LIMNOLOGY AND OCEANOGRAPHY
ASLO e-Lectures

Radioactivity in the Marine Environment

Cosmogenic and Anthropogenic Radionuclides

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ASLO e-Lectures

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

Claudia Benitez-Nelson

doi:10.1002/loe2.10008

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, $^{222, 224, 226, 228}Ra$ and ^{222}Rn.

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

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

3. **Anthropogenic radionuclides** – continuously being produced by humans.

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

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

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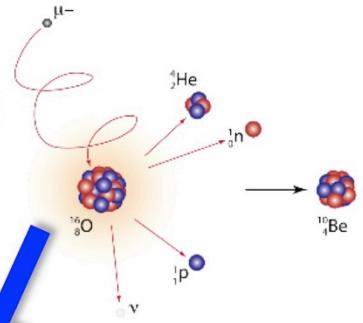
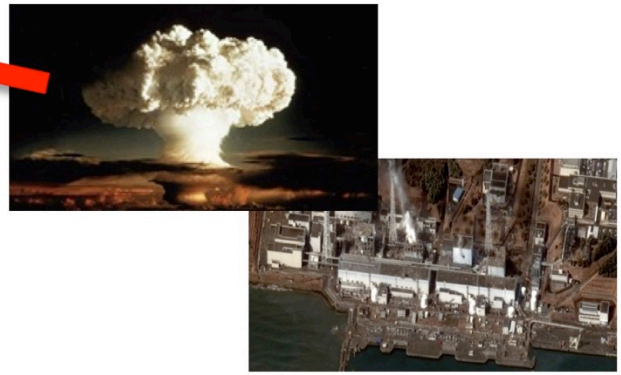
Three main sources of radionuclides to the Marine Environment:

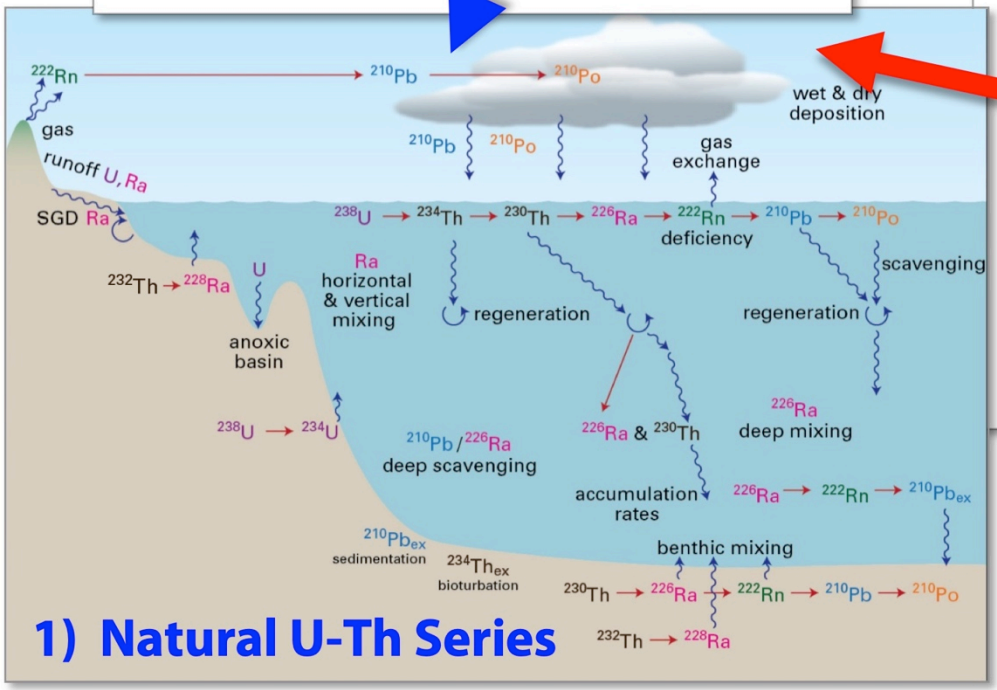
2) Cosmogenic (^{7}Be, ^{14}C)



3) Anthropogenic Radionuclides (^{90}Sr, ^{137}Cs)



1) Natural U-Th Series



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Cosmogenic Radionuclides

Table 2
Cosmogenic nuclide source functions in oceans, for nuclides of half-lives > 10 days, arranged in order of increasing half-lives

Nuclide	Half-life	Principal target element(s)		Global average surface injection rate[a]	Integrated in situ oceanic production rate[b]
		Atmosphere	In ocean water	(atoms/cm^2 per min)	
^{32}P	14.3 days	Ar	Cl, S, K	5.82×10^{-3}	7.6×10^{-4}
^{33}P	25.3 days	Ar	Cl, S, K	6.93×10^{-3}	2.9×10^{-4}
^{37}Ar	35.0 days	Ar	K, Ca	9.10×10^{-6}	8.1×10^{-6}
^{7}Be	53.3 days	N,O	O	1.27	6.0×10^{-3}
^{35}S	87.4 days	Ar	Cl, Ca, K	2.84×10^{-2}	5.1×10^{-4}
^{22}Na	2.6 years	Ar	Na	3.75×10^{-3}	3.9×10^{-4}
^{3}H	12.3 years	N,O	O, ^{2}H	1.39×10^{-1}	1.2×10^{-2}
^{32}Si	~ 150 years	Ar	S, Ca	9.60×10^{-3}	2.5×10^{-5}
^{39}Ar	269 years	Ar	K, Ca	2.00×10^{-1}	1.2×10^{-5}
^{14}C	5730 years	N,O	O	1.20×10^{2}	9.0×10^{-3}
^{41}Ca[c]	1.0×10^{5} years	–	Ca	–	2.4×10^{-5e}
^{81}Kr	2.1×10^{5} years	Kr	Sr	2.30×10^{-5}	1.9×10^{-8e}
^{36}Cl[d]	3.0×10^{5} yeatrs	Ar	Cl	6.60×10^{-2}	1.06×10^{-1e}
^{26}Al	7.2×10^{5} years	Ar	S, K, Ca	8.40×10^{-3}	6.8×10^{-6}
^{10}Be	1.6×10^{6} years	N,O	O	2.70	1.8×10^{-3}

[a] Based on atmospheric production rate estimates of Lal and Peters (1967). The mean stratosphere–troposphere exchange time is taken to be 2 years. Tropospheric residence time of isotopes that can be scavenged by wet precipitation is taken to be 40 days. Atmospheric residence times for ^{14}C, ^{39}Ar, and ^{81}Kr, for mixing with the mixed layer of the ocean, are taken to be 10, 270, and 170 years, respectively.

[b] Lal et al. (1988).

[c] Flux to oceans from rivers should be included to take into account production in rocks and soil by ^{40}Ca(n,γ) ^{41}Ca reaction; this estimate is not given here because of large uncertainties in these calculations.

[d] As above, due to ^{35}Cl(n,γ) ^{36}Cl reaction.

[e] The in situ production of ^{41}Ca, ^{81}Kr and ^{36}Cl in the oceans is primarily due to the relevant thermal neutron capture reaction. Note the 50% greater in situ production of ^{36}Cl in the oceans compared to its atmospheric source.

Lal, 1999

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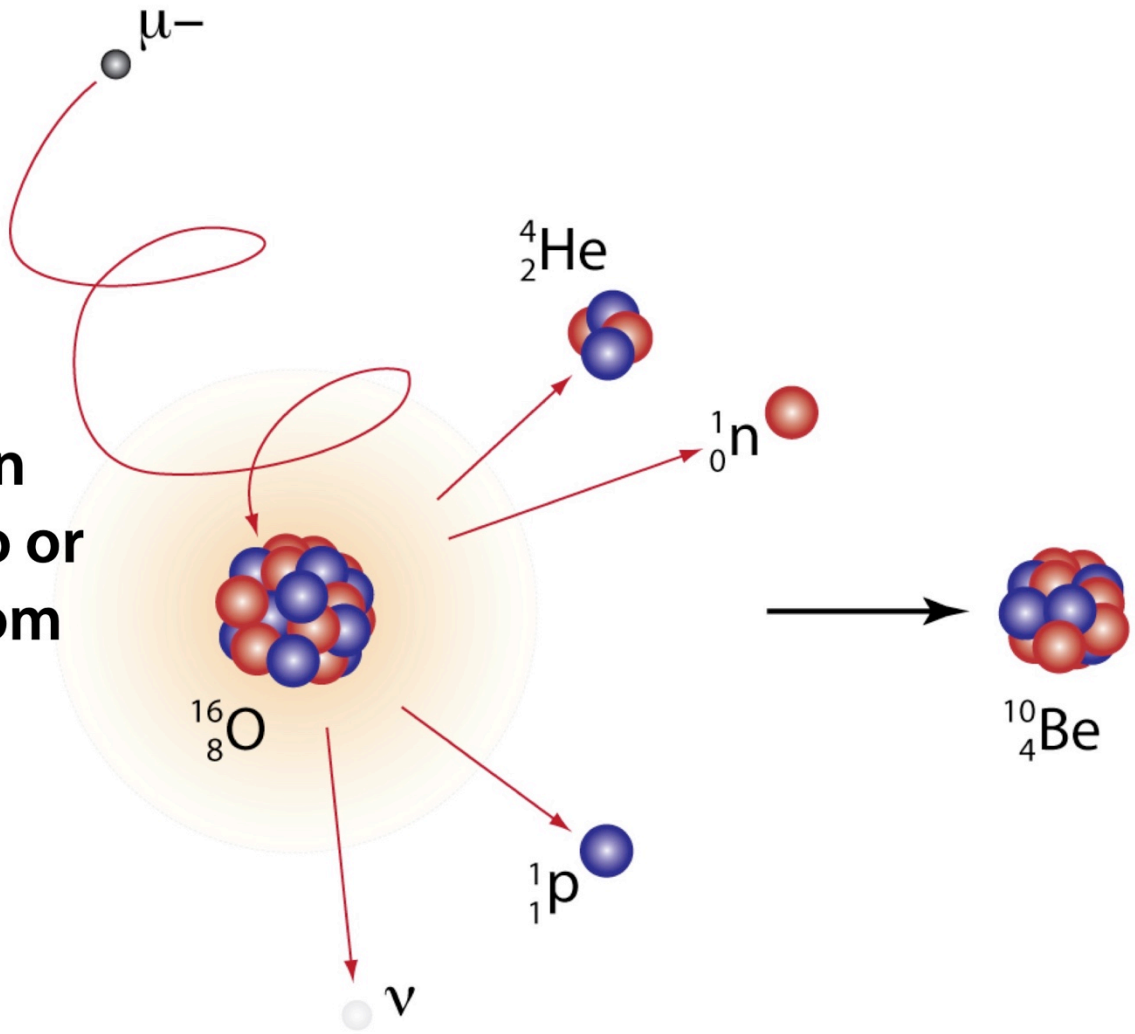
Cosmogenic Radionuclides

Sources:

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



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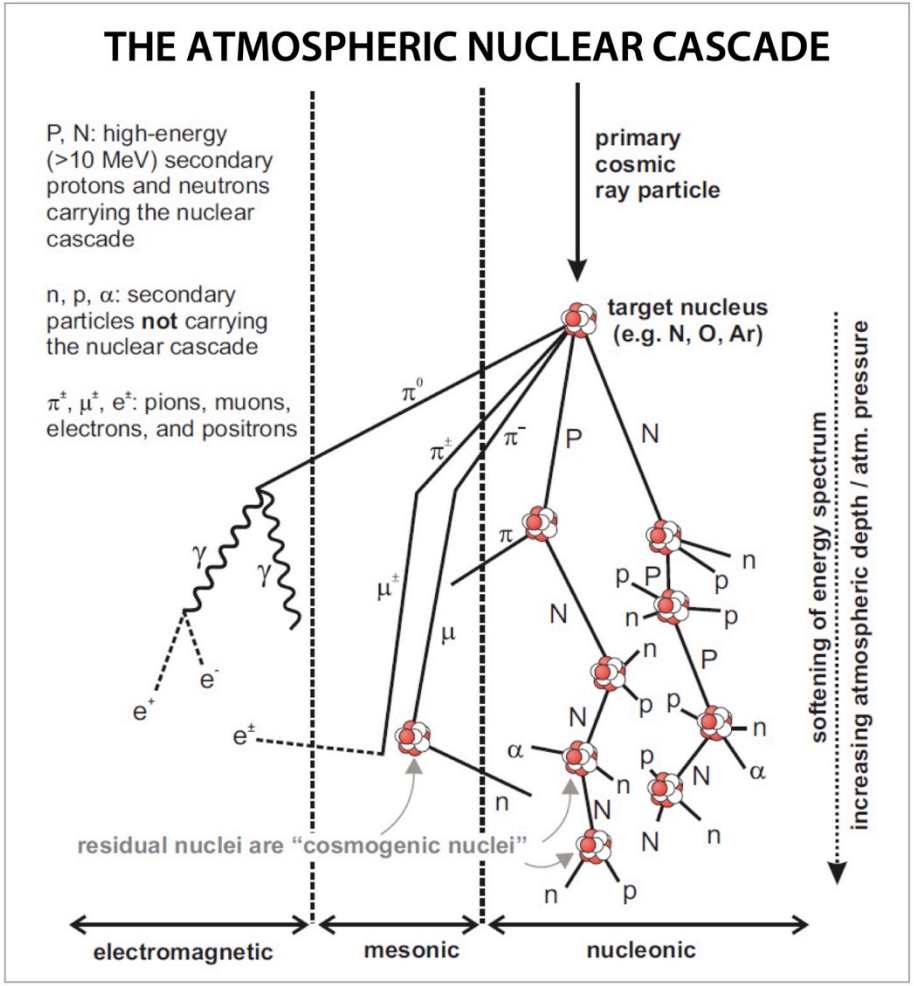
Cosmic Ray Shower

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 (protons)
10% helium, &
1% heavier elements.

Dunai and Lifton (2014)



Nuclear reactions in the atmosphere produce atmospheric cosmogenic nuclides as well as secondary particles, mostly neutrons. The latter are responsible for the cosmogenic nuclides measured in minerals. (Figure after Dunai, 2010)

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Cosmogenic Radionuclides

Cosmogenic nuclide production varies with latitude in the atmosphere!

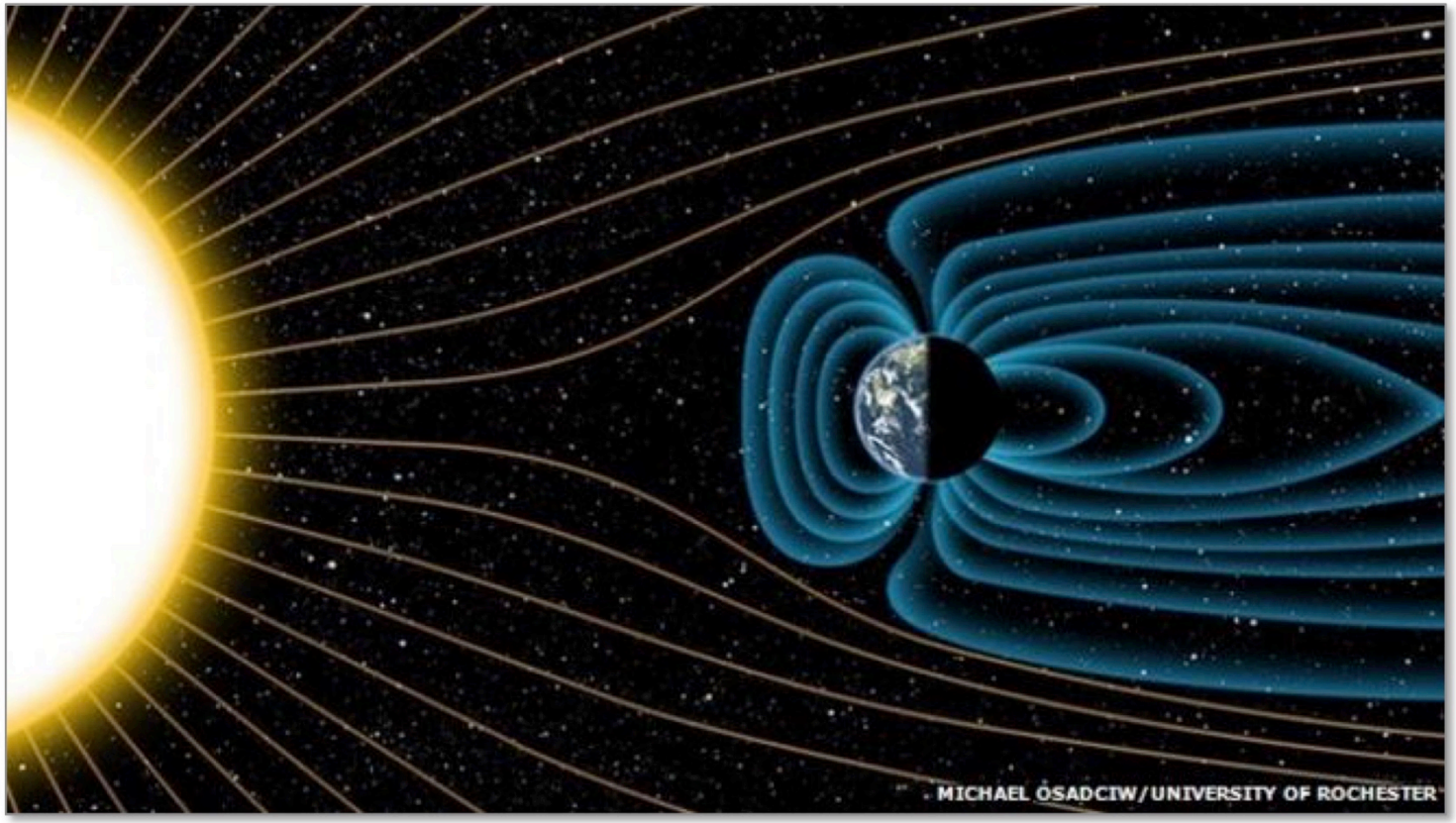

Image of solar winds deflected by Earth's magnetic field

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

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^{7}Be production in the Atmosphere

