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Radioactivity in the Marine Environment: Cosmogenic and Anthropogenic Radionuclides

Claudia Benitez-Nelson, Ken Buesseler, Minhan Dai, Michio Aoyama, Núria Casacuberta, Sabine Charmasson, José Marcus Godoy, Andy Johnson, Vladimir Maderich, Pere Masqué, et al.

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Radioactivity in the Marine Environment

Cosmogenic and Anthropogenic Radionuclides

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Andy Johnson, Vladimir Maderich, Pere Masqué,
Willard Moore, Paul J. Morris, John N. Smith

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 , ^{222}Rn , ^{224}Ra , ^{226}Ra , ^{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: ^3H , ^{14}C , ^7Be

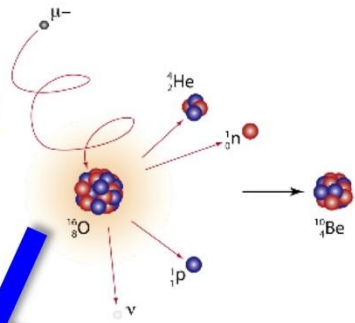
3. **Anthropogenic radionuclides** – continuously being produced by humans.

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

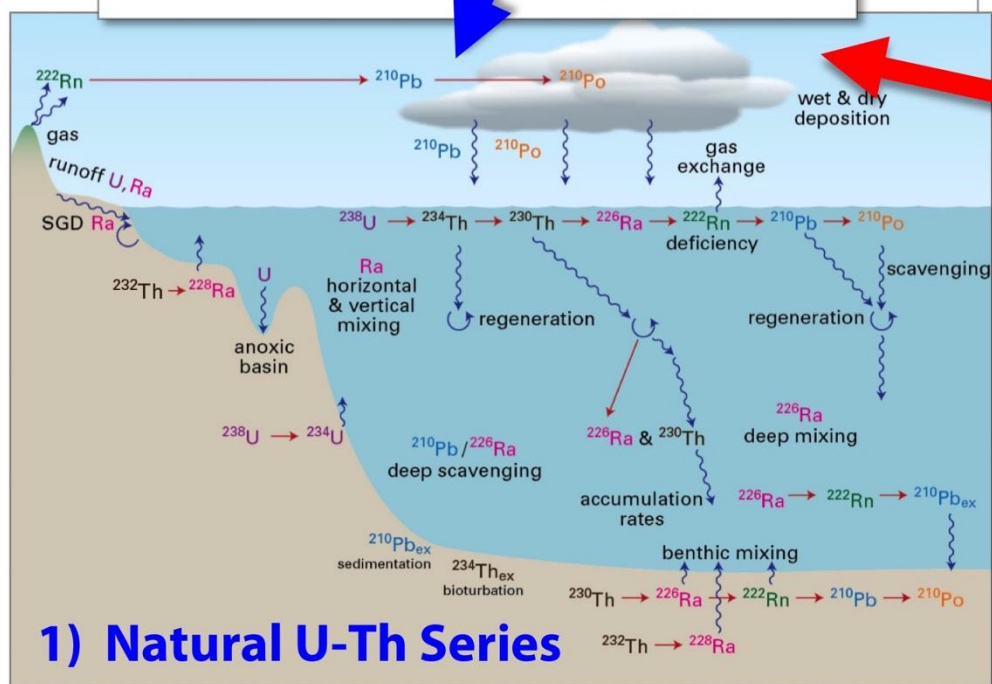
Note: Some radionuclides have both cosmogenic and anthropogenic sources (e.g., ^3H and ^{14}C).

Three main sources of radionuclides to the Marine Environment:

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



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



1) Natural U-Th Series

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 (atoms/cm ² per min) | Integrated in situ oceanic production rate ^b |
|-------------------------------|-------------------------|-----------------------------|-------------------|---|---|
| | | Atmosphere | In ocean water | | |
| 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 years | 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} |

^aBased 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 ¹⁴C, ³⁹Ar, and ⁸¹Kr, for mixing with the mixed layer of the ocean, are taken to be 10, 270, and 170 years, respectively.

^bLal et al. (1988).

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

^dAs above, due to ³⁵Cl(*n*, γ) ³⁶Cl reaction.

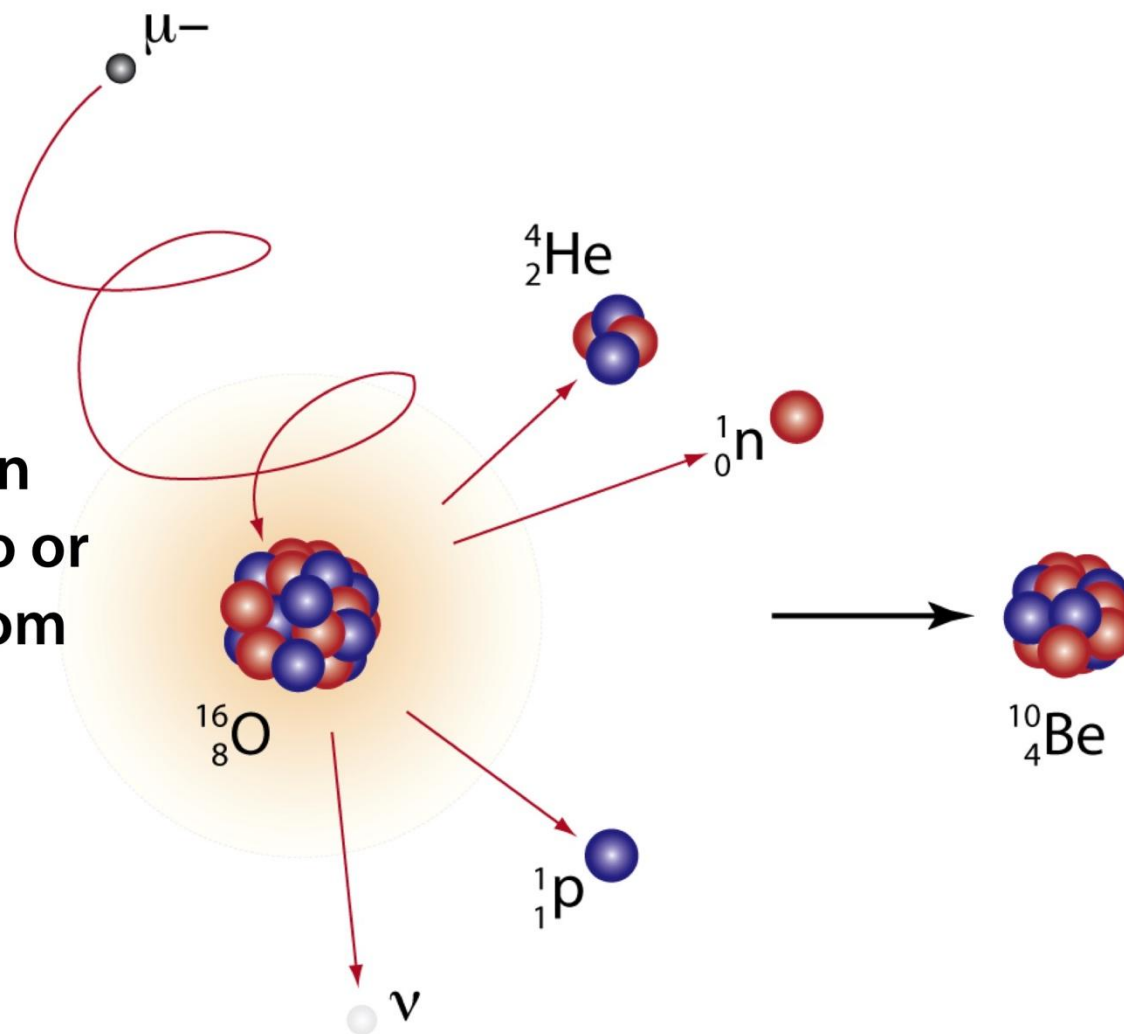
^eThe in situ production of ⁴¹Ca, ⁸¹Kr and ³⁶Cl in the oceans is primarily due to the relevant thermal neutron capture reaction. Note the 50% greater in situ production of ³⁶Cl in the oceans compared to its atmospheric source.

Lal, 1999

Cosmogenic Radionuclides

Sources:

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



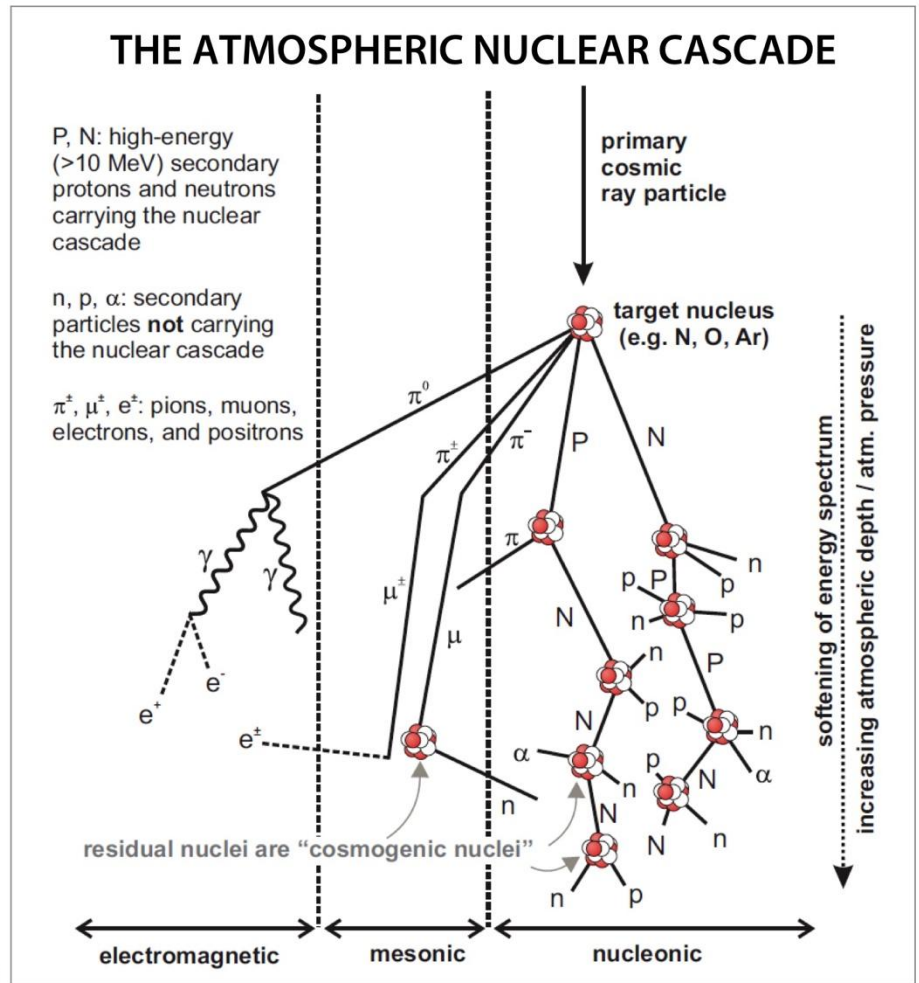
Cosmic Ray Shower

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)

Cosmogenic Radionuclides

Cosmogenic nuclide production varies
with **latitude** in the atmosphere!

