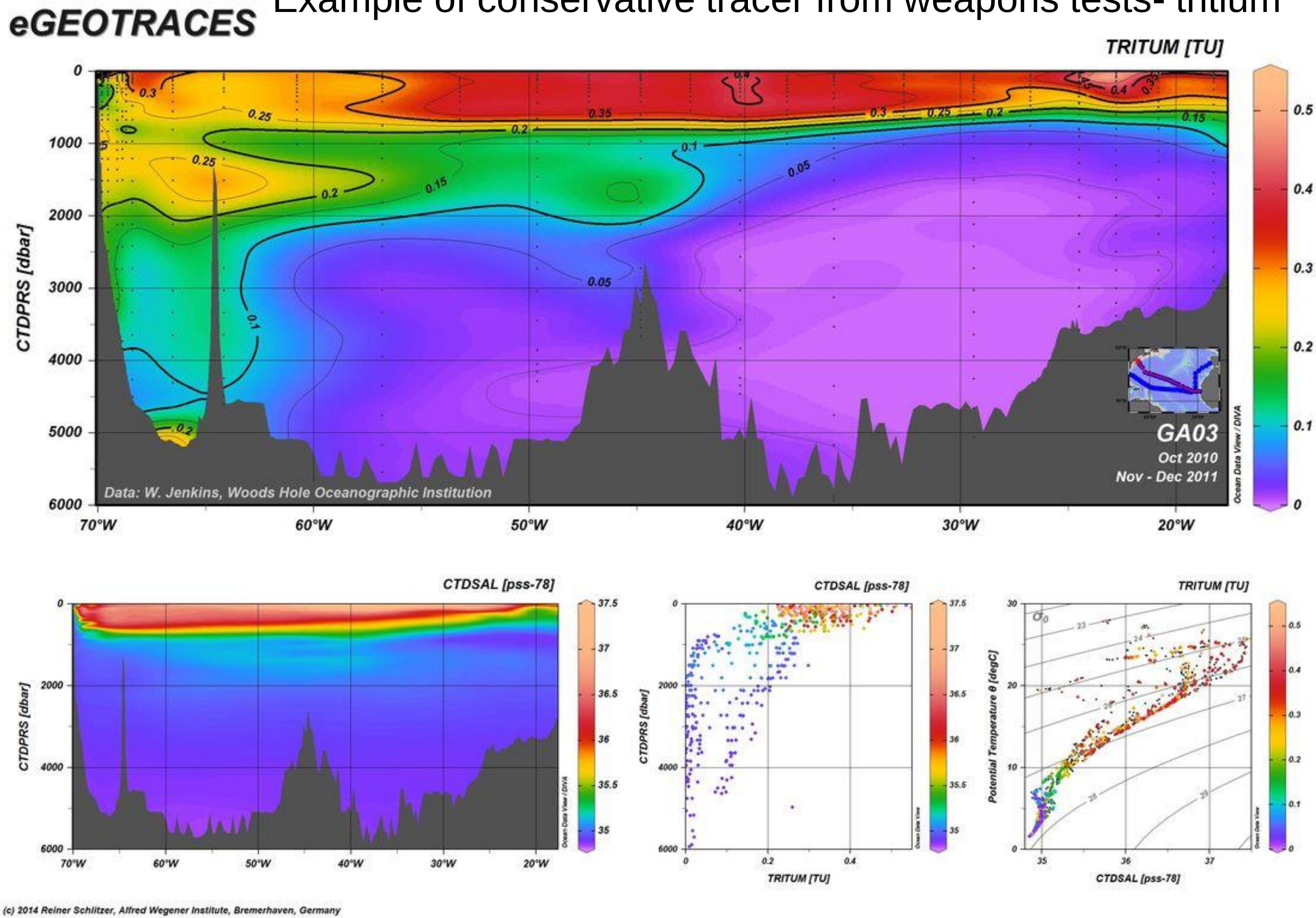
Pu, ^{137}Cs

4) ages/dating of specific elements/compounds