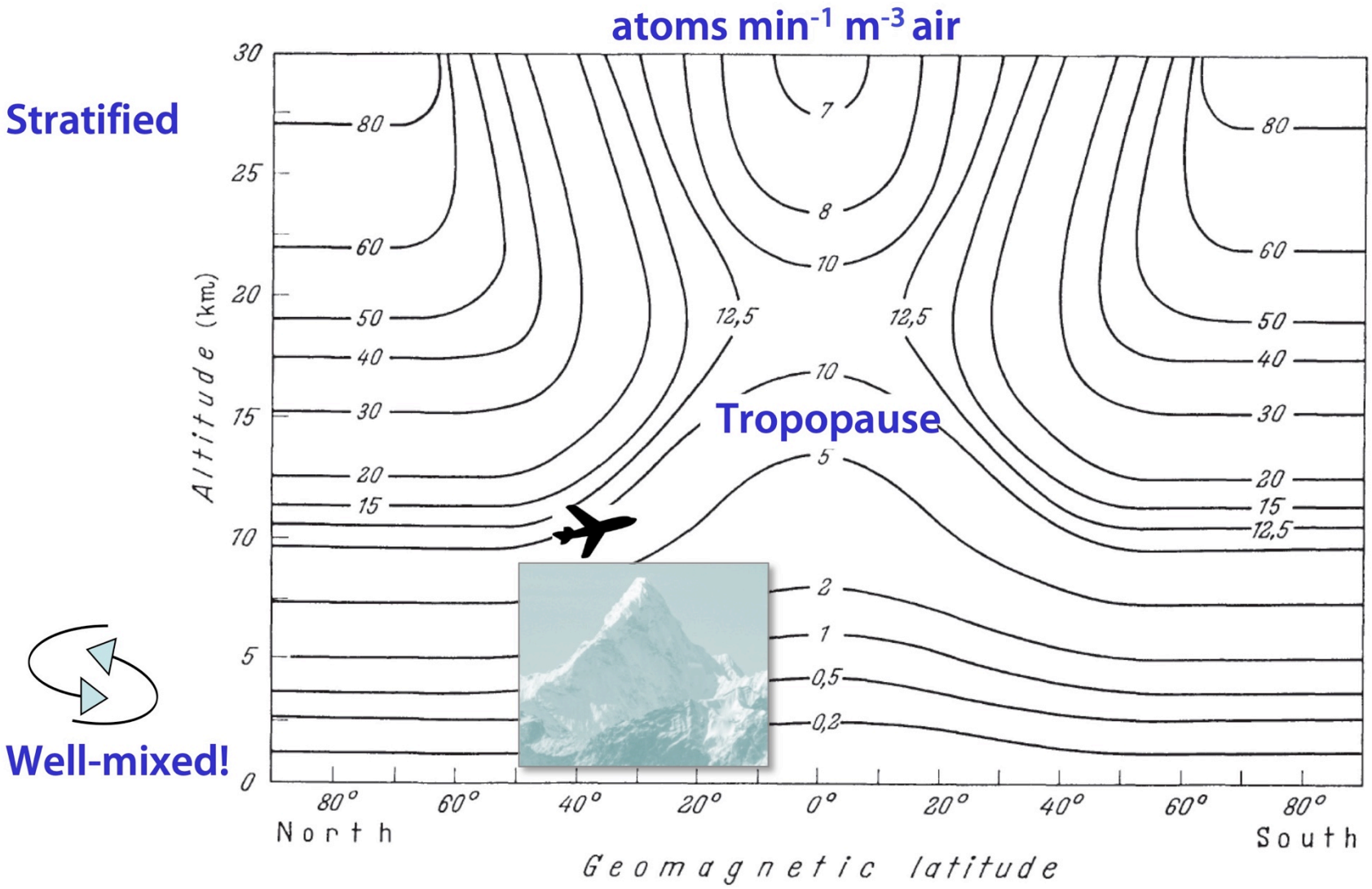
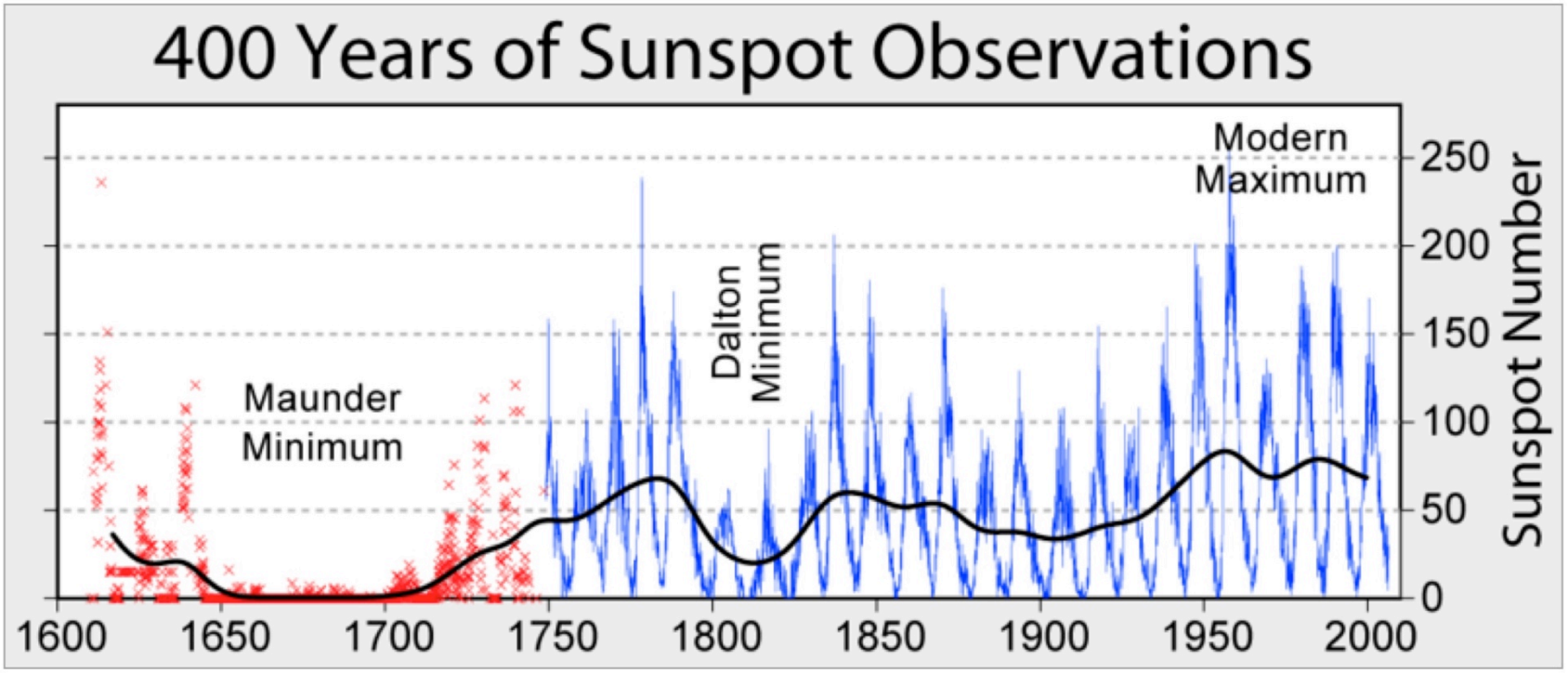


Fig. 20. A north-south section through the atmosphere showing surfaces of constant Be^{7} production (nuclei/min, m^{3} air at S.T.P.).

Lal and Peters (1967)

doi:10.1002/loe2.10008

Cosmogenic nuclide production varies over seasonal to multidecadal time scales



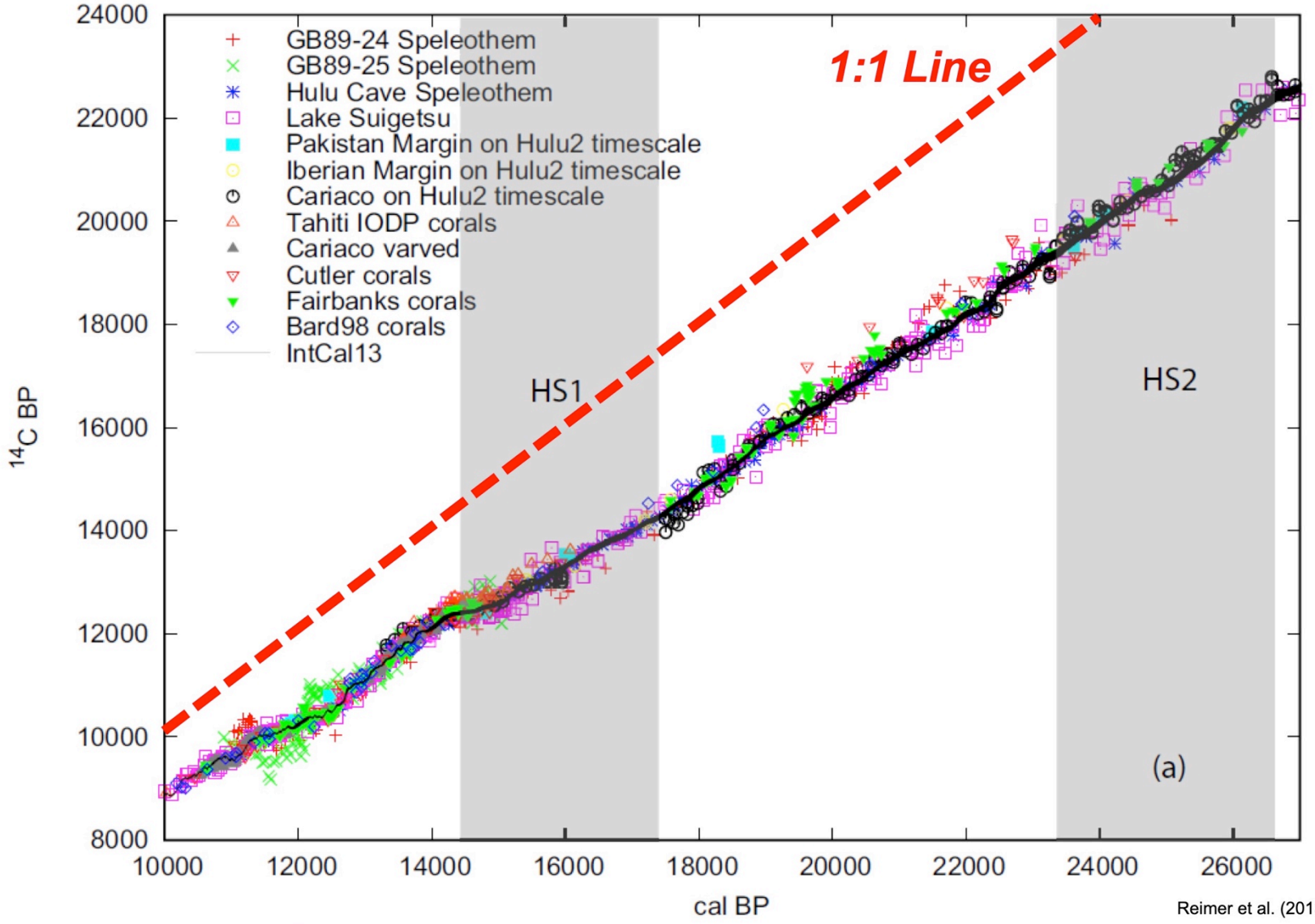
http://oneminuteastronomer.com/1042/sunspots-cycles/

"Little Ice Age"

Source variability complicates use of ^{14}C

doi:10.1002/loe2.10008

Cosmogenic Radionuclides: Correction Factors



Reimer et al. (2013)

doi:10.1002/loe2.10008

What are Cosmogenic Radionuclides used for?

Many studies have focused on ^{14}C (complicated by "bomb" ^{14}C and "Suess Effect") and ^{7}Be given their well-defined source (atmospheric) to the surface ocean.

Studies include:

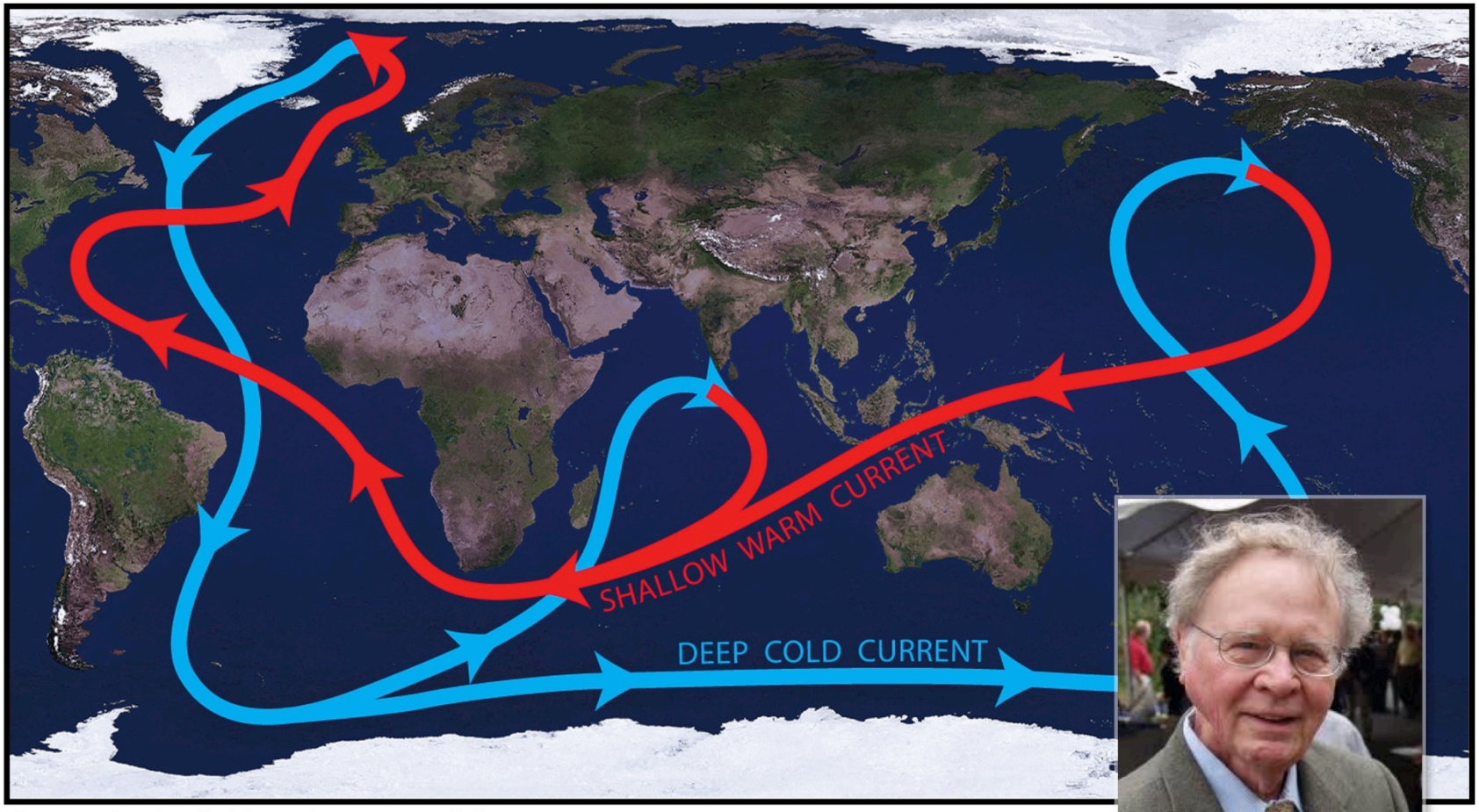
a) Large-scale ocean circulation
^{14}C ($t_{½}$ = 5,700 yr)

b) Short time scale upper ocean vertical mixing rates and atmospheric deposition
^{7}Be ($t_{½}$ = 53 d)

c) Age dating, mixing, particle transport and erosion
^{7}Be ($t_{½}$ = 53 d)
^{14}C ($t_{½}$ = 5,700 y)