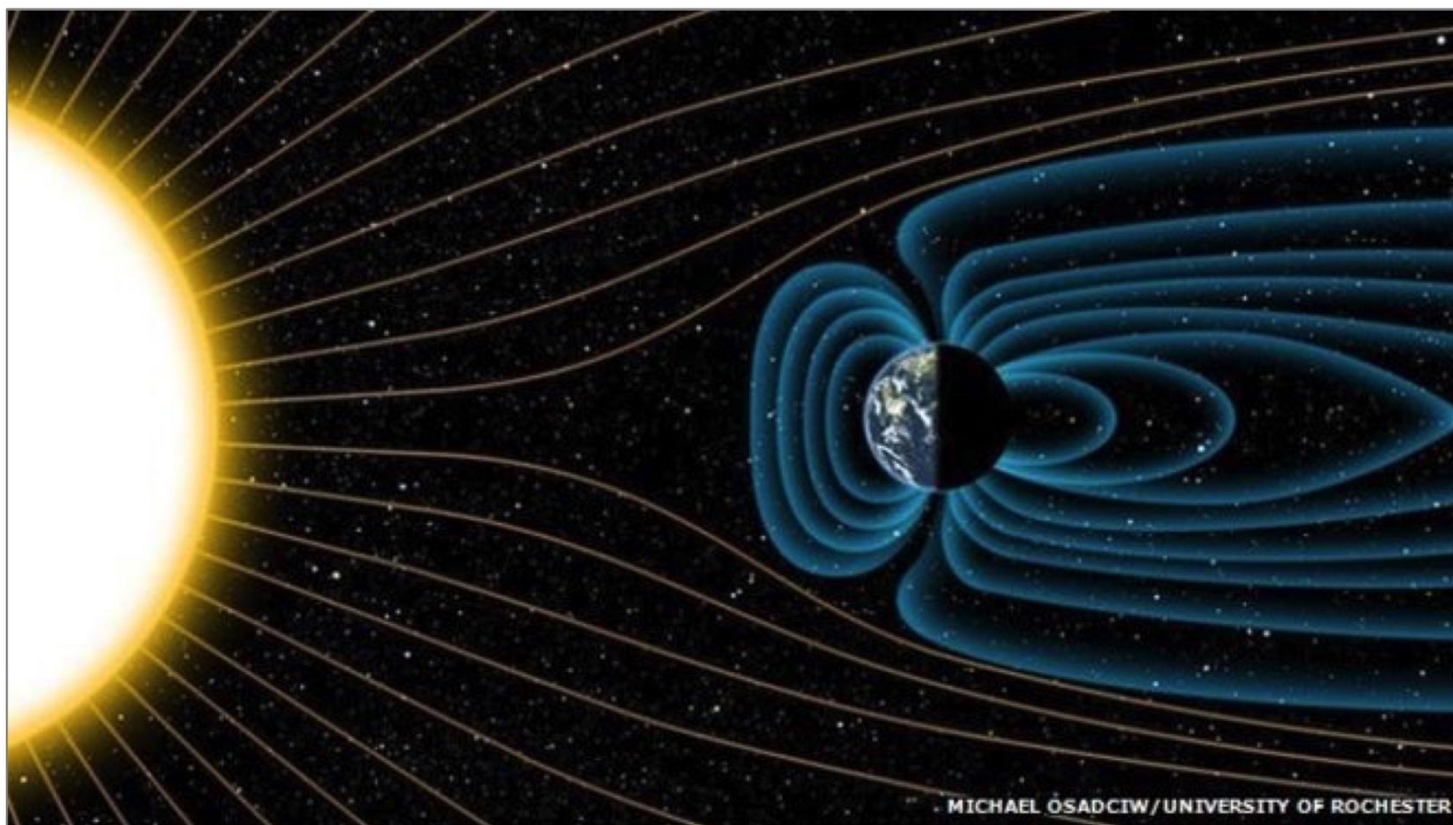


Image of solar winds deflected by Earth's magnetic field

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



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

Photo by Twitter user [@andrew_chester](#) via [lockerz.com](#)

^7Be production in the Atmosphere

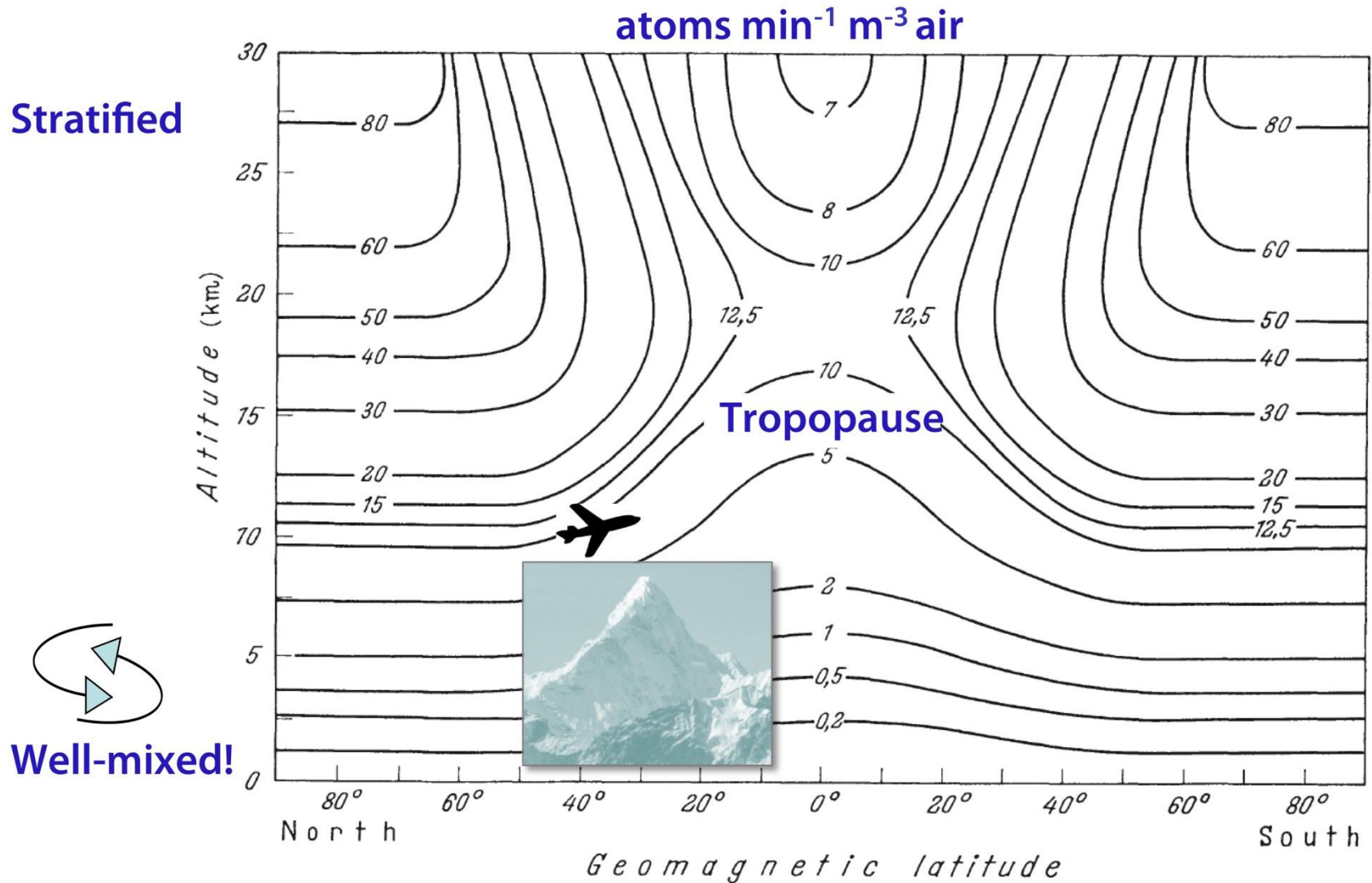
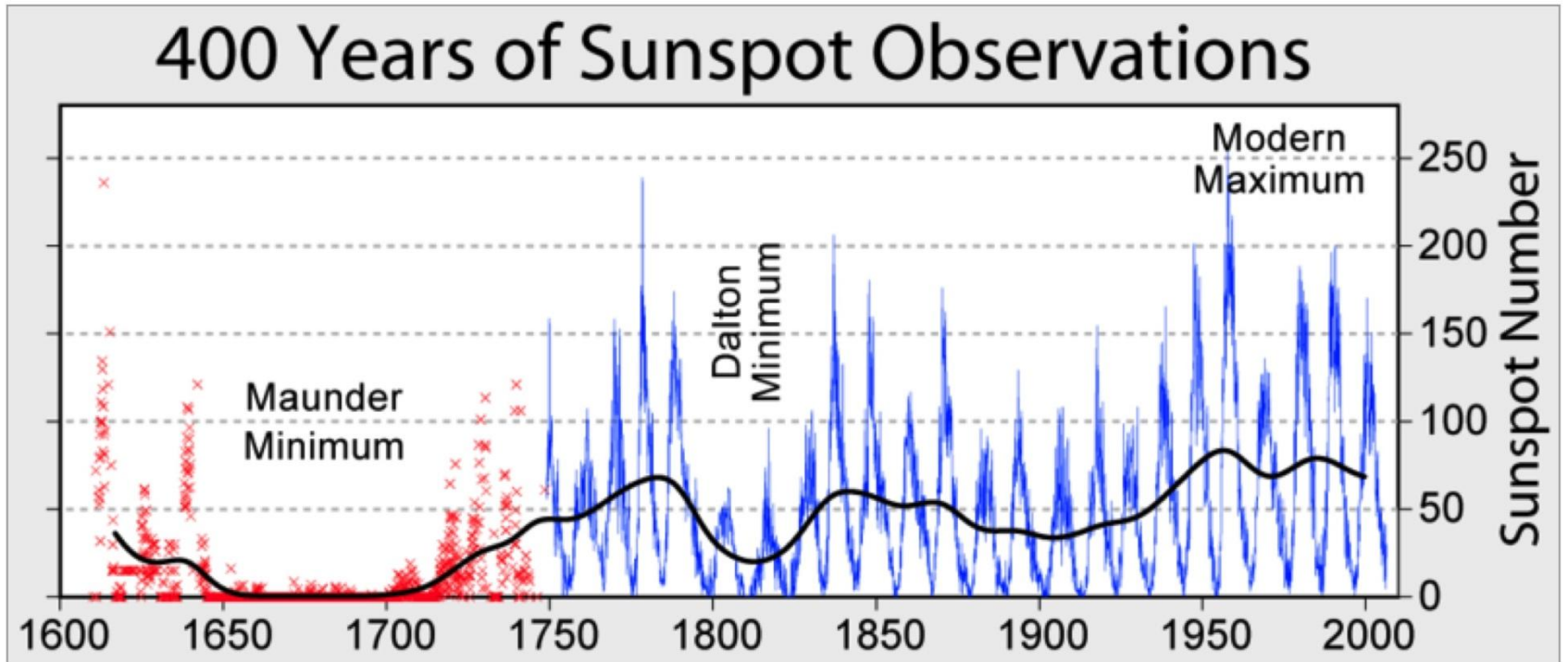