DOC/COC; compound specific ^{14}C

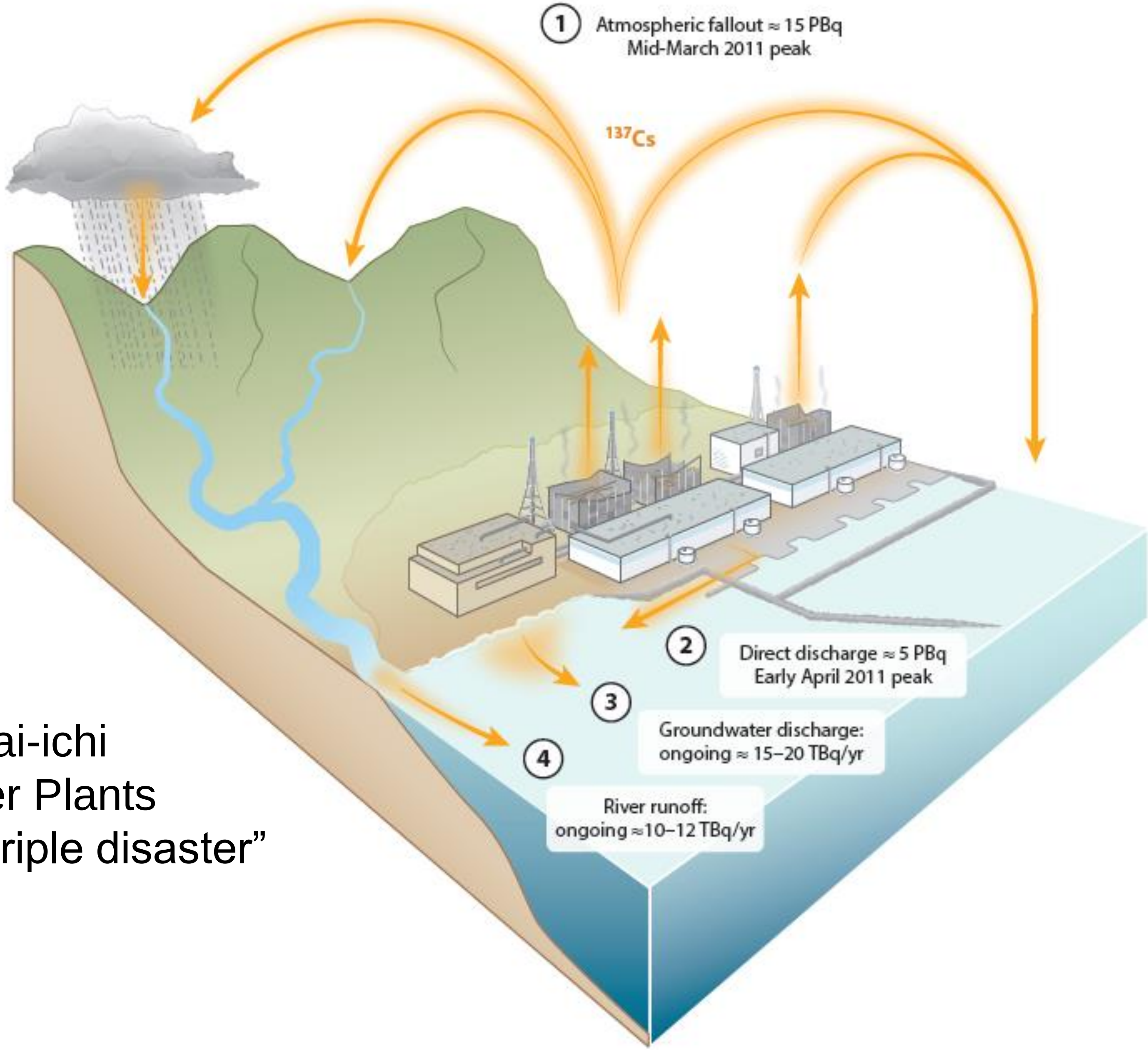
Fallout radionuclide records in corals; mollusks