doi:10.1002/loe2.10008

Cosmogenic Radionuclides: a) Ocean Circulation

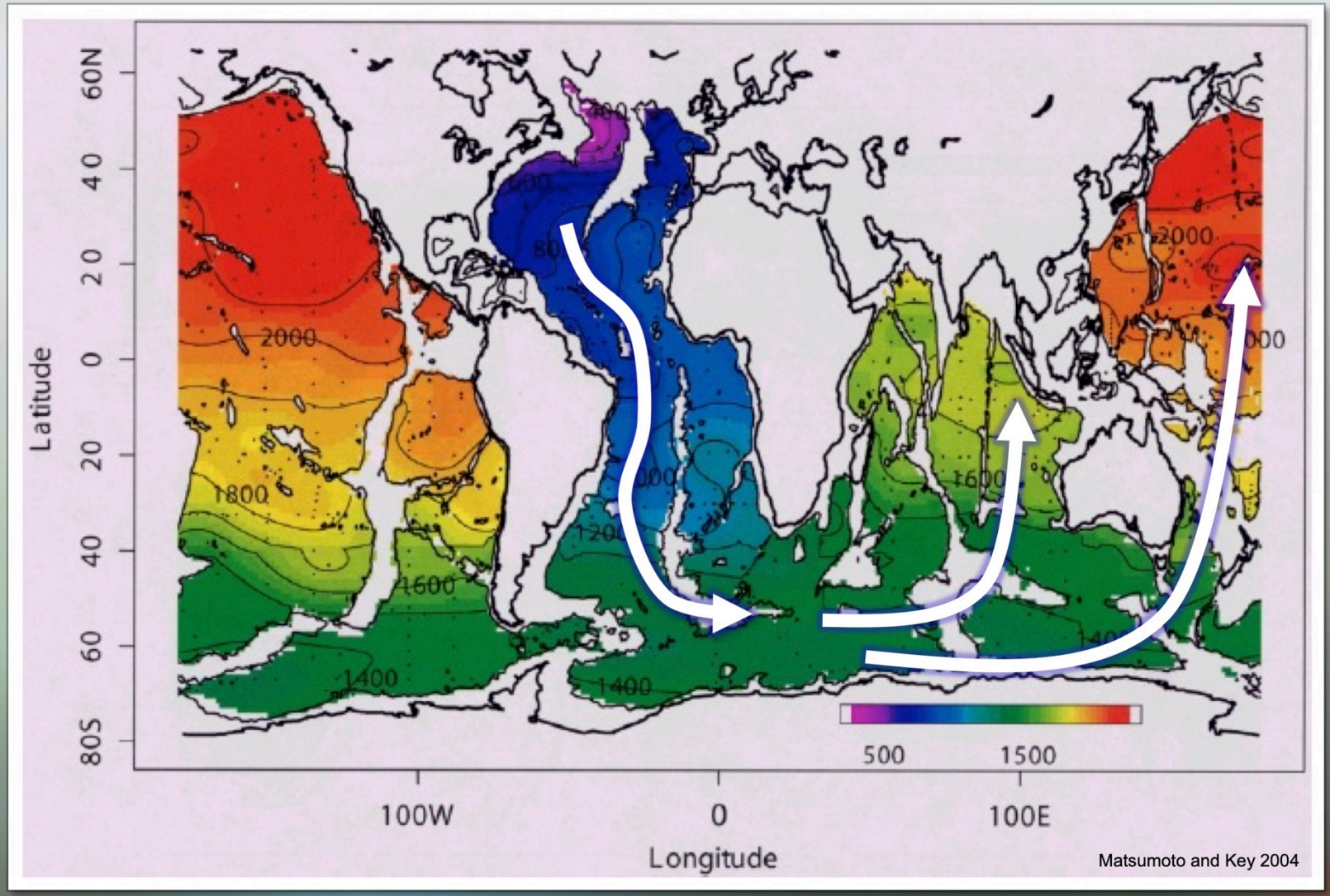


stempreacademy.hawaii.edu/c-more/ocean-conveyor-belt

W. Broecker

doi:10.1002/loe2.10008

Circulation and age of ocean waters at 3500 m determined from ^{14}C



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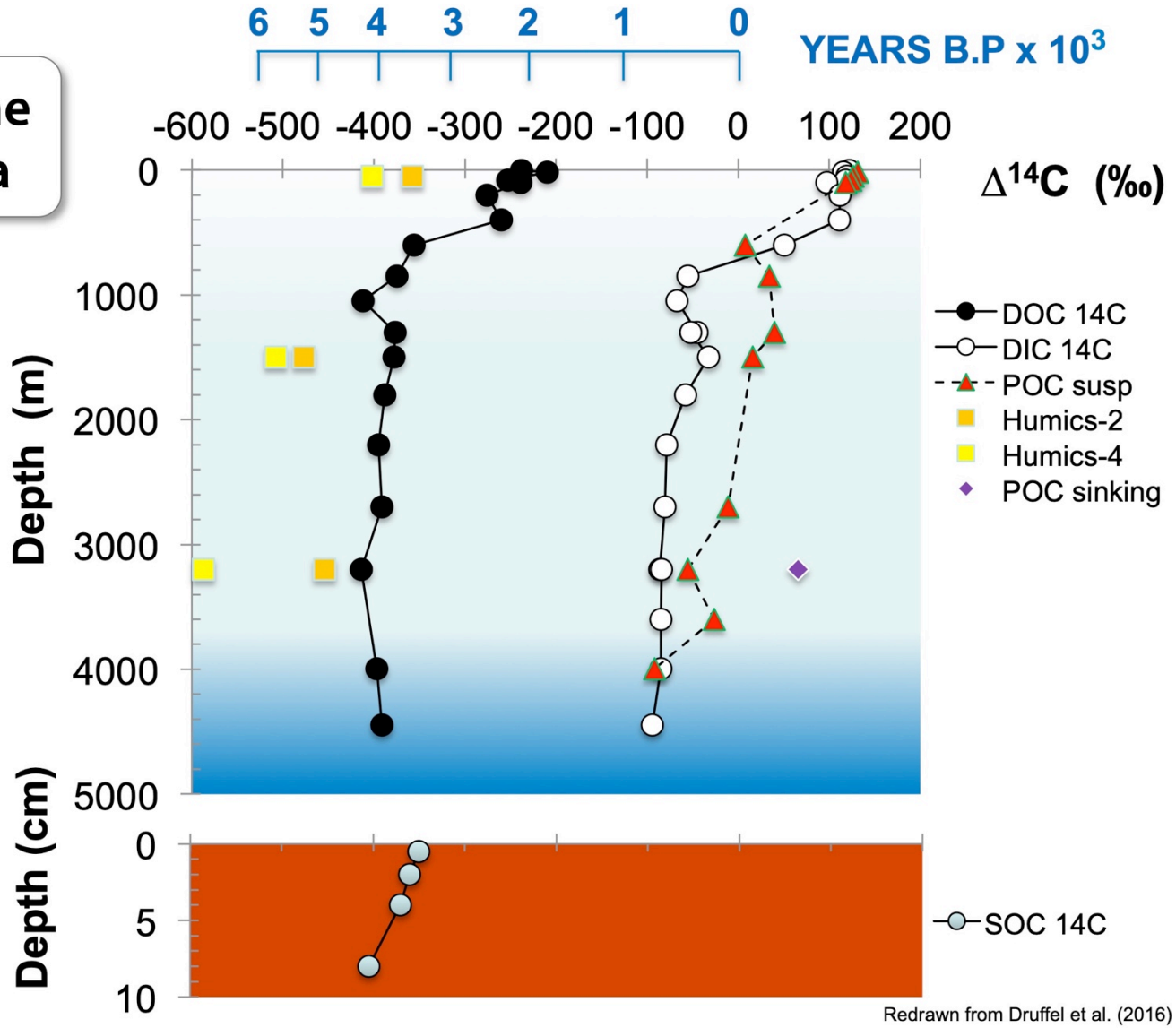
doi:10.1002/loe2.10008

Cosmogenic Radionuclides: c) Age Dating

Data from the Sargasso Sea

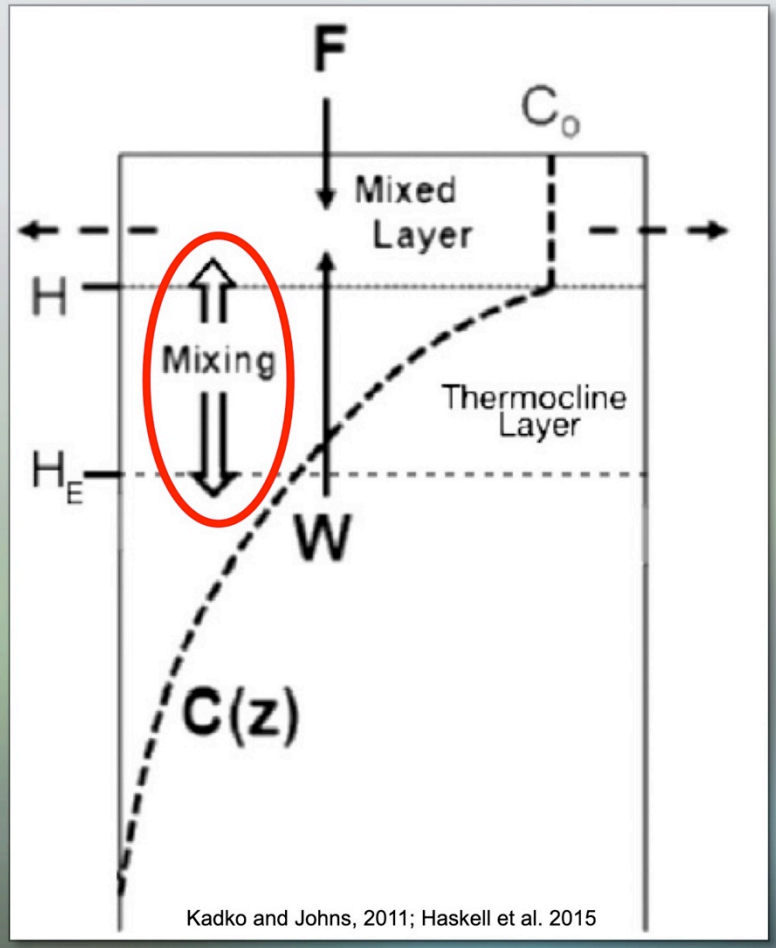


Redrawn from Druffel et al. (2016)

doi:10.1002/loe2.10008

Cosmogenic Radionuclides: b) Upper Ocean Mixing

^{7}Be ($t_{½}$=53 d) as conservative tracer for upper ocean mixing: conceptual model



Kadko and Johns, 2011; Haskell et al. 2015

$$H\frac{\partial C}{\partial t} = F_a - J - \lambda C_0 H + w_H C_o - P$$

$$J = w_H C_o - K_z\left(\frac{\partial C}{\partial z}\right)_H$$

- C_o = change in ^{7}Be concentration along a mixing line (dashed)
- F_a = Atmospheric ^{7}Be input (assume constant)
- P = Particle export (assume negligible)
- H = mixed layer depth
- H_E = depth of euphotic zone
- W_H = upwelling velocity
- Dashed arrows = horizontal flow

Vertical mixing or eddy diffusivity (K_z) is derived from the vertical profile of ^{7}Be activities.

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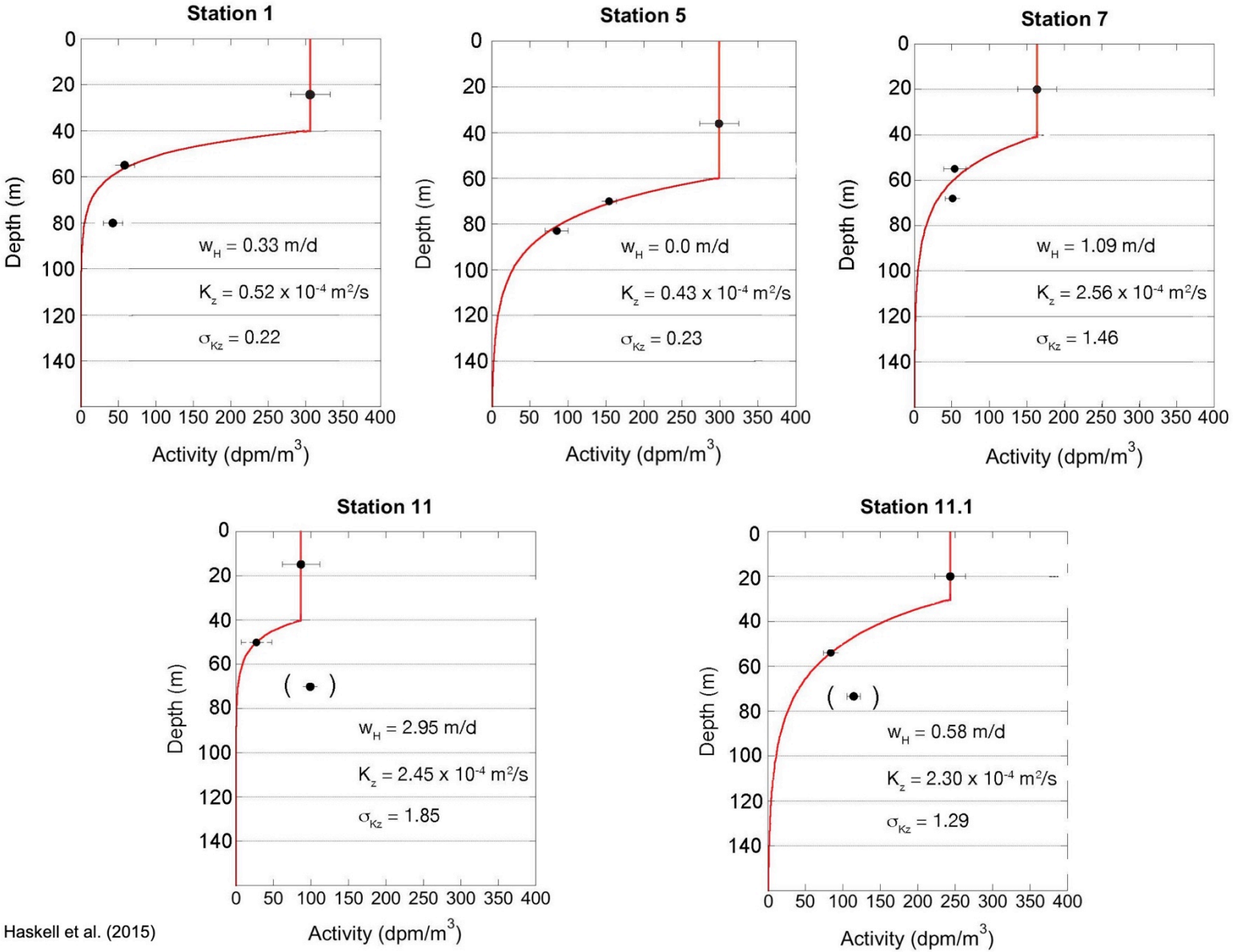

Haskell et al. (2015)

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Anthropogenic Radionuclides

Wikipedia.org

Anthropogenic Radionuclides are those radioactive nuclides that have been produced by humankind.

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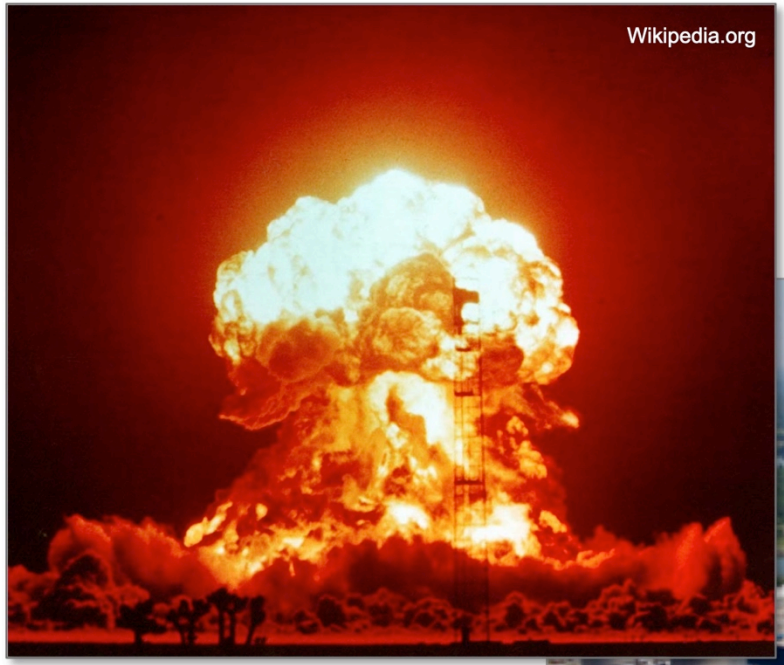
Major sources of Anthropogenic Radionuclides to the environment



1) Nuclear weapons

2) Nuclear fuel cycle

Tennessee Valley Authority

doi:10.1002/loe2.10008

Why produce Anthropogenic radionuclides?

Nuclear power

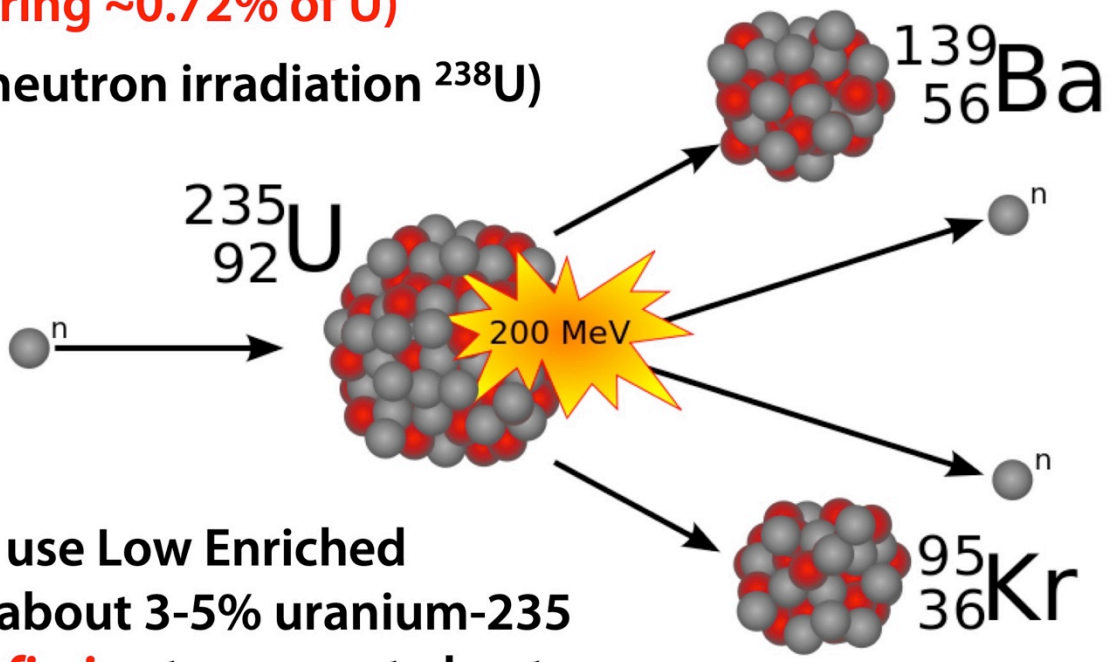
It's all about using a fission chain reaction to release energy!

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.