Fig. 20. A north-south section through the atmosphere showing surfaces of constant Be^7 production (nuclei/min, m^3 air at S.T.P.).

Lal and Peters (1967)

Cosmogenic nuclide production varies
over **seasonal to multidecadal time scales**

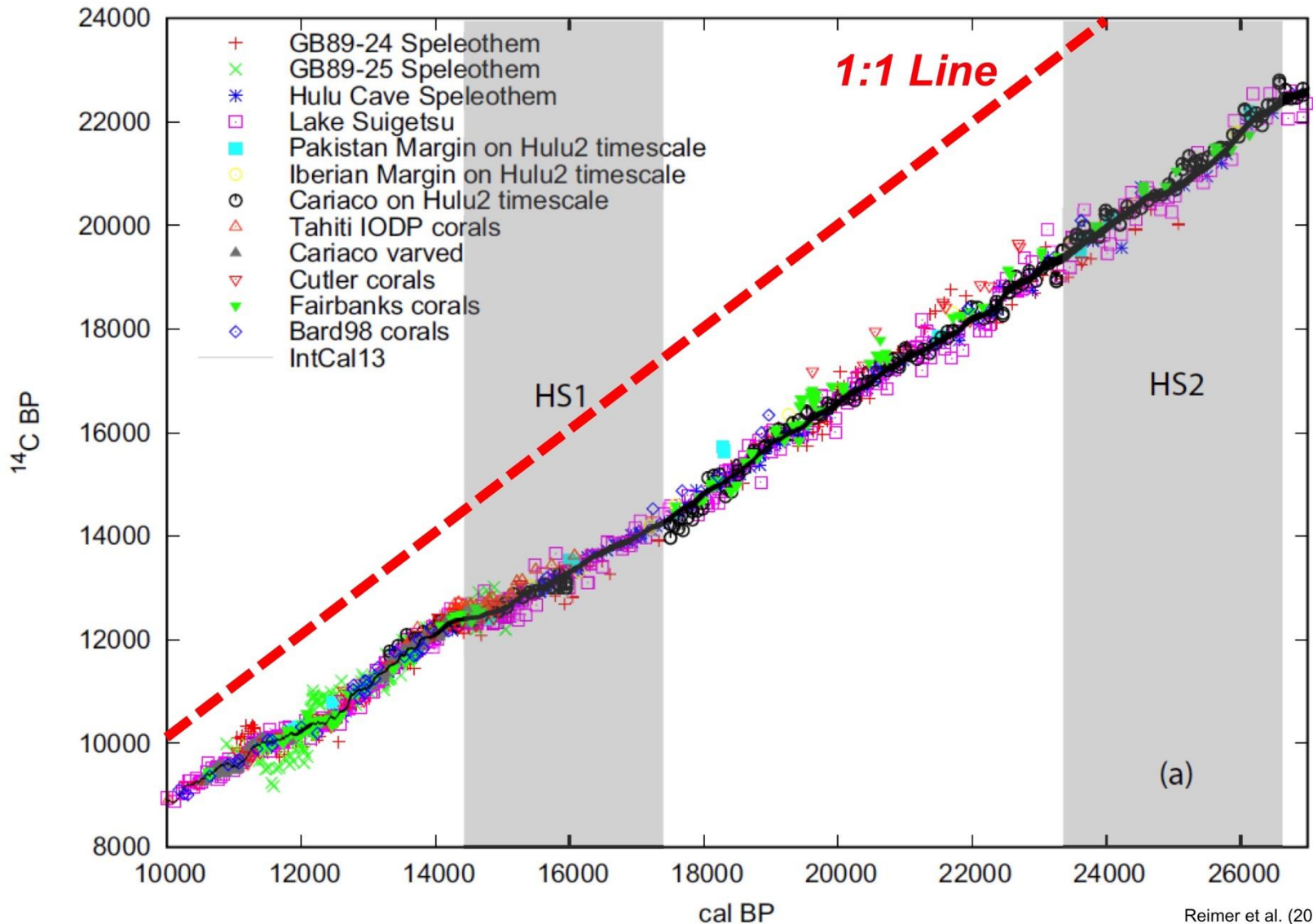


<http://oneminuteastronomer.com/1042/sunspots-cycles/>

{
"Little Ice Age"

Source variability complicates use of ^{14}C

Cosmogenic Radionuclides: Correction Factors



Reimer et al. (2013)

What are Cosmogenic Radionuclides used for?

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

Studies include:

a) Large-scale ocean circulation

^{14}C ($t_{1/2} = 5,700$ yr)

b) Short time scale upper ocean vertical mixing rates and atmospheric deposition

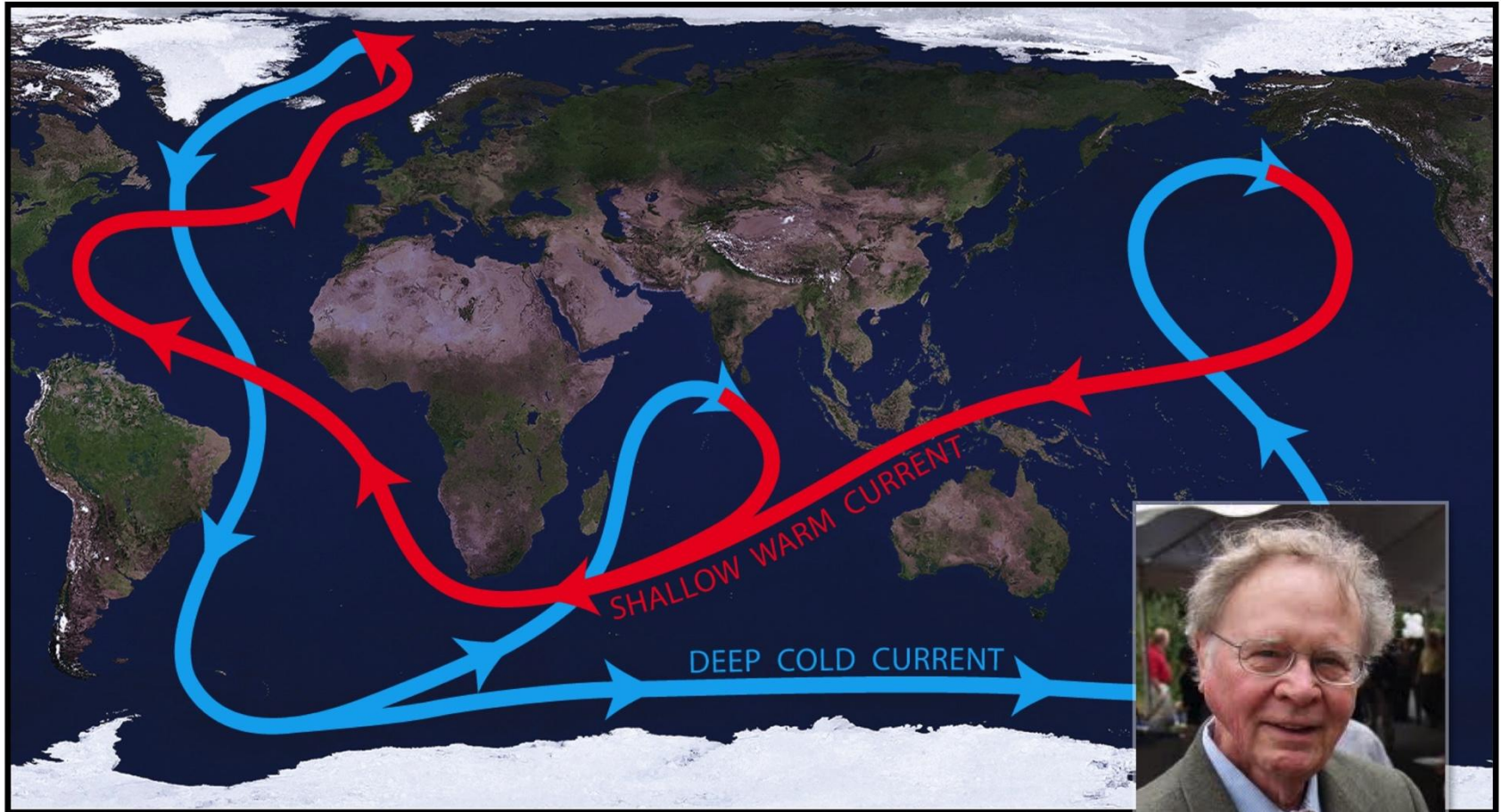
^7Be ($t_{1/2} = 53$ d)

c) Age dating, mixing, particle transport and erosion

^7Be ($t_{1/2} = 53$ d)