Example of conservative tracer from weapons tests- tritium



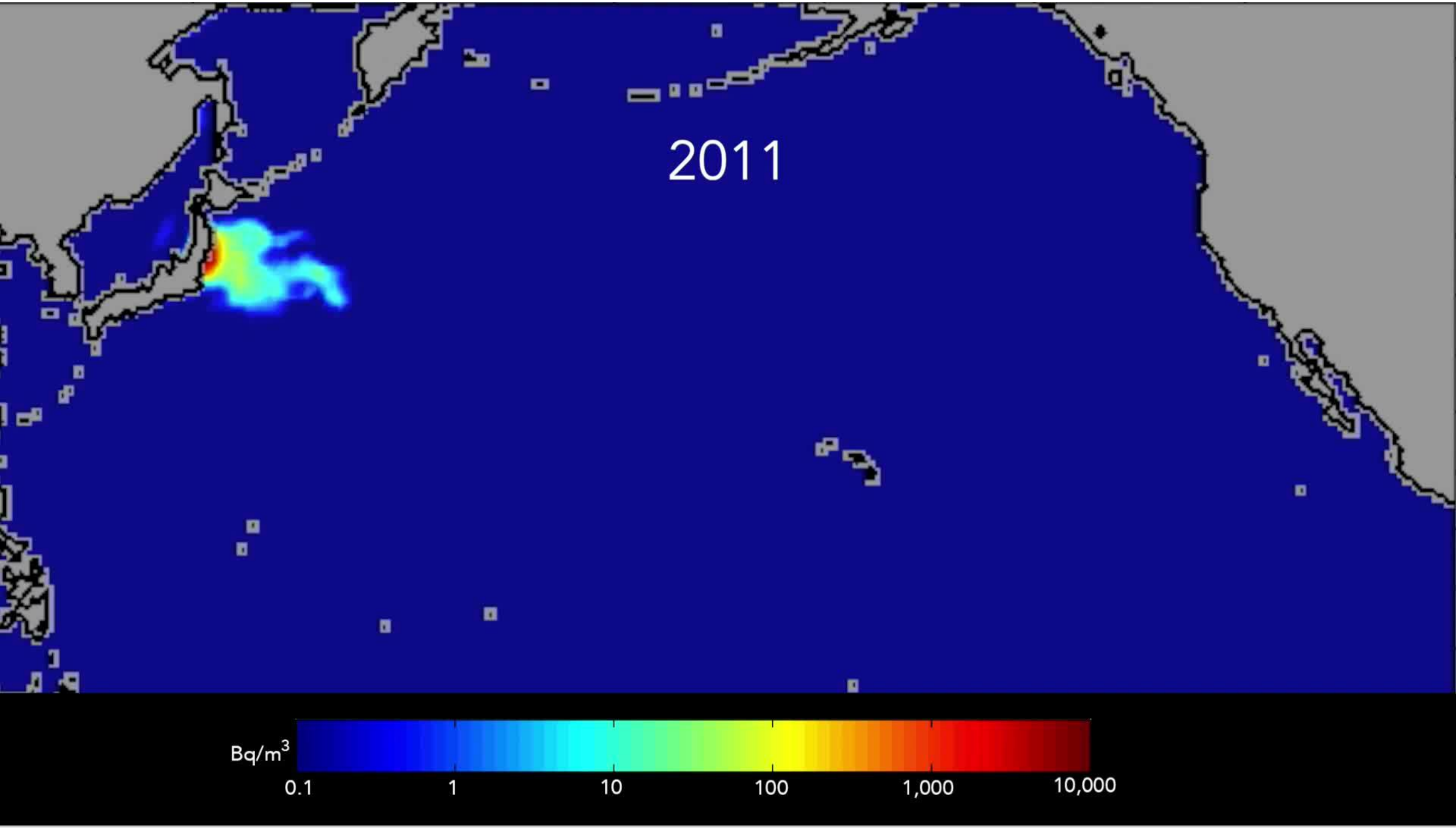
http://www.egeotraces.org/scenes.html

Example of artificial radionuclides from nuclear disaster



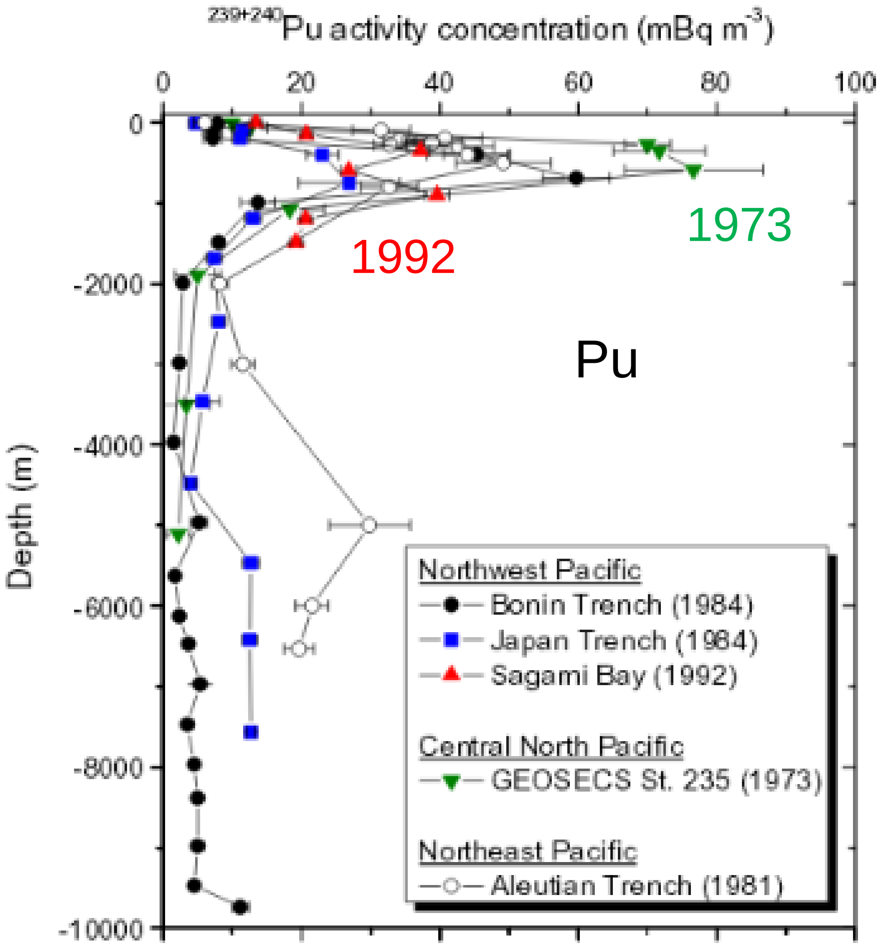
Fukushima Dai-ichi
Nuclear Power Plants