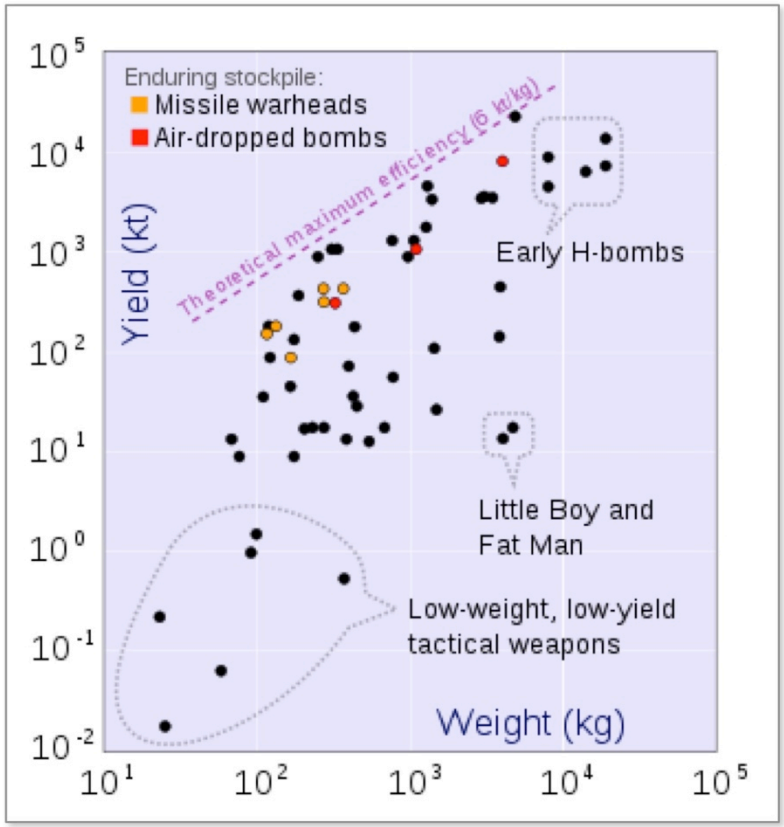
doi:10.1002/loe2.10008

Why produce Anthropogenic radionuclides?

Nuclear weapons

Reactions involved in nuclear weapons are characterized by *uncontrolled nuclear fission*.

High Enriched Uranium (HEU) is comprised of more than 20% ^{235}U 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 nuclear bomb.



Weapons-grade HEU is typically defined as 90% HEU or above. This minimizes weapon size. Smaller and lighter nuclear weapons are much easier to deliver. Some nuclear weapons also use Pu.

doi:10.1002/loe2.10008

Alternative uses of Anthropogenic Radionuclides

Useful for medical, scientific, and industrial applications as well as various consumer products.

Radioactive Isotope	Medical Applications
Cobalt-60	*Radiation therapy*
Iodine-131	*Location of brain tumors, monitoring of cardiac, liver, and thyroid activity*
Carbon-14	*Study of metabolism changes for patients with diabetes, gout, and anemia*
Sodium-24	*Studies of blood circulation*
mTechnecium-99	*Heart tissue damage, tumor detection, other medical diagnoses*
	Industrial/Scientific Applications
Americium-241	*Determination of oil wells, uniform steel and paper thickness*
Iridium-192	*Boiler and aircraft integrity*
Uranium-235	*Nuclear power*
Californium-252	*Soil moisture*
	Consumer Products
Americium-241	*Smoke Detectors*
Hydrogen-3 (Tritium)	*Watches*

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Major sources of *known* Anthropogenic Radionuclides to the marine environment

1) Nuclear weapons - atmospheric testing

1950's-1960's most above ground tests in the Pacific Ocean, the USA and the former USSR.

≈ 200,000 PBq total; ≈ 1,000 PBq ^{137}Cs of which *600 PBq to oceans.*

2) Planned releases from nuclear fuel reprocessing plants

Examples: Sellafield (UK) and Cap de la Hague (France); ≈45 PBq total; *≈ 40 PBq to oceans* ^{137}Cs (from 1950-2010, peaking in 1975).

1 PBq = peta-Becquerel = one million billion Bq = 10^{15} Bq

IAEA (1999), IAEA (2001), and Hu et al. (2010)

doi:10.1002/loe2.10008

Major sources of *known* Anthropogenic Radionuclides to the marine environment

3) Accidents related to nuclear power/production

- Chernobyl (April 26, 1986) ≈ 5000 PBq total;
  ≈ 85 PBq ^{137}Cs of which *15-20 PBq to oceans.*
- Fukushima (March 11, 2011) ≈ 500 PBq total;
  ≈ 20-40 PBq ^{137}Cs of which *15-20 PBq to oceans.*

4) Other main accidents with known releases in the oceans

- Satellite with nuclear power supply: SNAP-9A (1964)
  ≈ 0.630 PBq ^{238}Pu mainly in Southern Hemisphere
- Undetonated weapons in Palomares, Spain (6 Jan 1966)
  ≈ 1.4 PBq Pu isotopes
- Undetonated weapons in Thule, Greenland (21 Jan 1968)
  ≈ 0.03 PBq Pu isotopes

1 PBq = peta-Becquerel = one million billion Bq = 10^{15} Bq

IAEA (1999), IAEA (2001), and Hu et al. (2010)

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Major *potential (not well constrained)* sources of Anthropogenic Radionuclides to the marine environment

1) Disposal at sea of radioactive waste (wastes from nuclear industry, medical and scientific centers)

- Known nuclear waste dumping to ocean (1946-1993)

  ≈ *85 PBq total*

2) Accidents and losses at sea (nuclear powered submarines or vessels, undetonated nuclear weapons, nuclear powered lighthouses, sealed sources, etc.)

≈ *1 to 2000 PBq* (e.g., Kursk submarine reactors)

≈ *45 PBq* Other categories

1 PBq = peta-Becquerel = one million billion Bq = 10^{15} Bq

IAEA (1999), IAEA (2001), and Hu et al. (2010)

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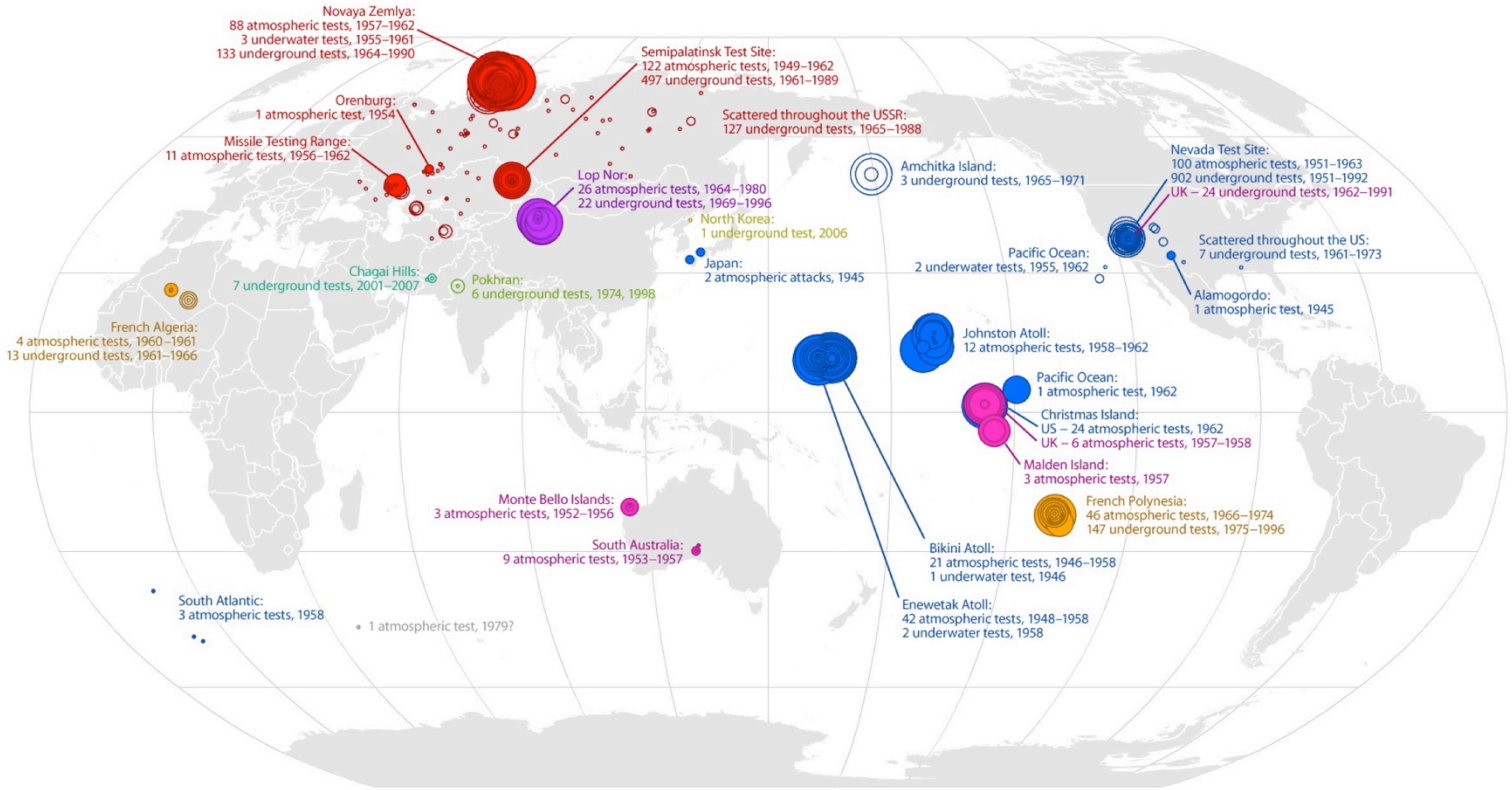
Nuclear Weapons Testing 1945 - 2007



Country:	Year of first detonation:	Number of detonations: atmospheric	underground	underwater
United States	1945	206	912	5
USSR	1949	223	756	3
United Kingdom	1952	21	24	
France	1960	50	160	
China	1964	22	26	
Israel?	1967 ?			
India	1974		6	
South Africa?	1979 ?	1 ?		
Pakistan	1998		7	
North Korea	2006		1	

not all data is official, and some locations are approximate. data source: http://www.johnstonsarchive.net/nuclear/tests

Each explosion is represented by a circle.
Many of these circles overlap.

Filled circles are atmospheric detonations

Hollow circles are underground or underwater tests

The size of each circle represents the yield of the blast.
The scale is not linear:

more than 20 megatons
2.5–5.1 megatons
160–320 kilotons
less than 15 kilotons

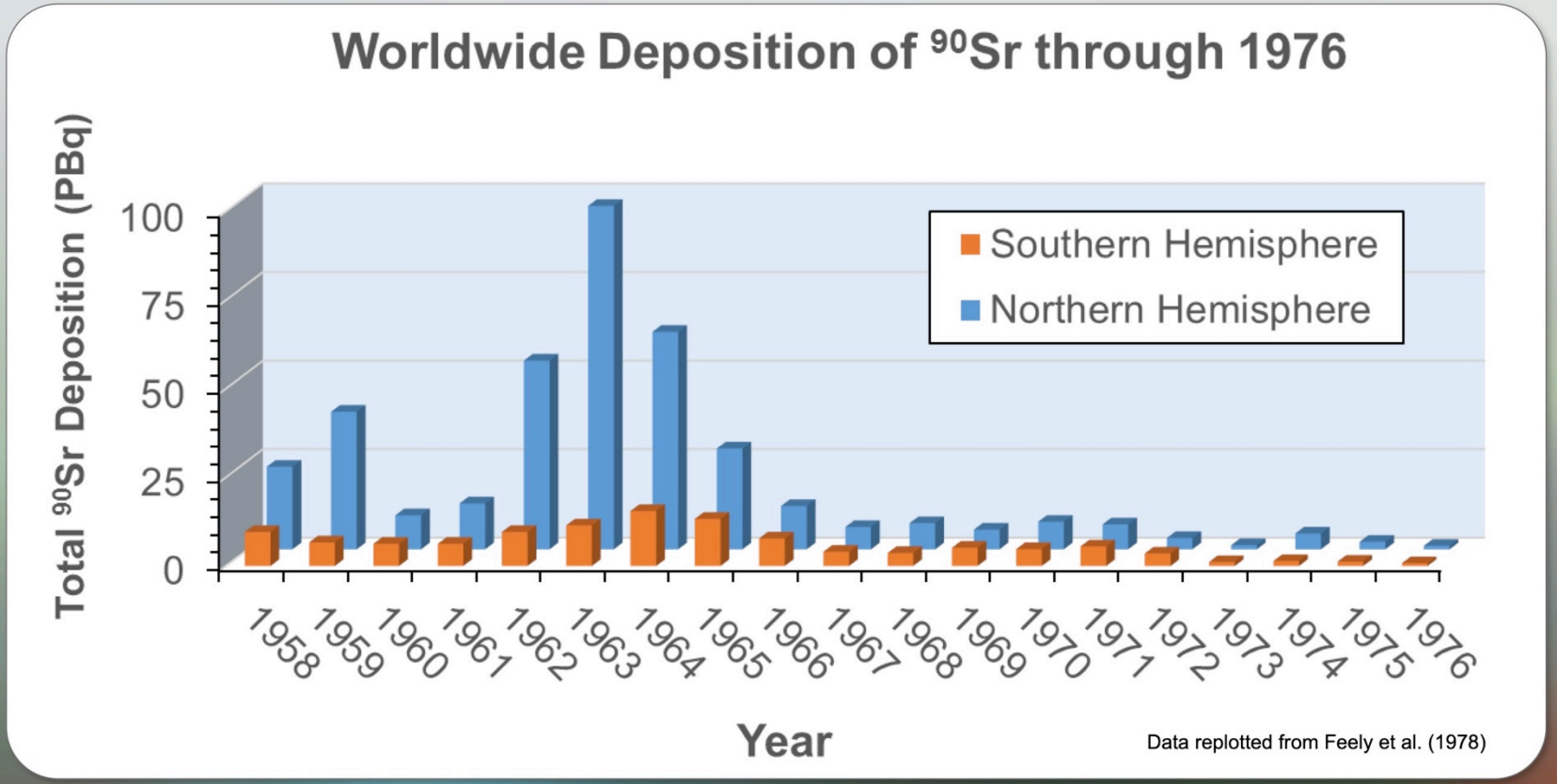
Map created by radicalcartography.net

doi:10.1002/loe2.10008

Atmospheric Nuclear Weapons Testing 1945 - 1980

Atmospheric nuclear weapons testing led to temporal and spatial variability in *global* radioactive fallout by atmospheric deposition.

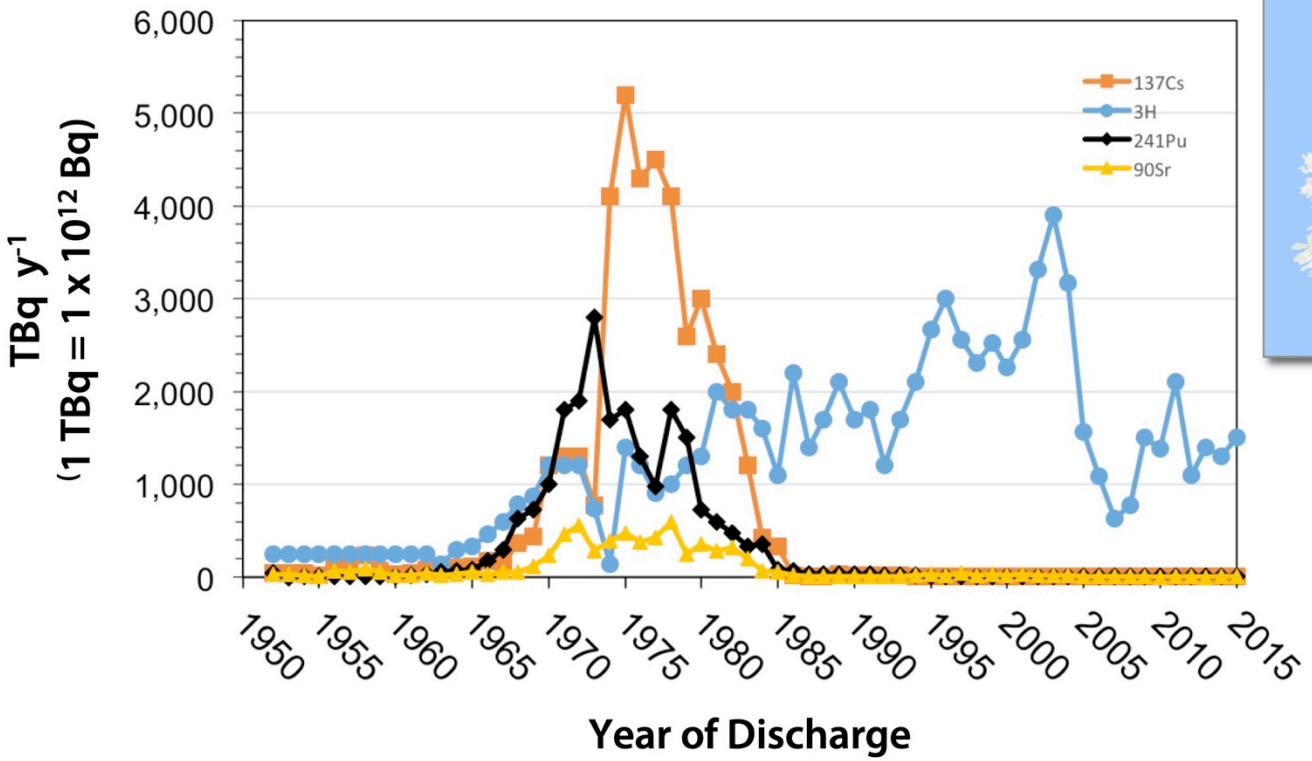
12% of the fallout deposited as **"close-in" fallout** near the test site
10% percent has ended up around the same latitude as the test site- "**intermediate**"
78% percent is **"global" fallout**, most of which ended up in the ***same*** hemisphere



doi:10.1002/loe2.10008

Planned Releases

Discharges of major beta gamma emitters in liquid effluent from Sellafield: 1952-2015