^{14}C ($t_{1/2} = 5,700$ y)

Cosmogenic Radionuclides: a) Ocean Circulation

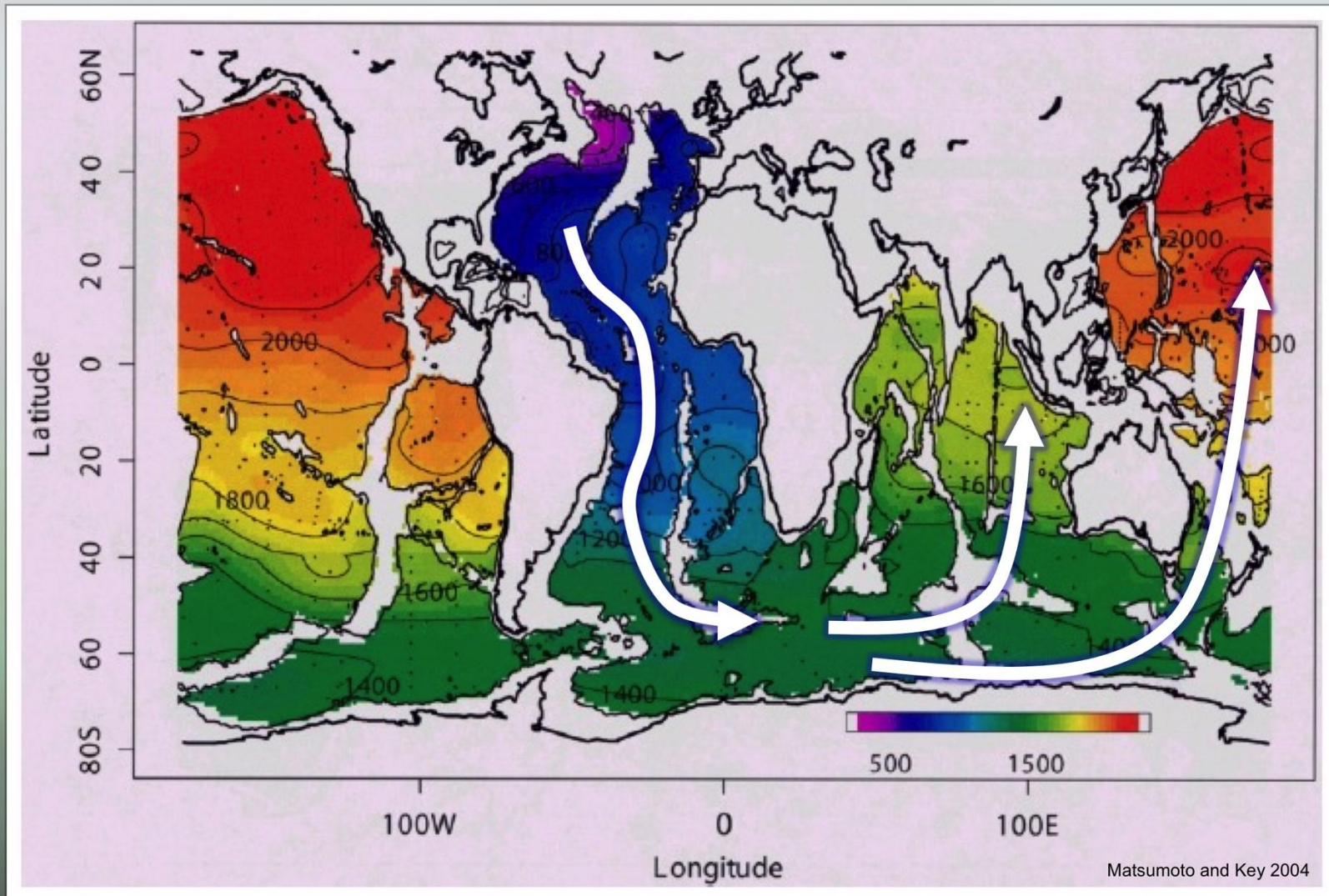


stempreacademy.hawaii.edu/c-more/ocean-conveyor-belt

W. Broecker

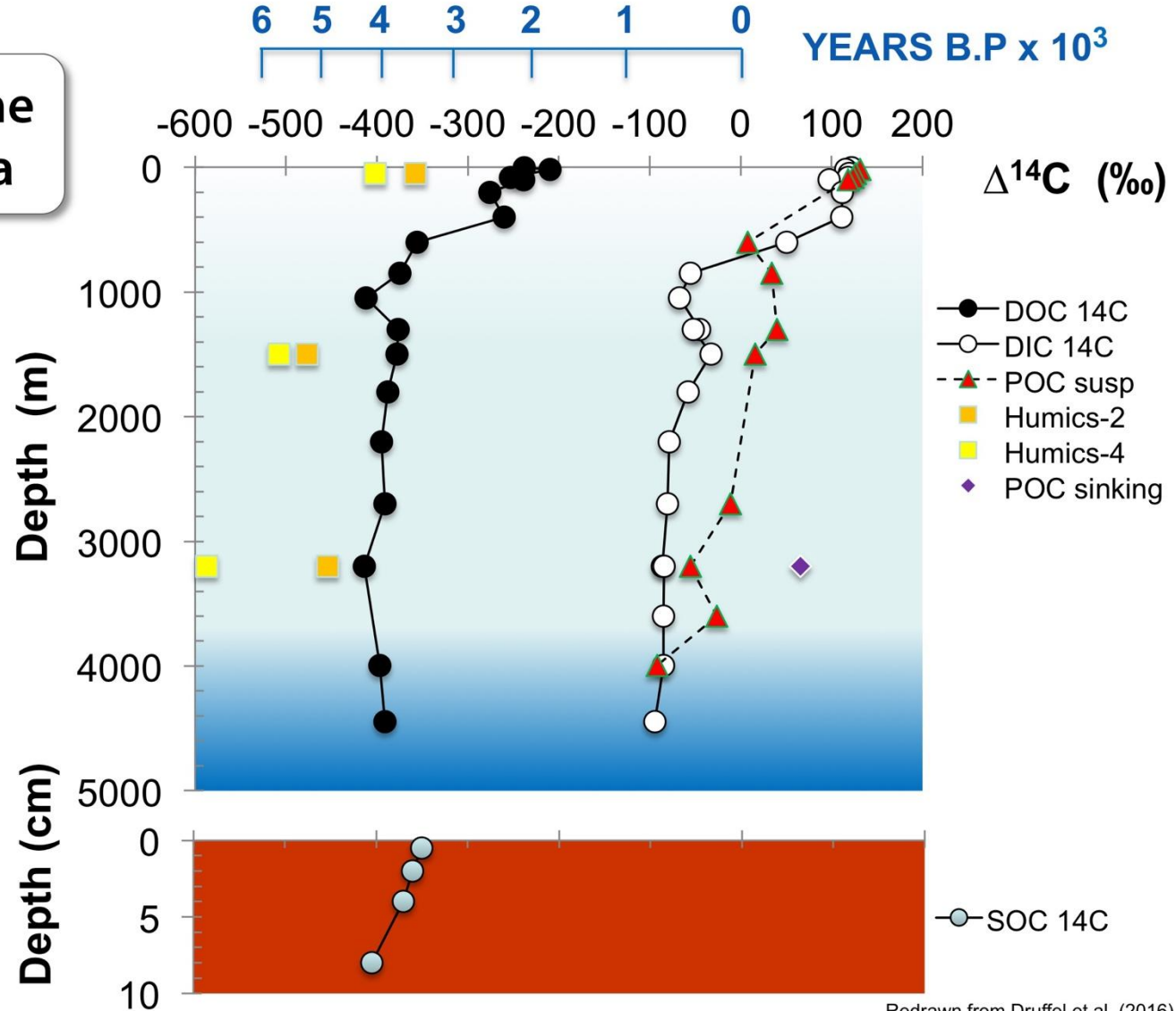


Circulation and age of ocean waters at 3500 m determined from ^{14}C



Cosmogenic Radionuclides: c) Age Dating

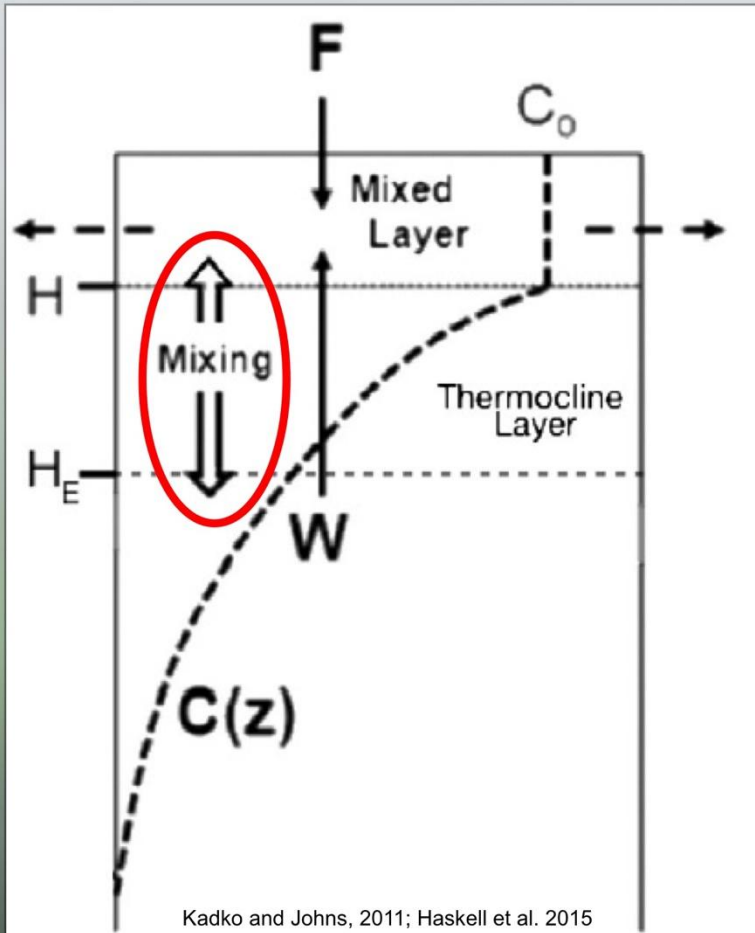
Data from the
Sargasso Sea



Redrawn from Druffel et al. (2016)

Cosmogenic Radionuclides: **b) Upper Ocean Mixing**

^7Be ($t_{1/2}=53$ d) as conservative tracer for upper ocean mixing: conceptual model

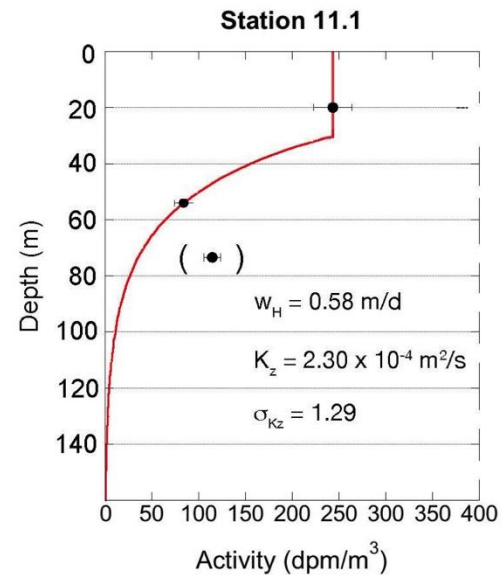
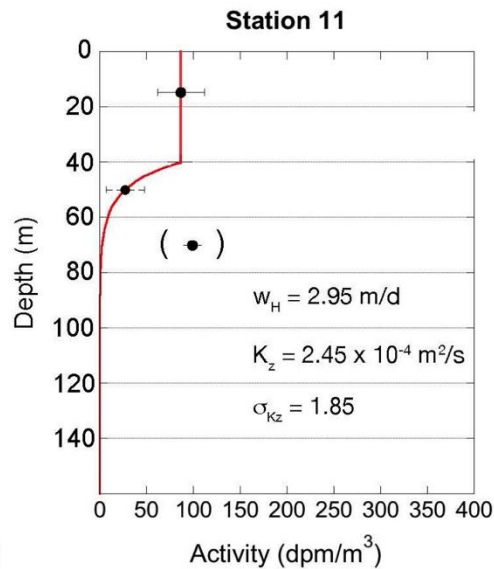
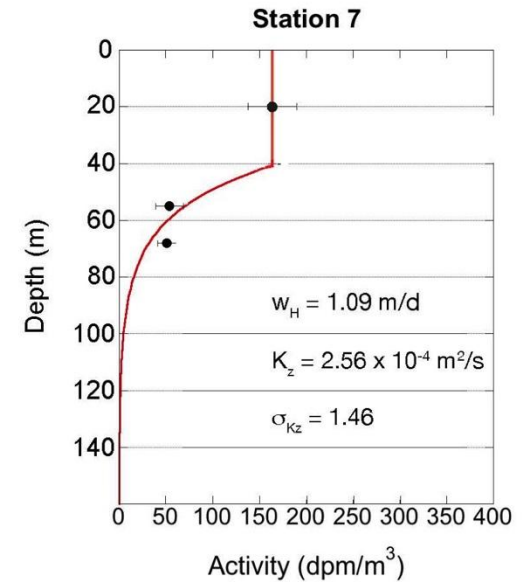
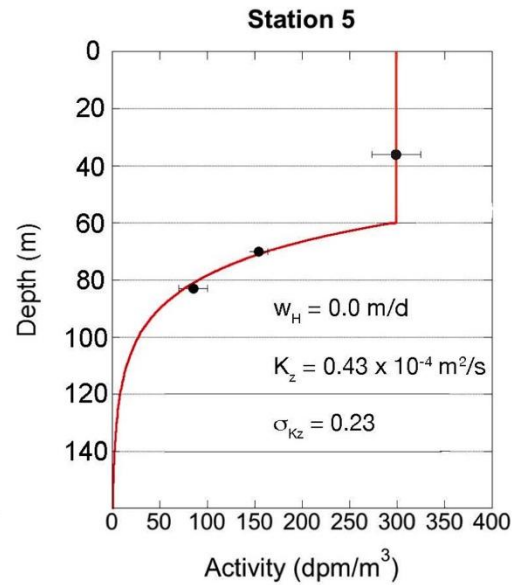
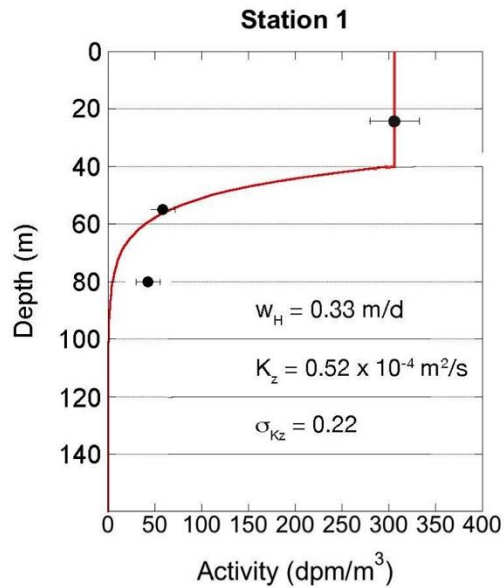