March 2011 "triple disaster"

Fukushima cesium as tracer of currents and mixing

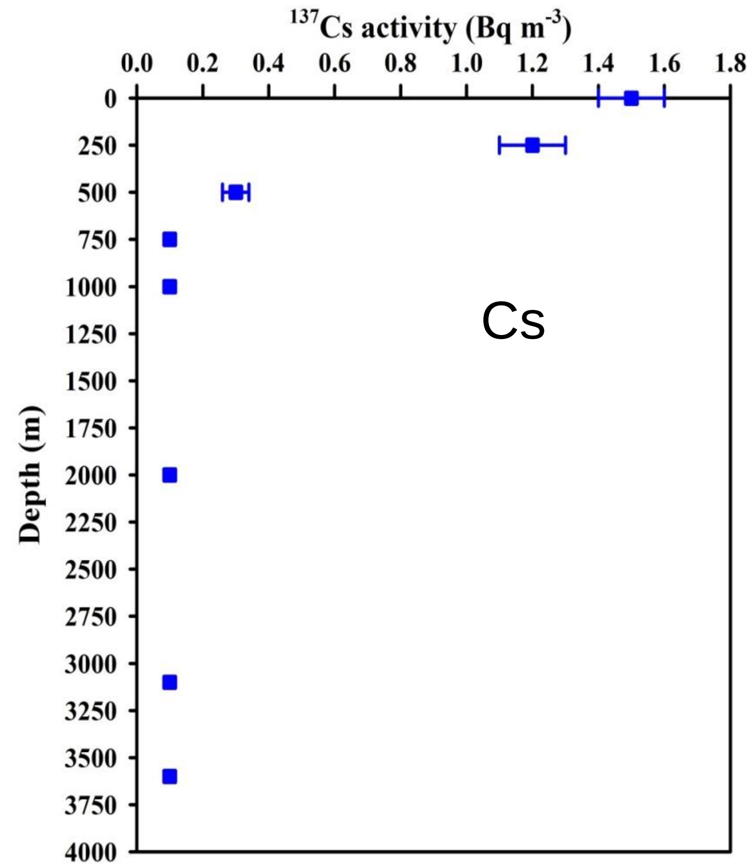


Rossi et al., 2013 & 2014

Fallout Pu - particle scavenging leads to decrease with time
- contrasts with Cs and other conservative fallout isotopes