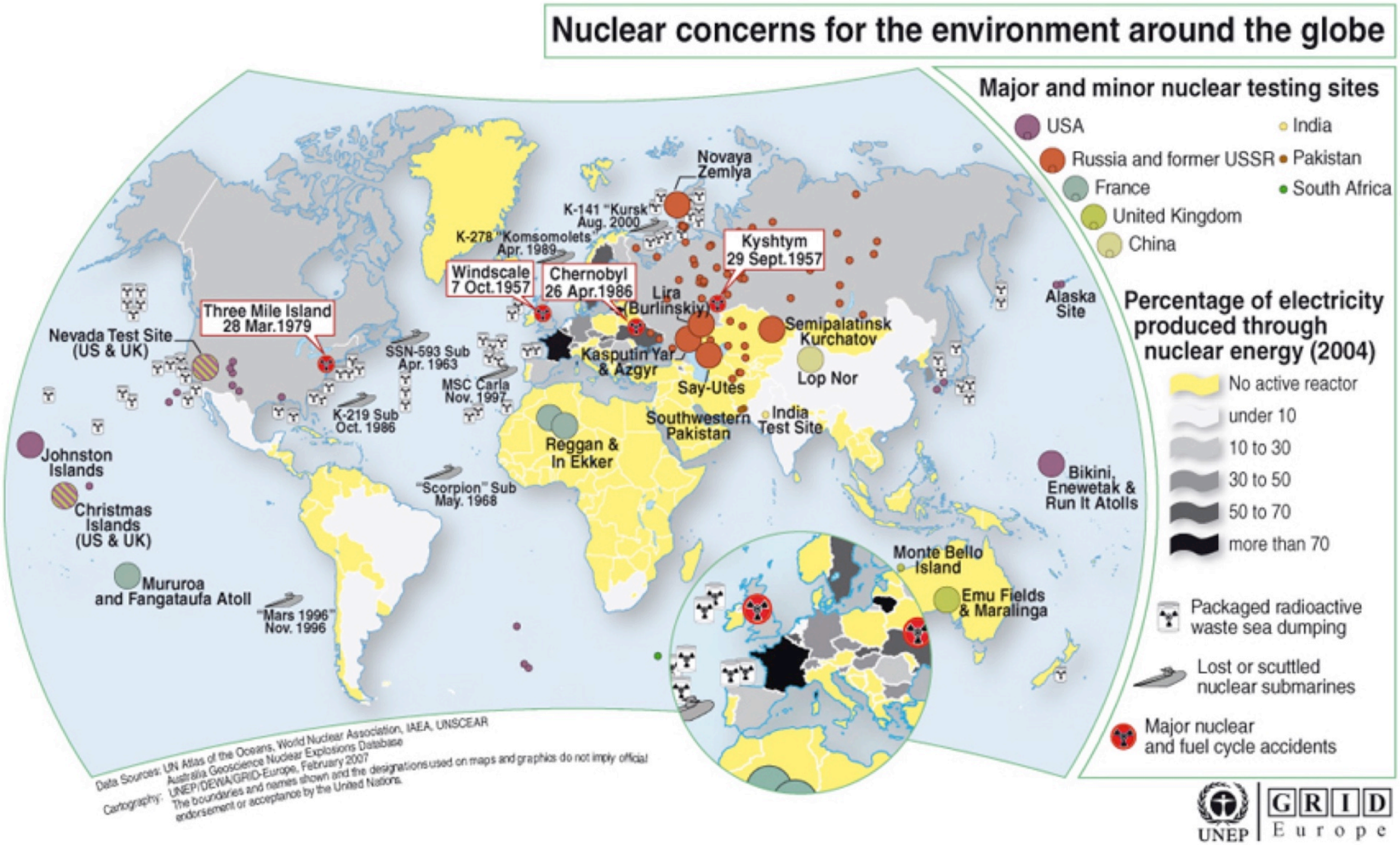
1 TBq = tera-Becquerel = 10^{12} Bq

Adapted from Gray et al. (2005)

Source from nuclear fuel reprocessing discharges to Irish sea from Sellafield

doi:10.1002/loe2.10008

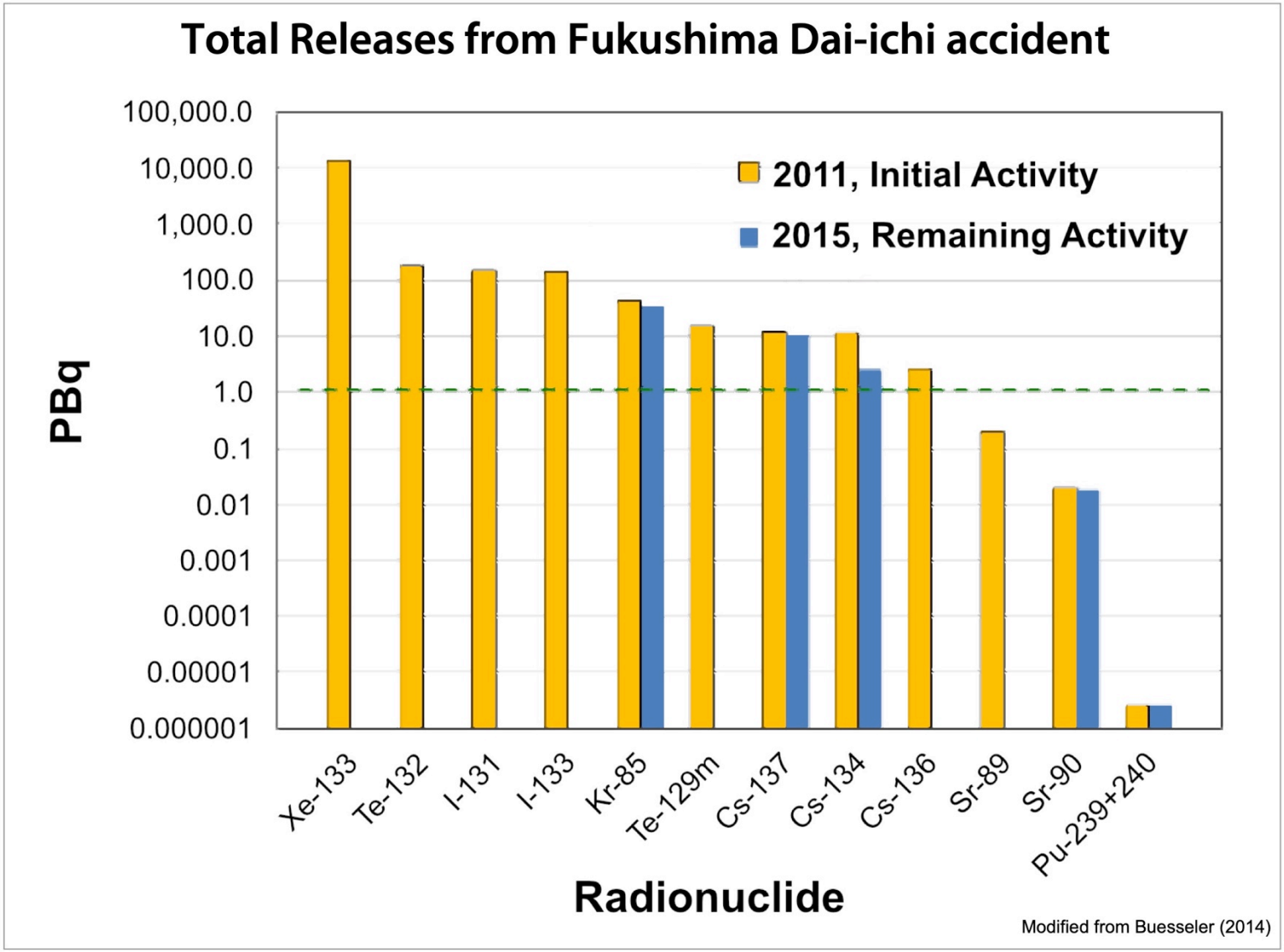
Nuclear Power Plant Accidents (not comprehensive)



http://www.grid.unep.ch/products/4_Maps/nuclear_concernsb.jpg

doi:10.1002/loe2.10008

Why so much attention on cesium?



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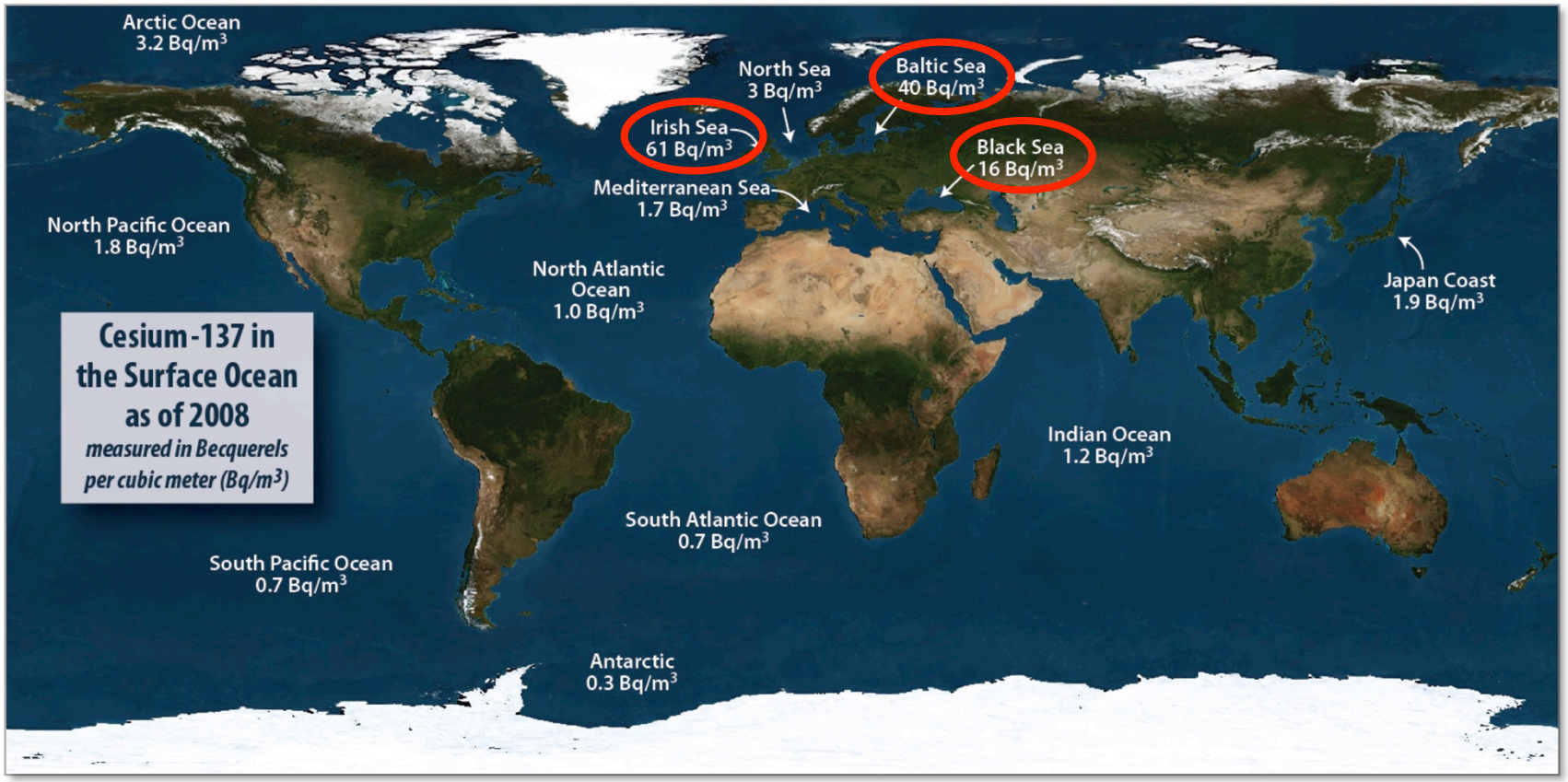
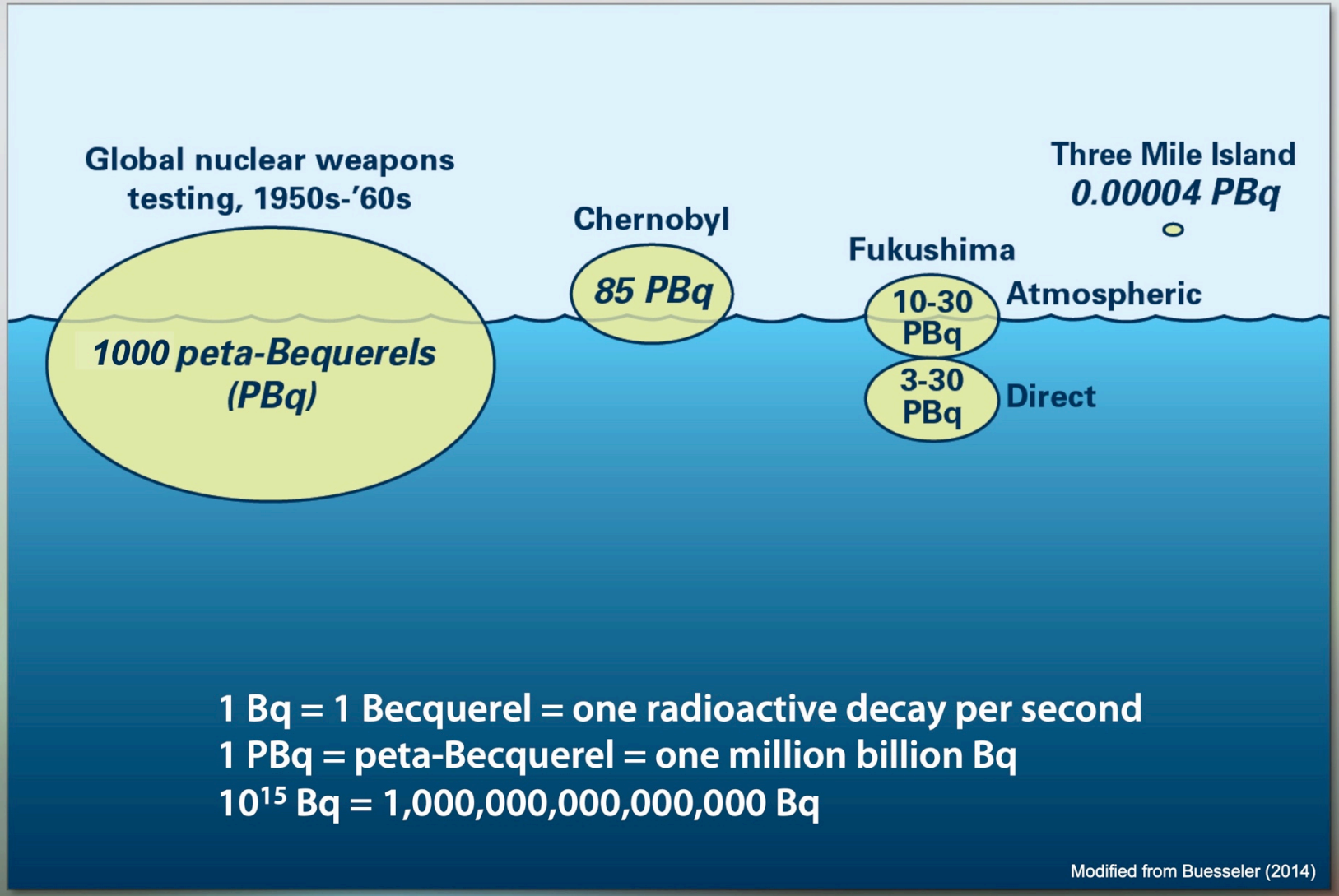
Uneven distributions of radionuclides in the ocean



Modified from Buesseler (2014)

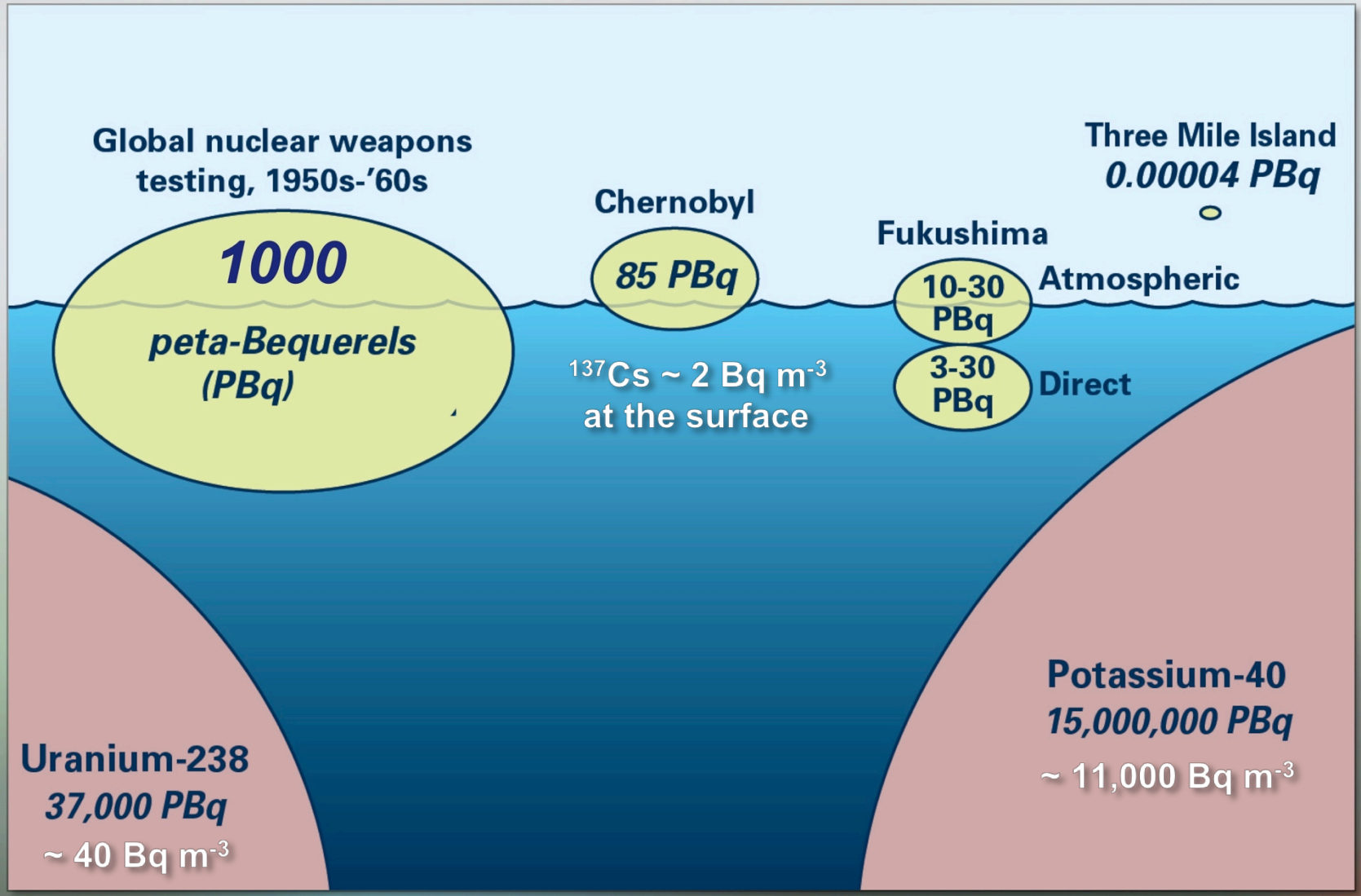
doi:10.1002/loe2.10008

Total cesium-137: What are the largest sources?



doi:10.1002/loe2.10008

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



Updated from Buesseler (2014) based on Buesseler et al. (2017)

doi:10.1002/loe2.10008

Tracer applications of Anthropogenic Radionuclides in the ocean

1

Water/Atmospheric mixing rates (i.e., ocean, groundwater etc.):

^{90}Sr ^{14}C ^{3}H ^{129}I ^{36}Cl $^{134,137}Cs$

2

Particle scavenging & transport rates in the water column:

^{241}Am ^{144}Ce ^{106}Ru $^{239,240}Pu$

3

Biological uptake: ^{14}C

(see slide 16)

4

Sediment accumulation & mixing:

^{134}Cs ^{137}Cs ^{241}Am $^{239,240}Pu$

5

Age dating:

^{14}C ^{134}Cs ^{137}Cs $^{239,240}Pu$

doi:10.1002/loe2.10008

Case studies

Ocean Circulation

Anthropogenic radionuclides derived from (1) atmospheric fallout (weapons tests), (2) point sources from nuclear facilities (Sellafield & La Hague) and (3) nuclear accidents (Fukushima) used to trace ocean circulation.