$$H \frac{\partial C}{\partial t} = F_a - J - \lambda C_0 H + w_H C_0 - P$$

$$J = w_H C_0 - K_z \left(\frac{\partial C}{\partial z} \right)_H$$

- C_0 = change in ^7Be concentration along a mixing line (dashed)
- F_a = Atmospheric ^7Be 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 ^7Be activities.



Haskell et al. (2015)

Anthropogenic Radionuclides

Bikini island 1946



Wikipedia.org

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

Major sources of Anthropogenic Radionuclides to the environment



1) Nuclear weapons

2) Nuclear fuel cycle



Tennessee Valley Authority

Why produce Anthropogenic radionuclides?

Nuclear power

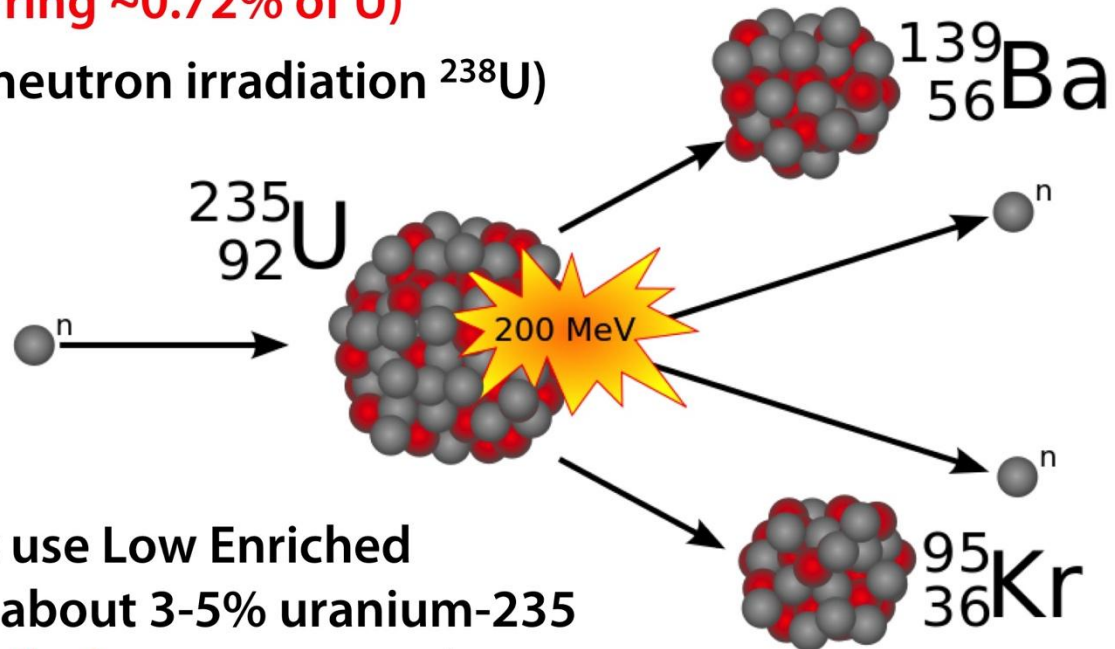
It's all about using a fission chain reaction to release energy!

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.