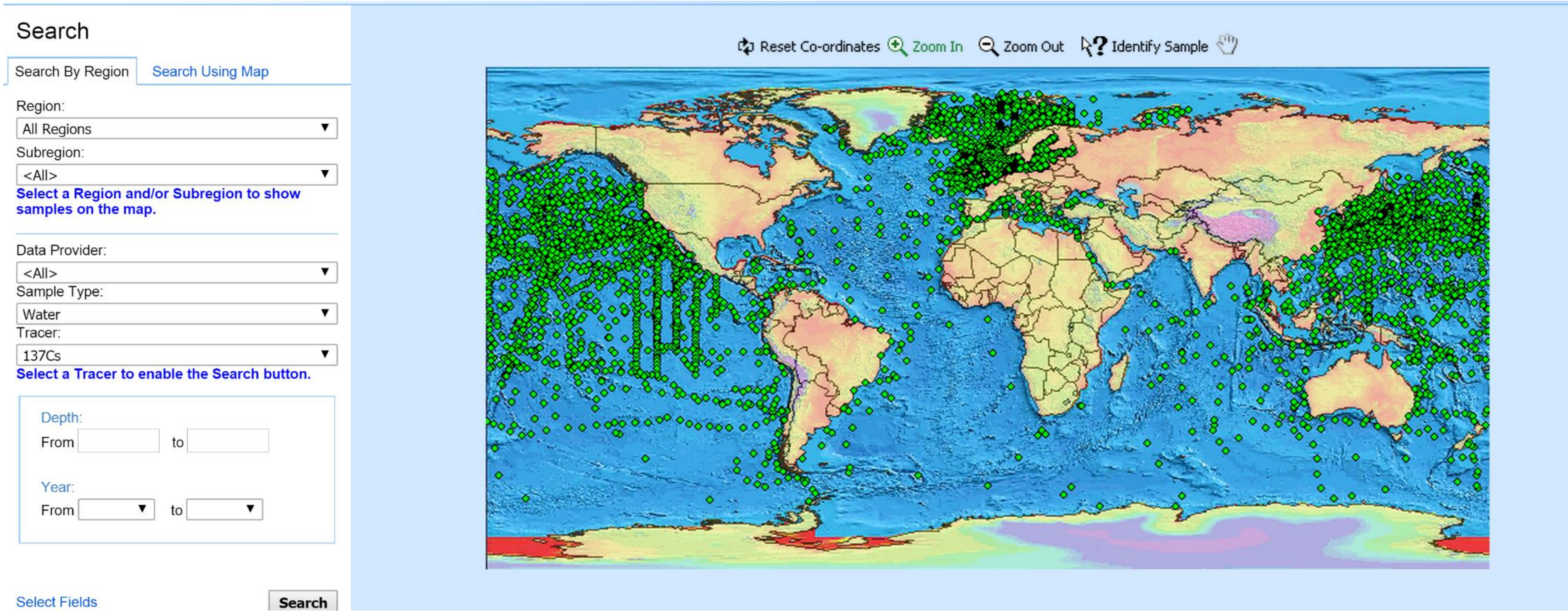


Lindahl et al., 2010



Povinec et al., 2003

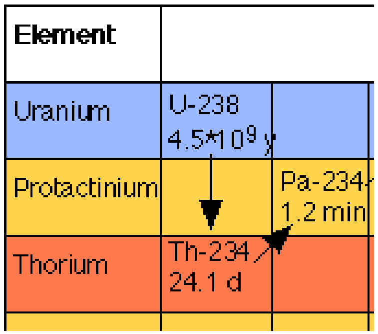
How to find marine radioactivity data?
e.g. IAEA MARiS Database for Artificial Radionuclides

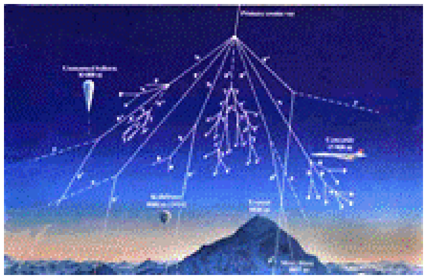


http://maris.iaea.org

Radionuclides in the Marine Environment- SUMMARY

1) The 3 main sources of radionuclides to the marine environment





2) Key examples of how radionuclides can be used to trace a variety of ocean processes- scavenging and conservative tracers