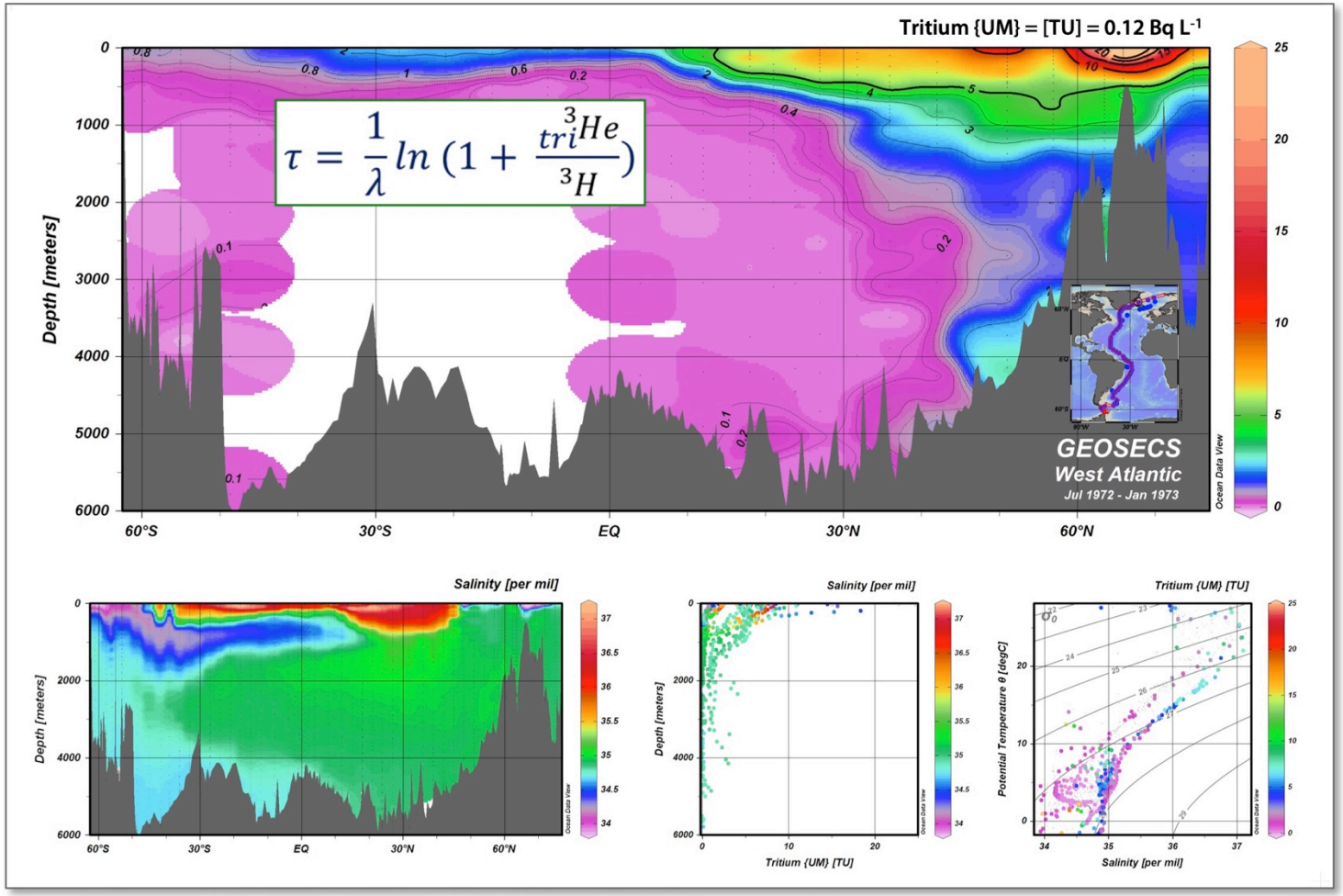
Particle Transport and Burial

Anthropogenic radionuclides that are particle reactive may be used to determine particle transport pathways, as well as sediment geochronologies.

doi:10.1002/loe2.10008

1) Circulation from atmospheric fallout - weapons tests: ^3H ($t_{½}$ = 12.3 y)

Measuring how much ^3H has penetrated into the ocean since nuclear weapons testing allows for an estimate of water mass age and circulation patterns.

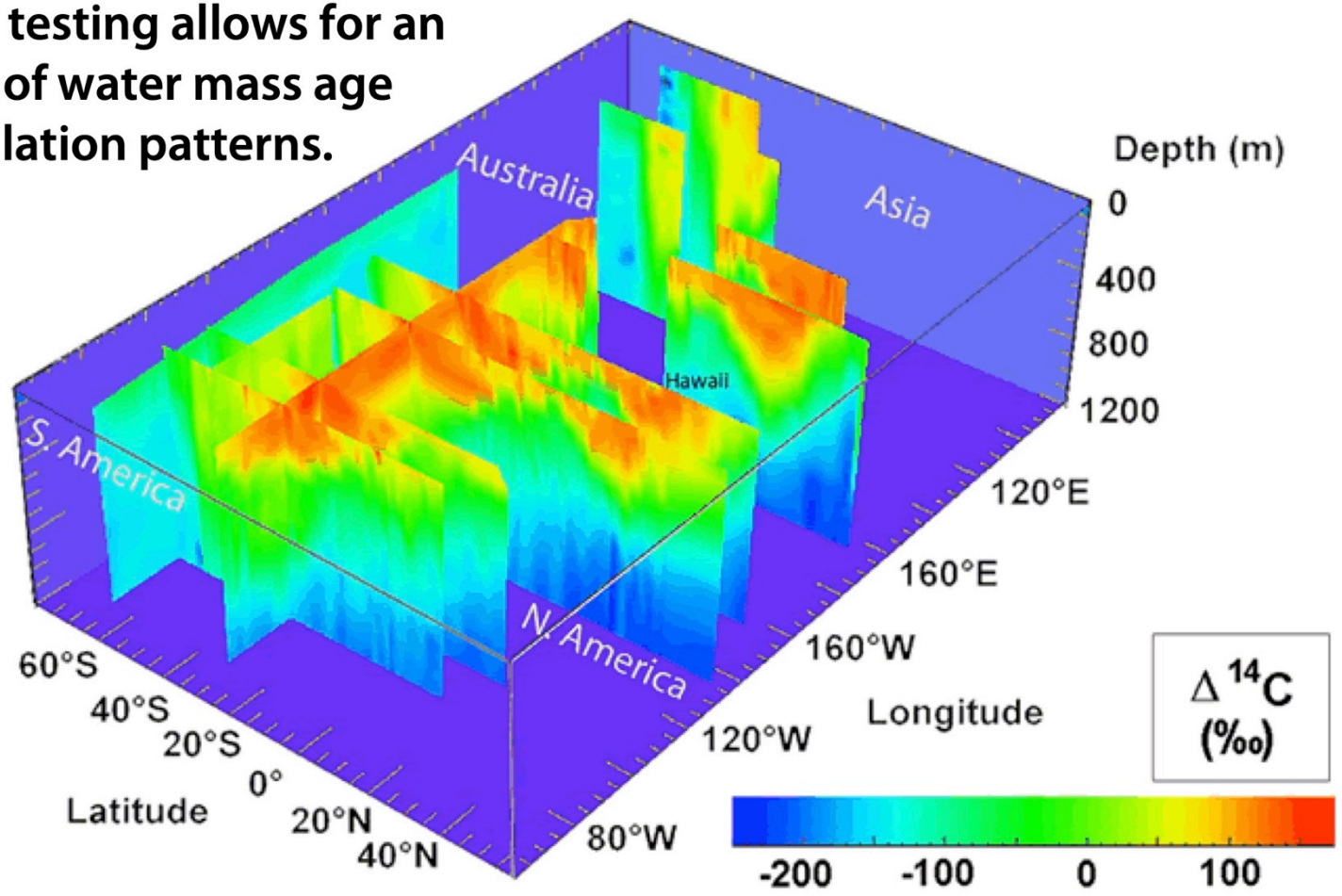


Claudia Benitez-Nelson

doi:10.1002/loe2.10008

1) Circulation from atmospheric fallout - weapons tests: ^{14}C ($t_{½}$ = 5730 y)

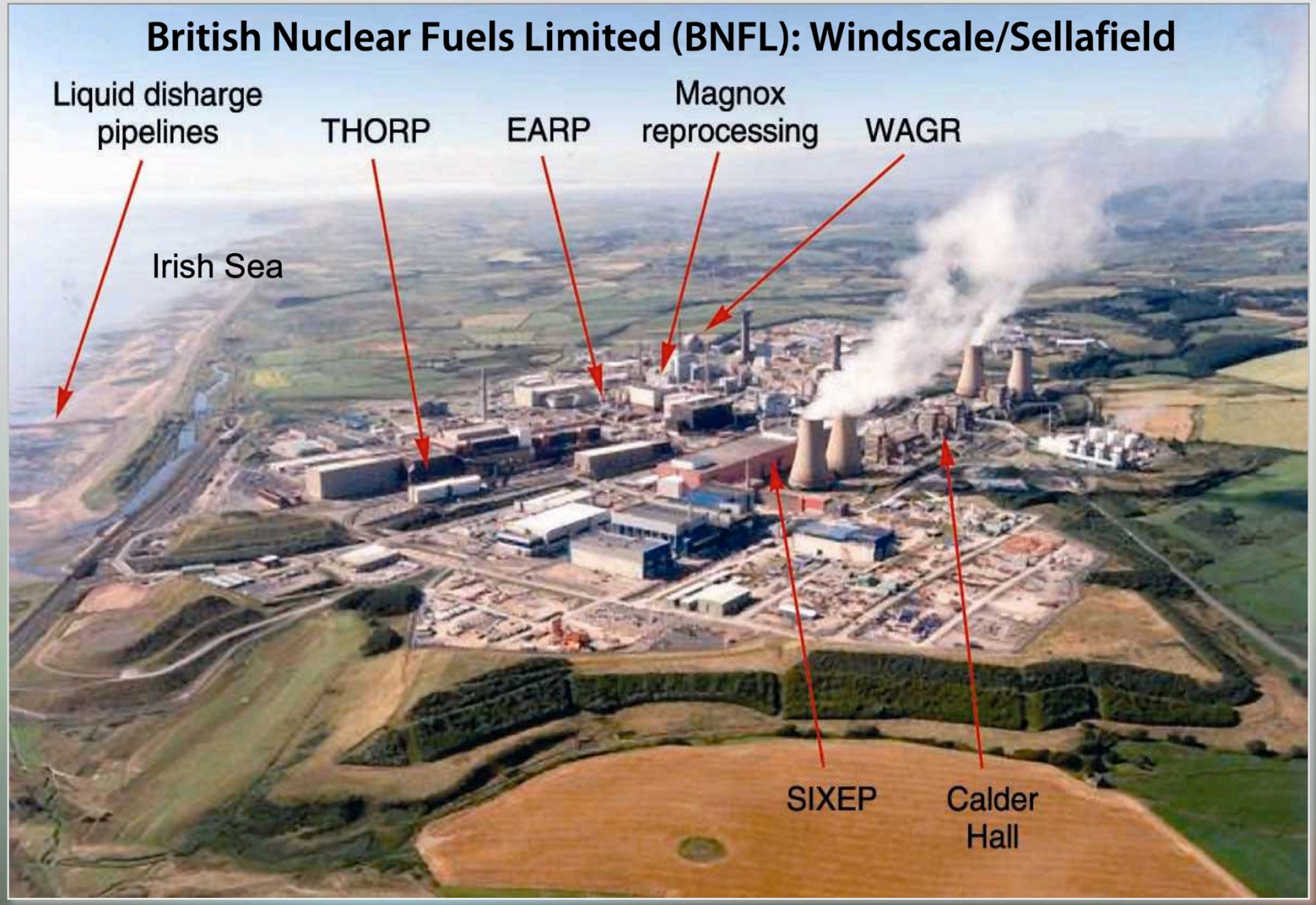
Measuring how much ^{14}C has penetrated into the ocean since weapons testing allows for an estimate of water mass age and circulation patterns.



Povinec et al. (2009)

doi:10.1002/loe2.10008

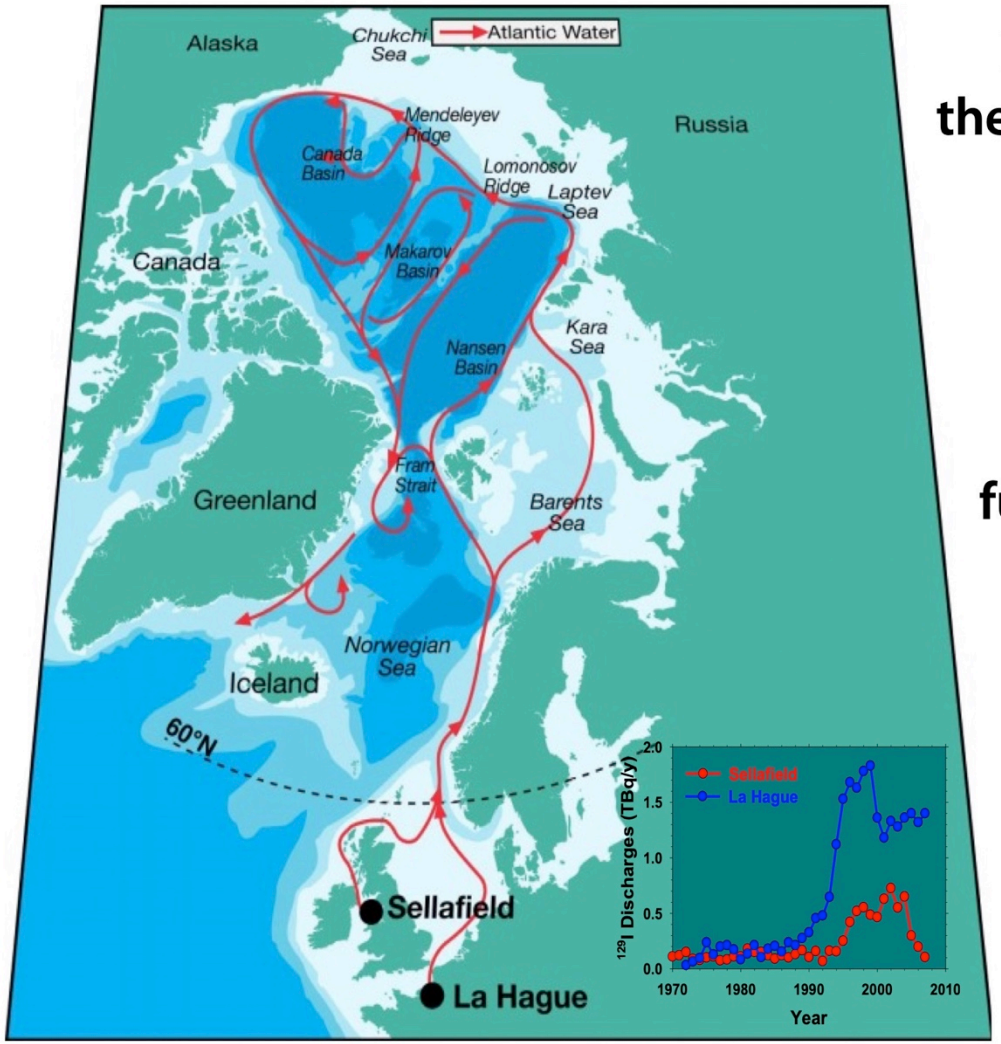
2) Circulation from point sources - nuclear facilities



doi:10.1002/loe2.10008

2) Circulation from point sources - nuclear facilities

^{129}I ($t_{½}$ = 1.6 x 10^7 y)



Pathways (red arrows) for the transport of tracers from Sellafield and La Hague nuclear facilities through the Arctic Ocean.