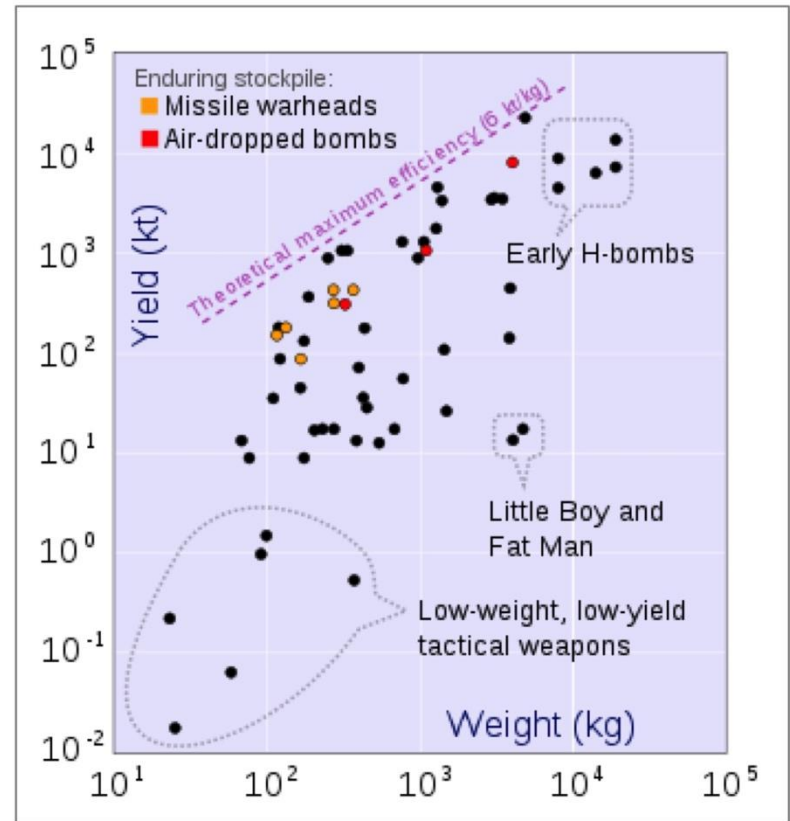
Why produce Anthropogenic radionuclides?

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.



Alternative uses of Anthropogenic Radionuclides

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

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

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)

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

3) Accidents related to nuclear power/production

- Chernobyl (April 26, 1986) \approx 5000 PBq total;
 \approx 85 PBq ^{137}Cs of which **15-20 PBq to oceans.**
- Fukushima (March 11, 2011) \approx 500 PBq total;
 \approx 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)
 \approx **0.630 PBq** ^{238}Pu mainly in Southern Hemisphere
- Undetonated weapons in Palomares, Spain (6 Jan 1966)
 \approx **1.4 PBq** Pu isotopes
- Undetonated weapons in Thule, Greenland (21 Jan 1968)
 \approx **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)

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)

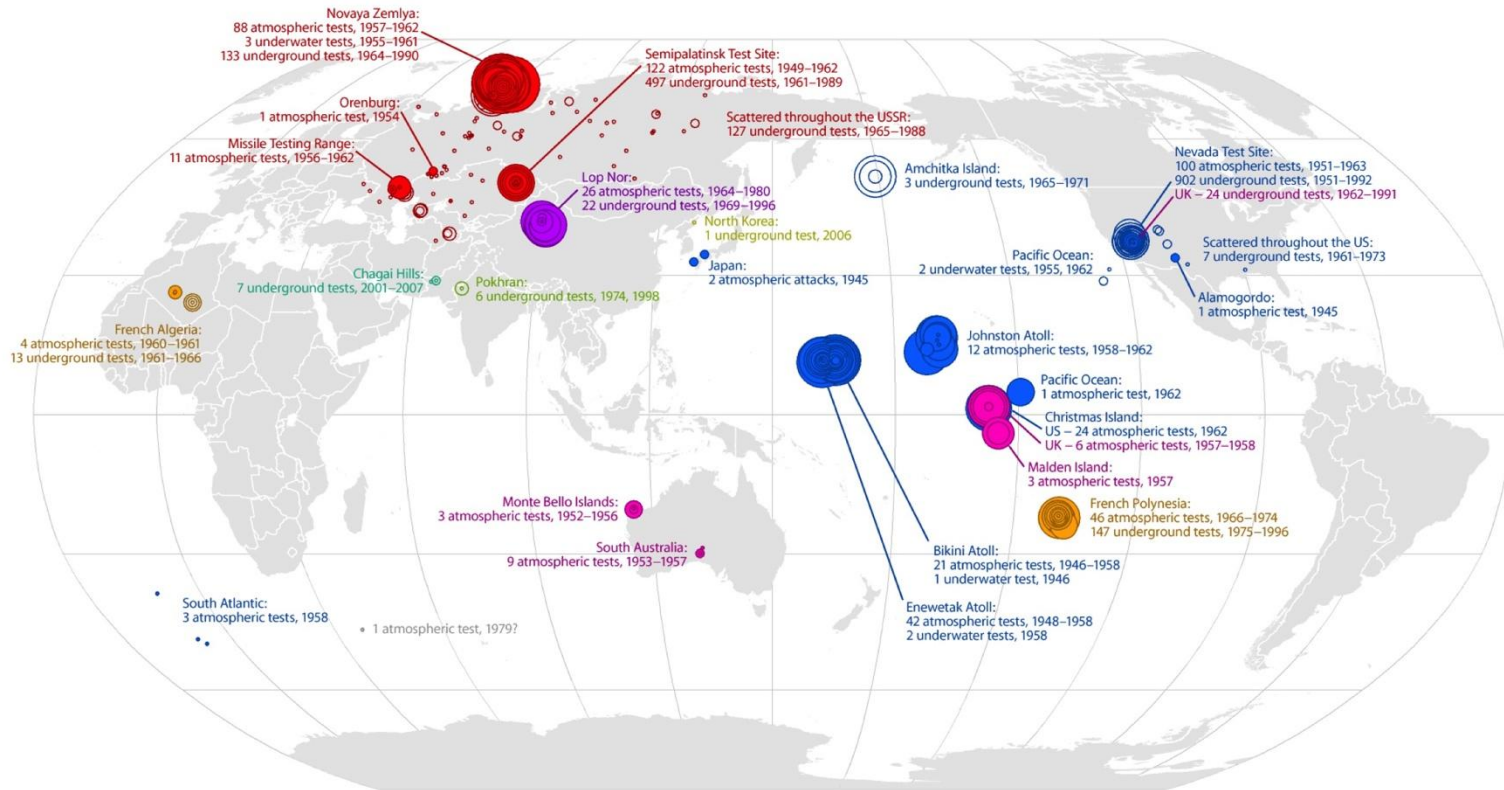
≈ 45 PBq Other categories



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

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

Nuclear Weapons Testing 1945 - 2007



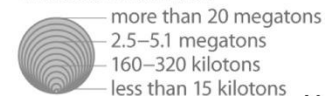
| Country: | Year of first detonation: | Number of detonations: | | |
|-----------------------|---------------------------|------------------------|-------------|------------|
| | | atmospheric | underground | underwater |
| United States | 1945 | 206 | 912 | 5 |
| USSR | 1949 | 223 | 756 | 3 |
| United 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:



Map created by radicalcartography.net

Atmospheric Nuclear Weapons Testing 1945 - 1980

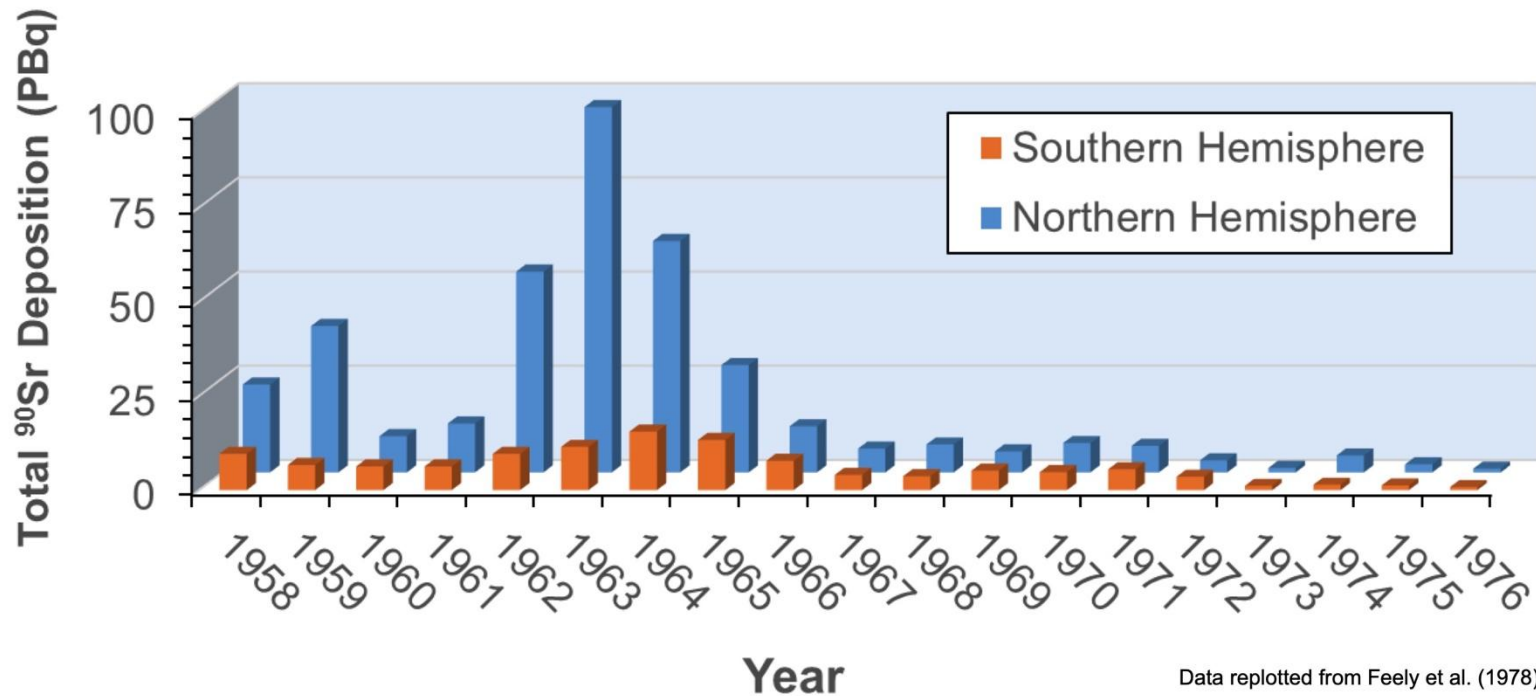
Atmospheric nuclear weapons testing led to temporal and spatial variability in *global* radioactive fallout by atmospheric deposition.