Inset shows ^{129}I input functions from the nuclear fuel reprocessing plants.

● ^{129}I ($t_{½}$ = 1.6 x 10^7 y)

Karcher et al. (2012)

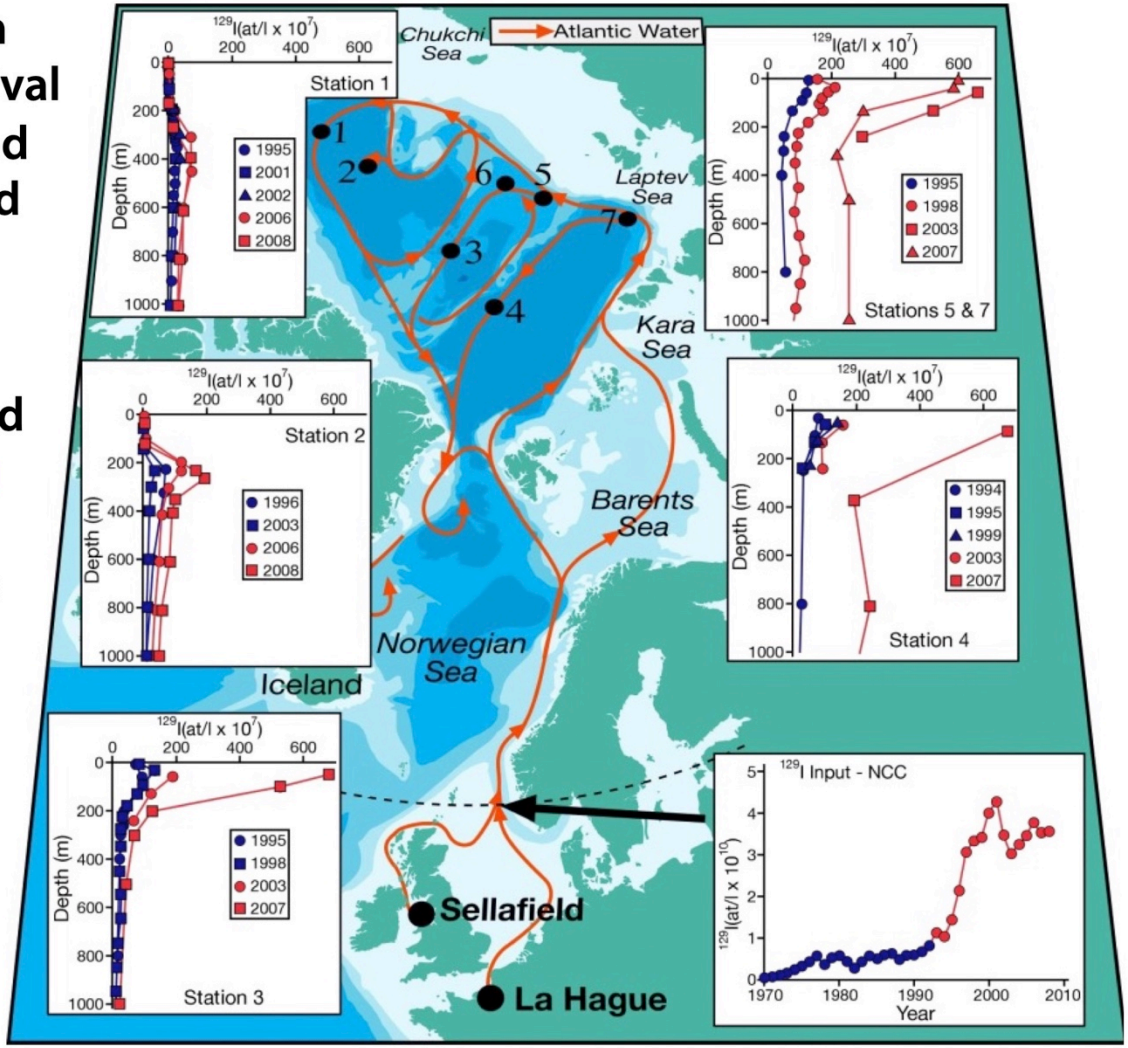
doi:10.1002/loe2.10008

2) Circulation from point sources - nuclear facilities

^{129}I ($t_{1/2}$ = 1.6 x 10^7 y)

^{129}I water-depth profiles in Arctic Ocean show the arrival of the large pulse produced in the 1990s from Sellafield and La Hague.

The arrival of the lower, pre-1993 signal is indicated at each station by blue ^{129}I profiles. The arrival of the higher, post-1993 signal is indicated by red profiles and provides a constraint on transit times (e.g., 10 y for transport from 60°N to St. 4 at the North Pole).



Smith et al. (2011)

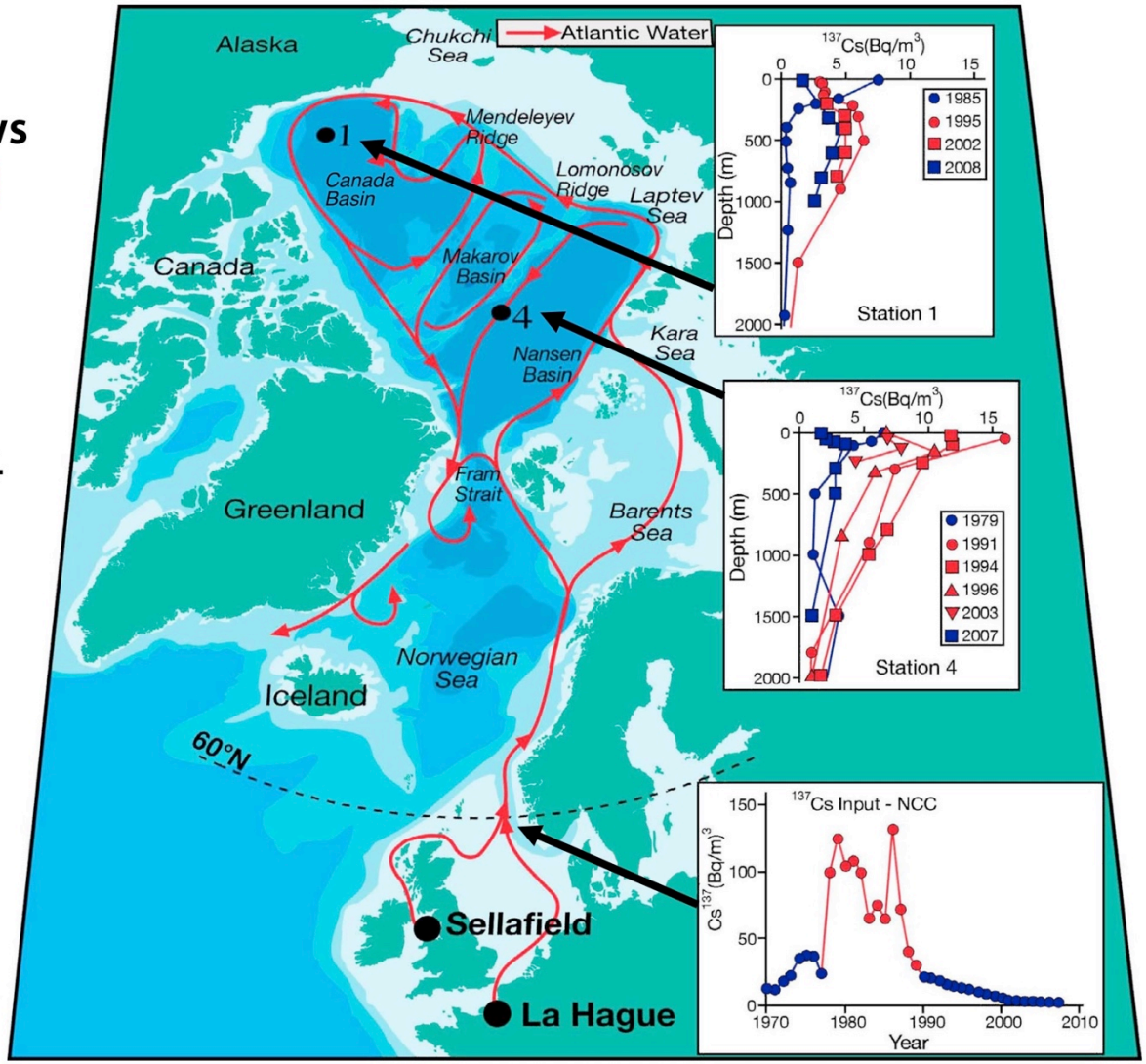
doi:10.1002/loe2.10008

2) Circulation from point sources - nuclear facilities

^{137}Cs ($t_{1/2}$ = 30.2 y)

^{137}Cs input function *(lower right inset)* **shows maximum values (red symbols) between 1978 and 1990.**

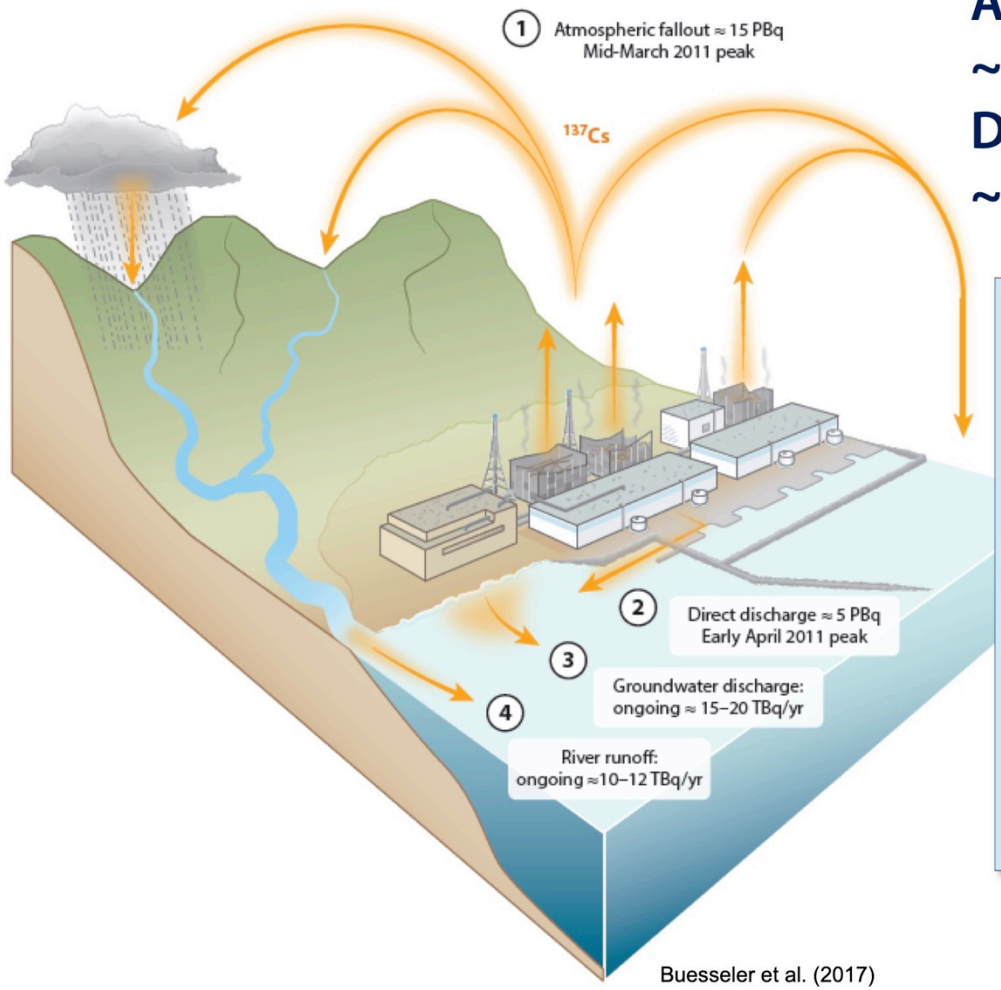
These elevated ^{137}Cs levels appear at Sta. 4 (red lines) between 1991 and 2003 which also indicates a 10 y transit time to Sta. 4 from 60°N.



Smith et al. (2011)

doi:10.1002/loe2.10008

3) Circulation from nuclear accidents: Fukushima, March 2011

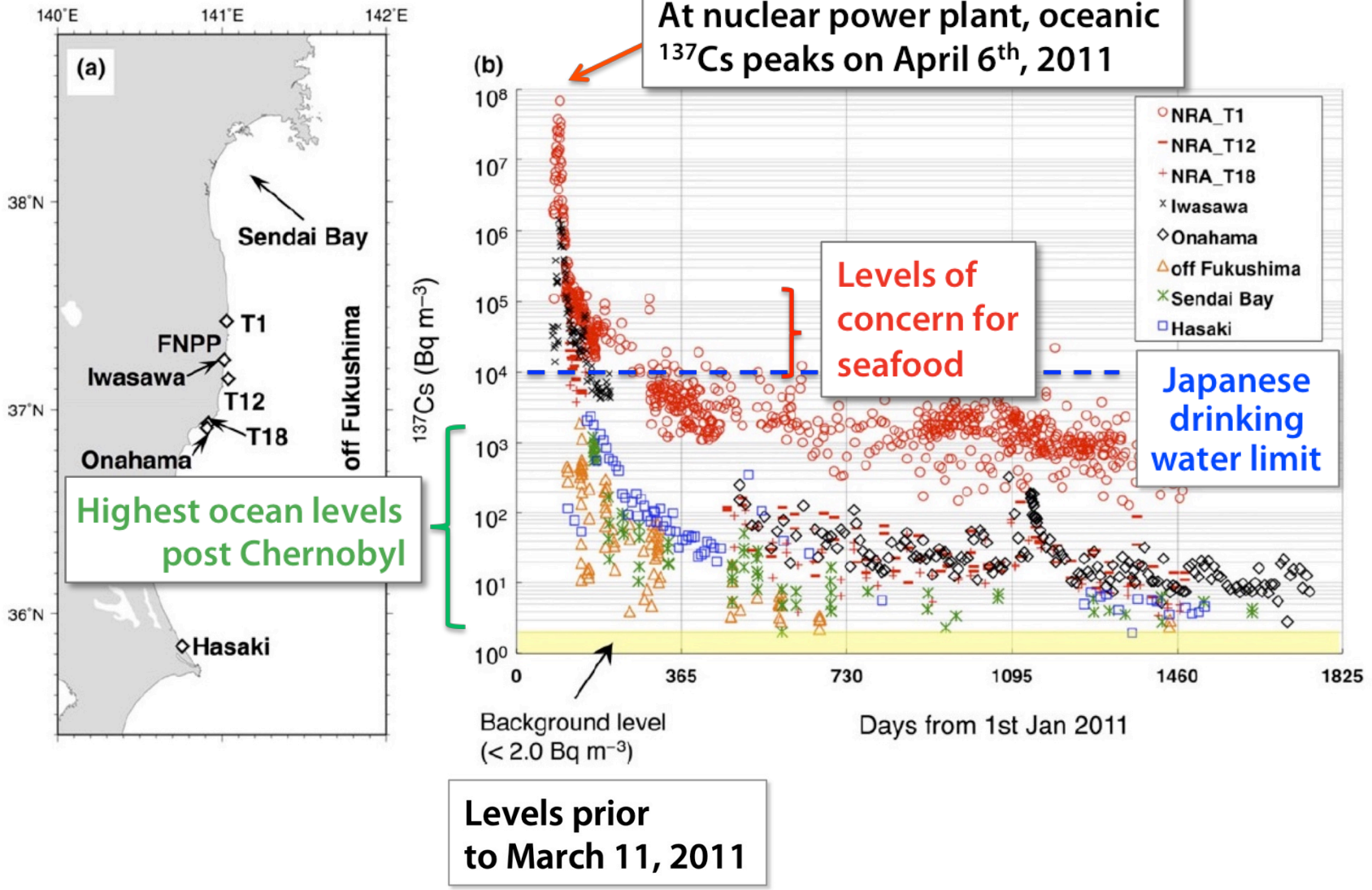


Buesseler et al. (2017)