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”**

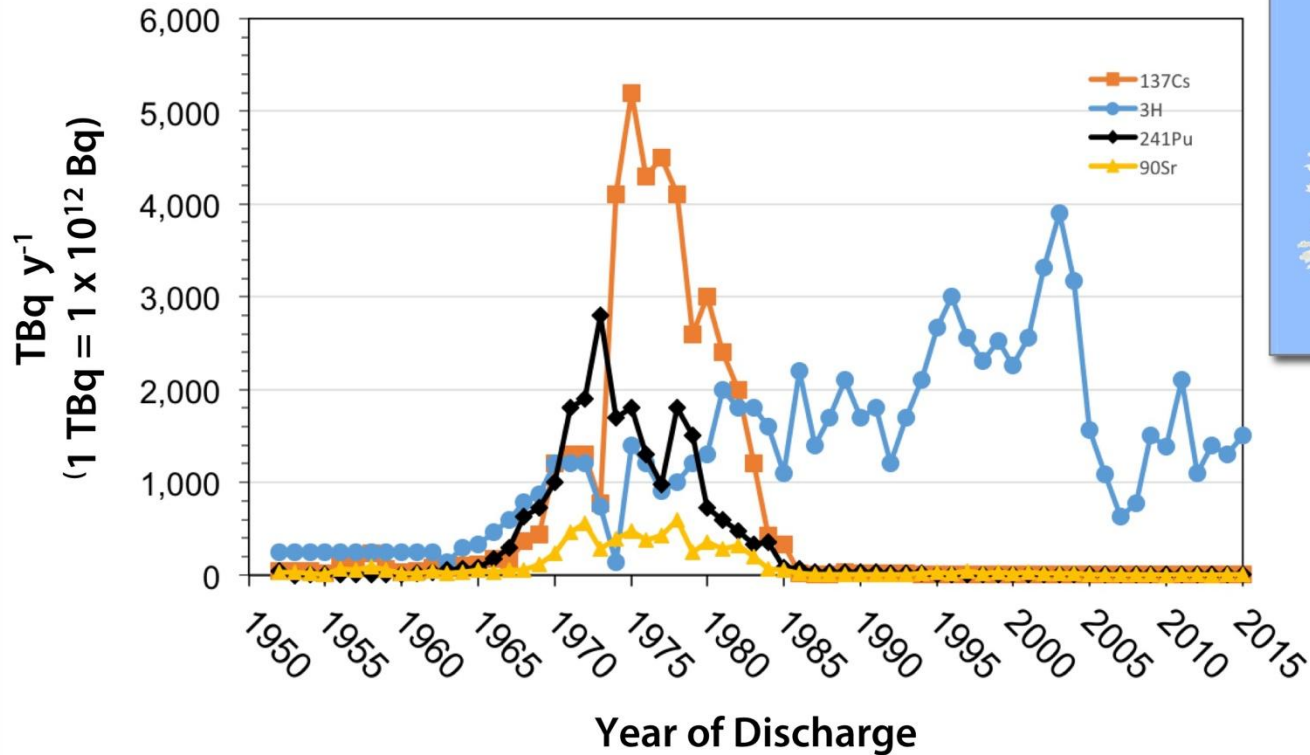
78% percent is **“global” fallout**, most of which ended up in the **same** hemisphere

Worldwide Deposition of ^{90}Sr through 1976



Planned Releases

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



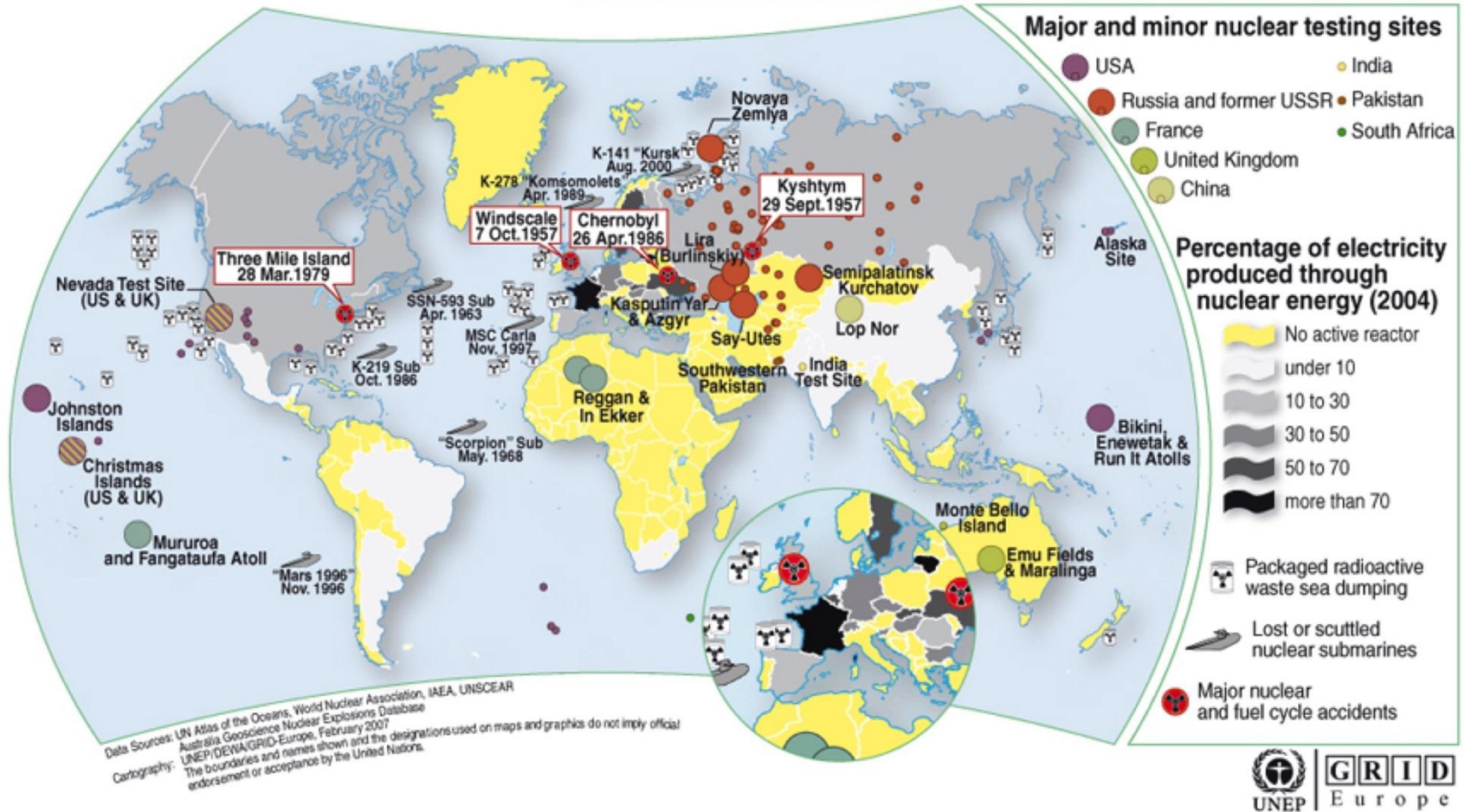
Source from nuclear fuel reprocessing discharges to Irish sea from Sellafield

1 TBq = tera-Becquerel = 10¹² Bq

Adapted from Gray et al. (2005)

Nuclear Power Plant Accidents (not comprehensive)