Atmospheric Fallout to ocean:
~ 15 PBq ^{137}Cs
Direct Discharge to ocean:
~ 5 PBq ^{137}Cs

doi:10.1002/loe2.10008

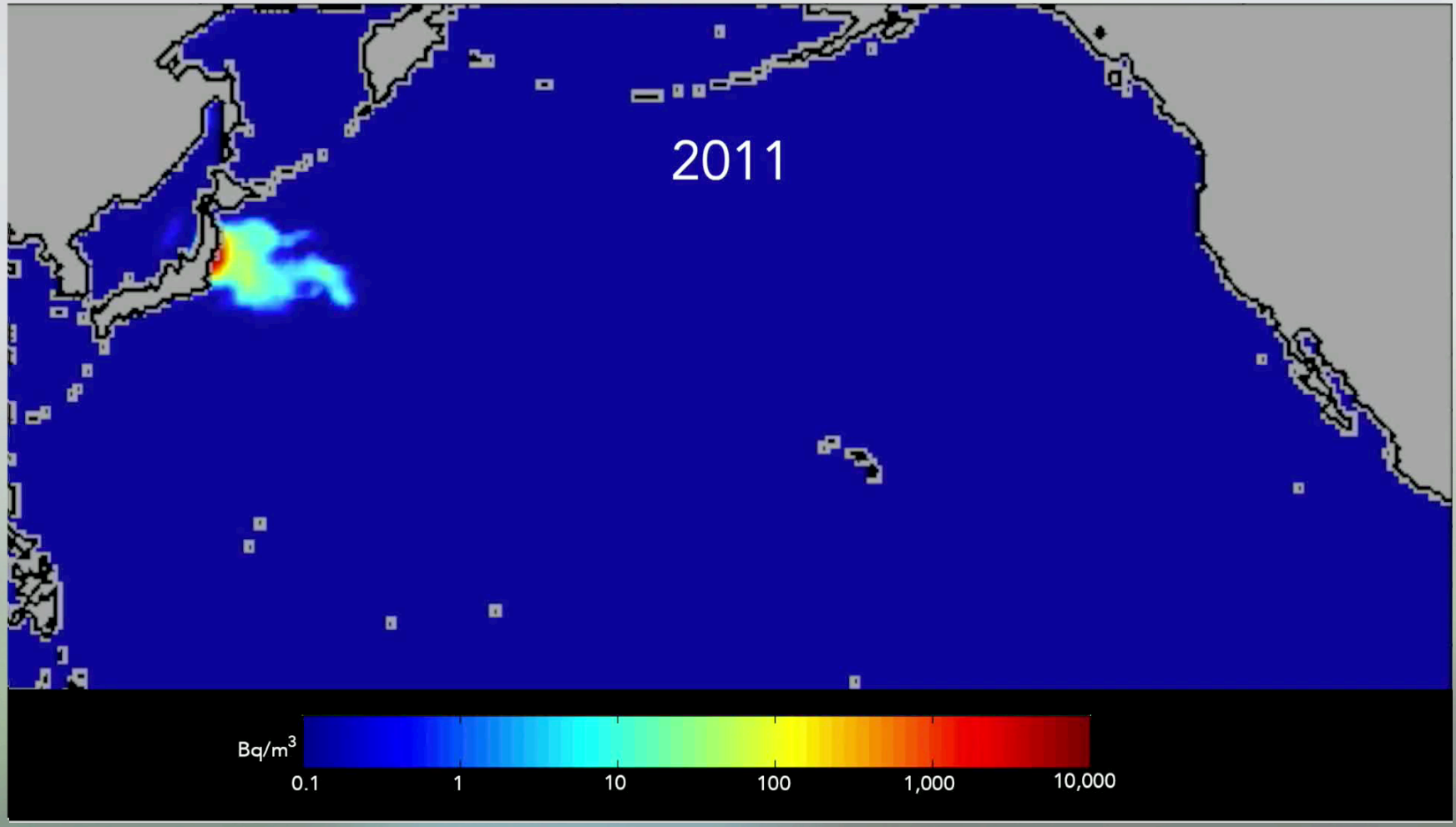
History of ^{137}Cs activity in ocean immediately off Fukushima



Kaeriyama (2016)

doi:10.1002/loe2.10008

3) Circulation from nuclear accidents: Fukushima, March 2011



Rossi et al., 2013 & 2014

(Hover over image and click to play video)

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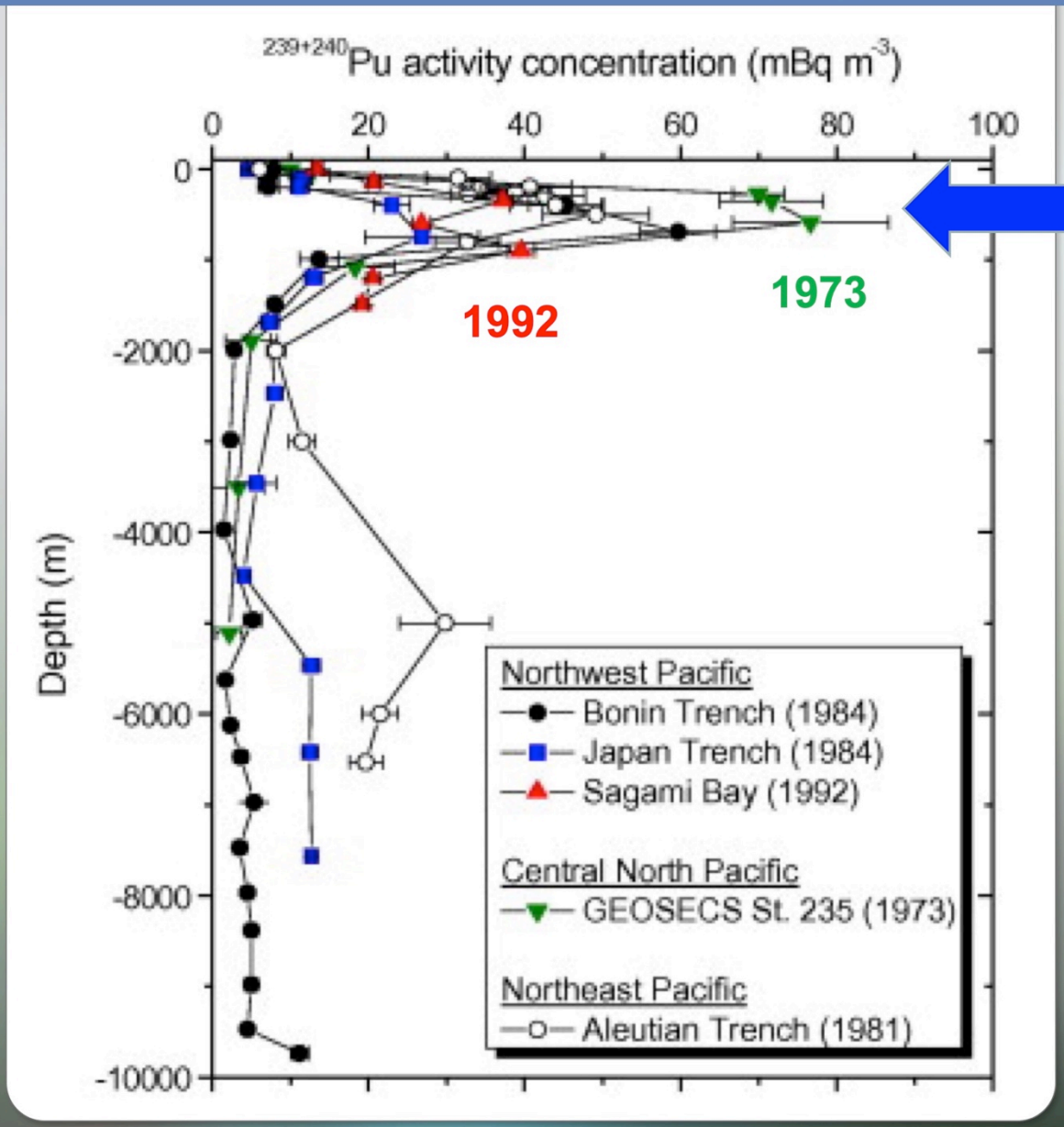
4) Scavenging Tracers



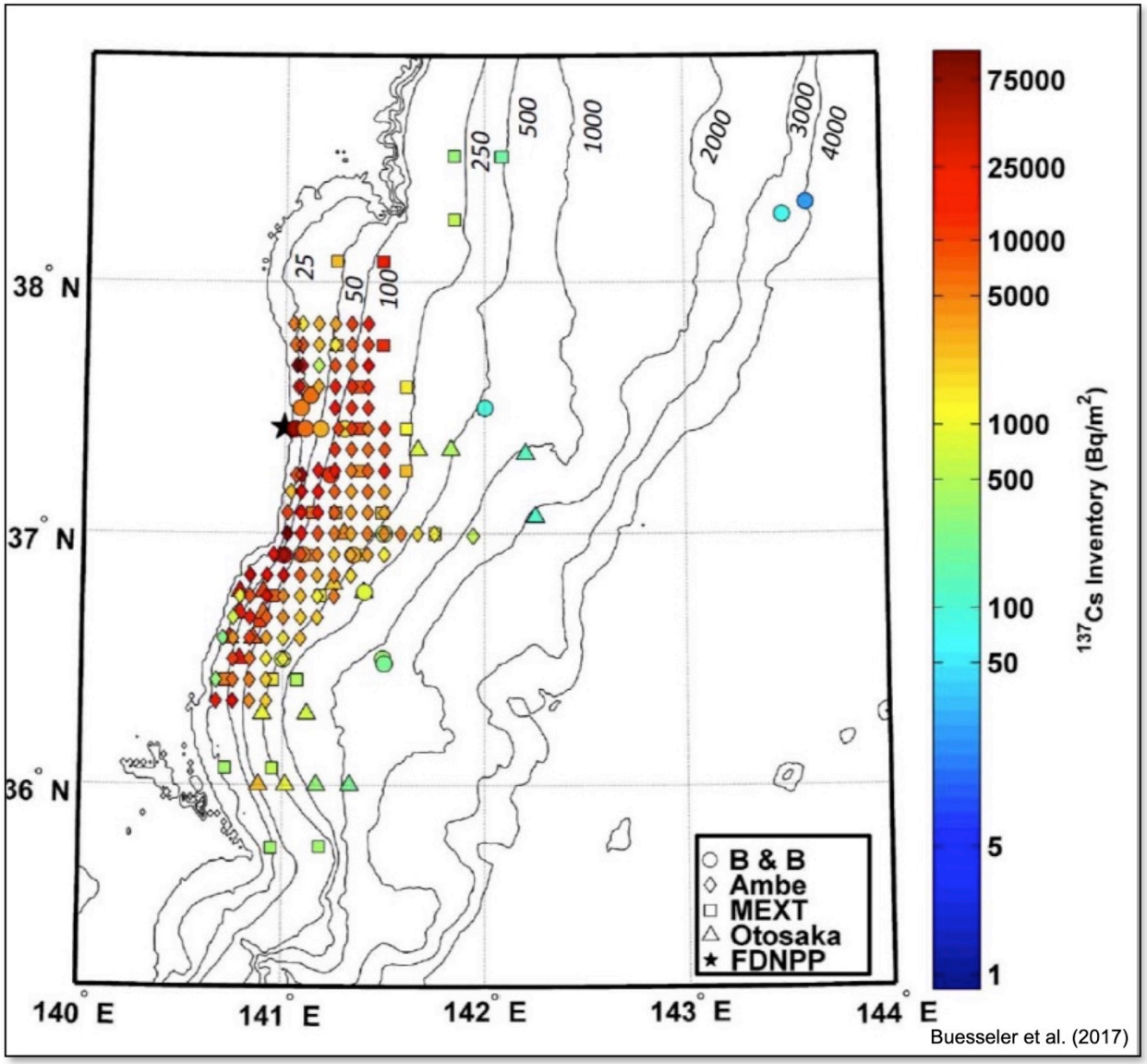
Particle scavenging leads to decrease in atmospherically derived $^{239+240}$Pu over time.

Subsurface peak at depth is hypothesized to be due to vertical transport followed by subsurface biological remineralization.

Lindahl et al. (2010)

doi:10.1002/loe2.10008

4) Scavenging Tracers and Age Dating



Buesseler et al. (2017)

doi:10.1002/loe2.10008

5) Age Dating: Event Markers

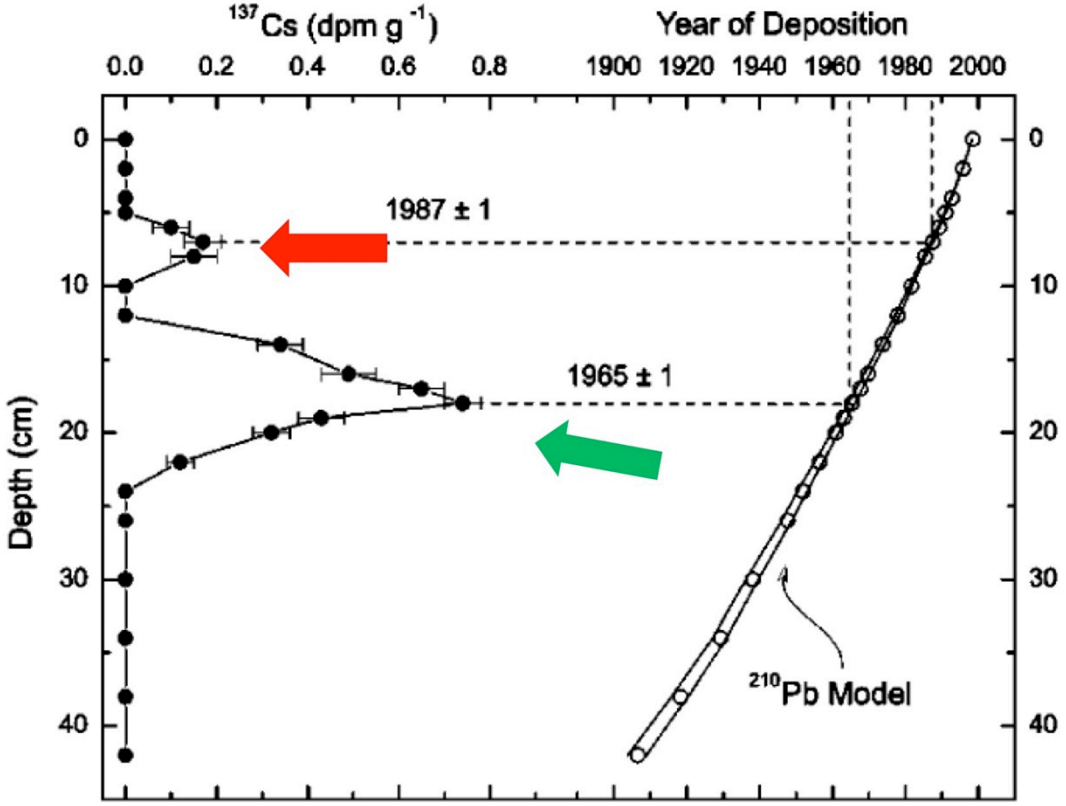


FIGURE 2. ^{137}Cs activity (closed circles) and year of deposition (calculated using unsupported ^{210}Pb, open circles) plotted versus sediment depth. Samples corresponds to 0.5-cm-thick sediment slices. Years of deposition are shown with an envelope of uncertainty ($\pm 1\sigma$).

Lima et al. (2005)

Chernobyl Accident

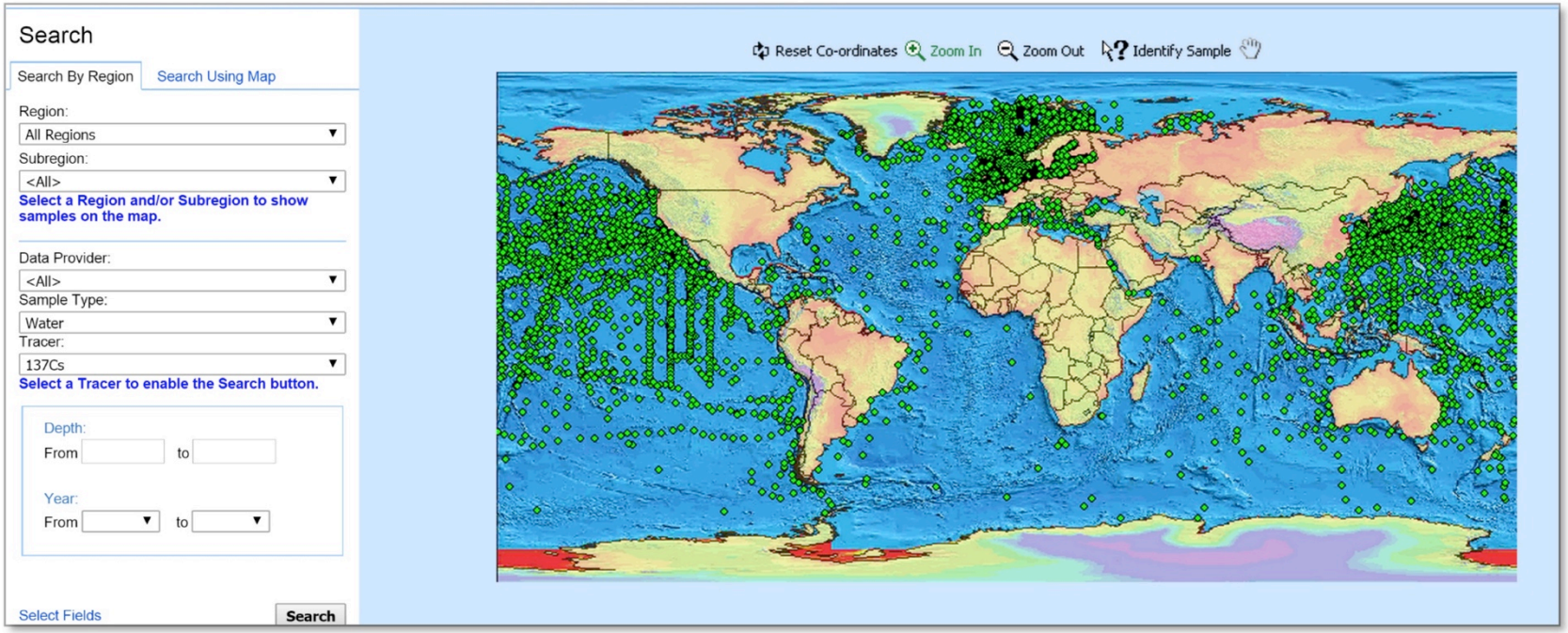
Peak in Atmospheric Nuclear Weapons testing

Sediment core was collected from the Pettaquamscutt River Basin, Rhode Island, USA

doi:10.1002/loe2.10008

How to find marine radioactivity data?

e.g. IAEA MARiS Database for Anthropogenic Radionuclides



http://maris.iaea.org

http://maris.iaea.org Geotraces: http://www.egeotraces.org/

doi:10.1002/loe2.10008

In summary

Cosmogenic and *anthropogenic radionuclides* are very useful tools as *tracers* to study a *large variety of processes* in the oceans at various *time scales* from days to thousands of years, such as:

- Inputs and outputs (e.g., ^{14}C, ^{137}Cs, ^{3}H, ^{129}I)
- Circulation (e.g., ^{7}Be, ^{14}C, ^{137}Cs, ^{3}H, ^{129}I)
- Transfer of substances (e.g., ^{14}C, $^{239+240}Pu$)
- Sedimentary record (e.g., ^{7}Be, ^{14}C)

doi:10.1002/loe2.10008

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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.

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