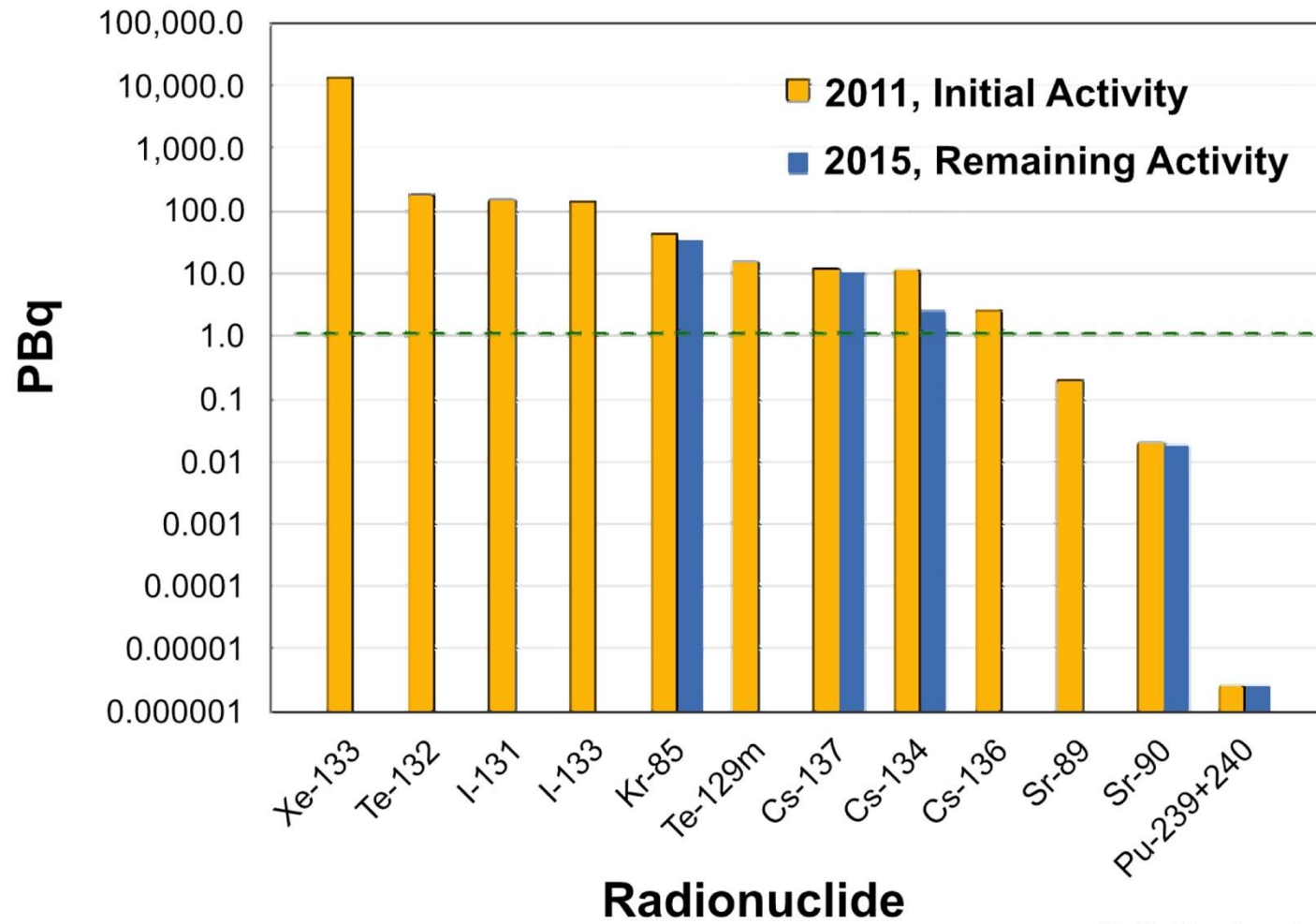
Nuclear concerns for the environment around the globe



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

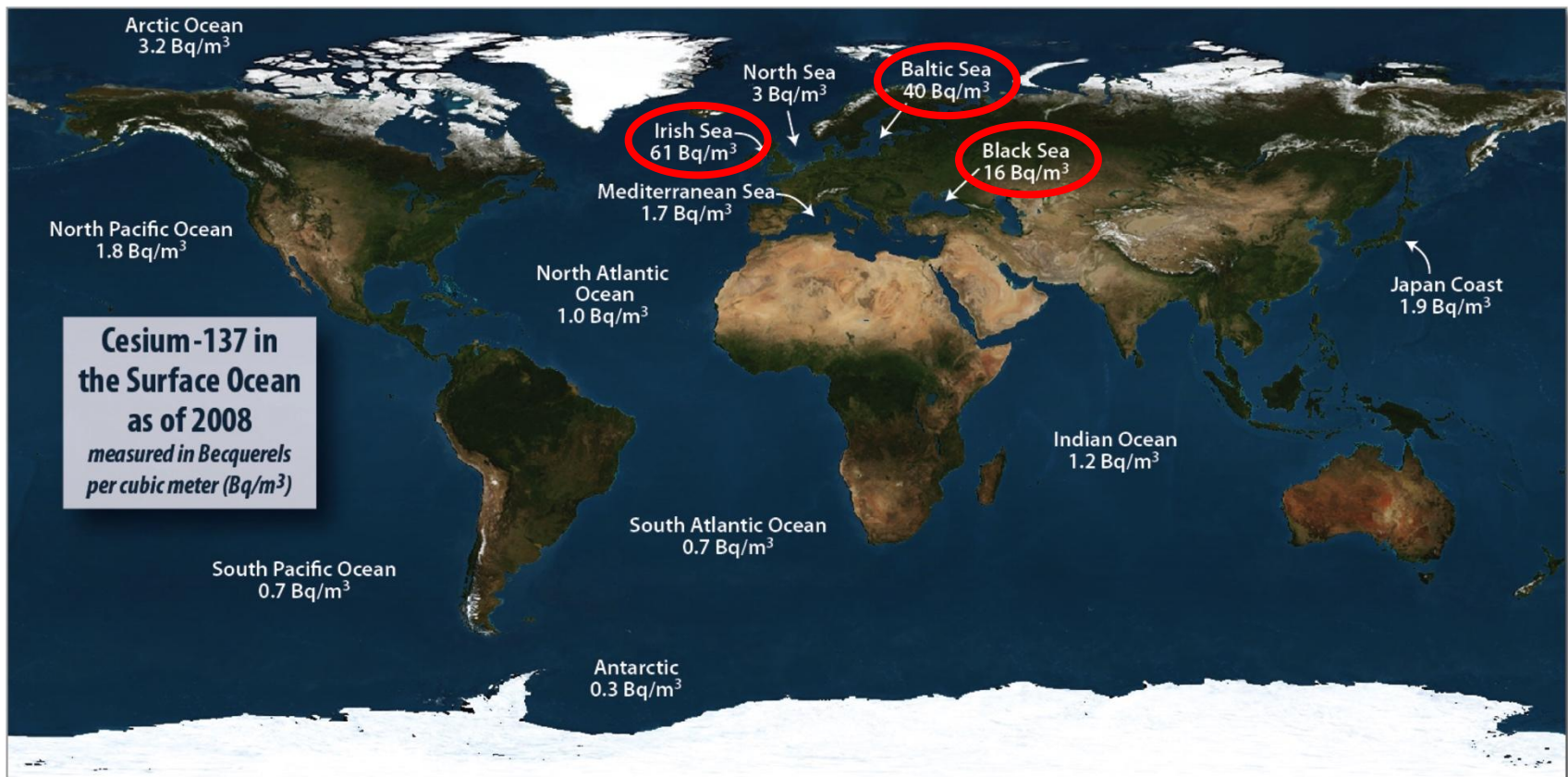
Why so much attention on cesium?

Total Releases from Fukushima Dai-ichi accident



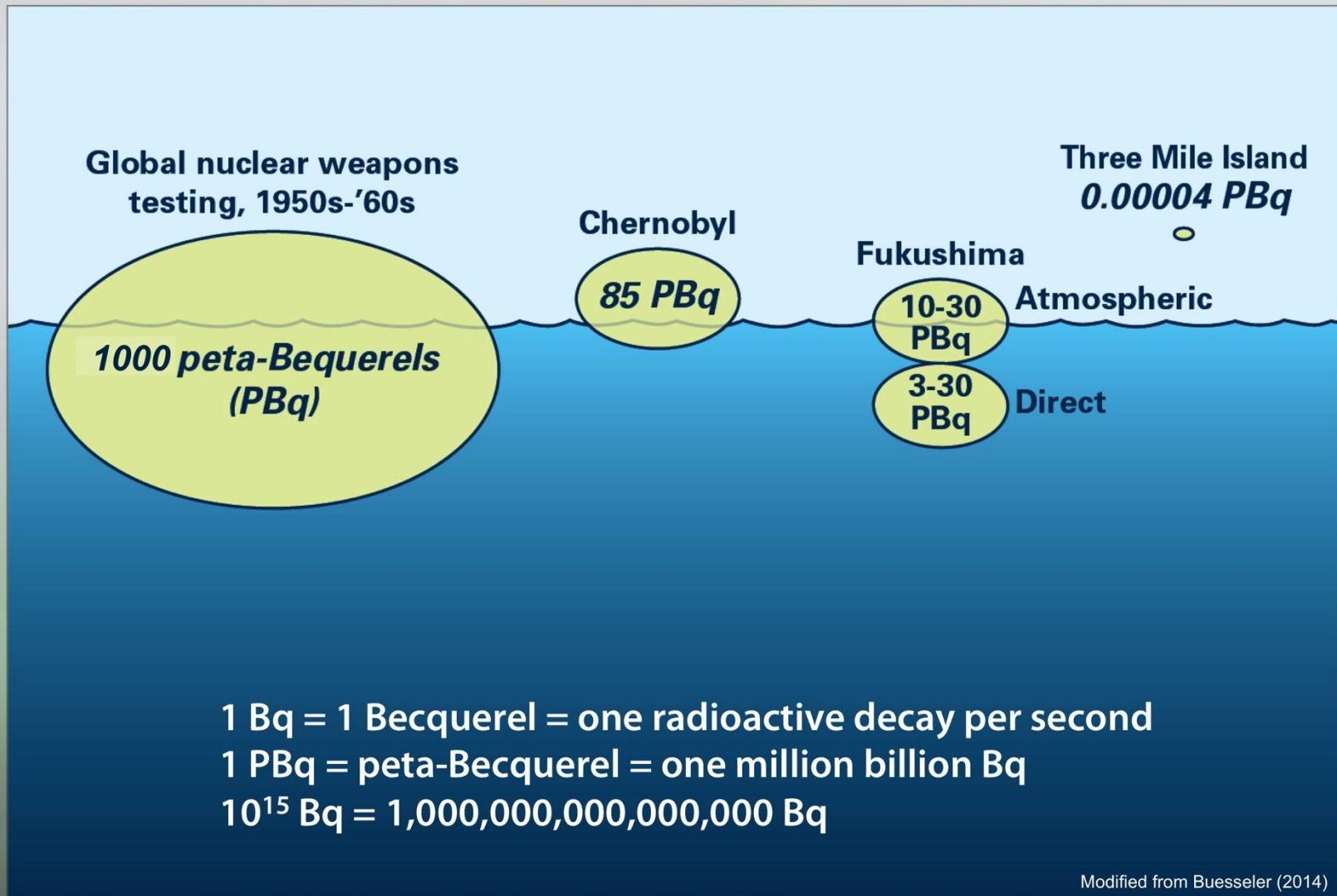
Modified from Buessler (2014)

Uneven distributions of radionuclides in the ocean

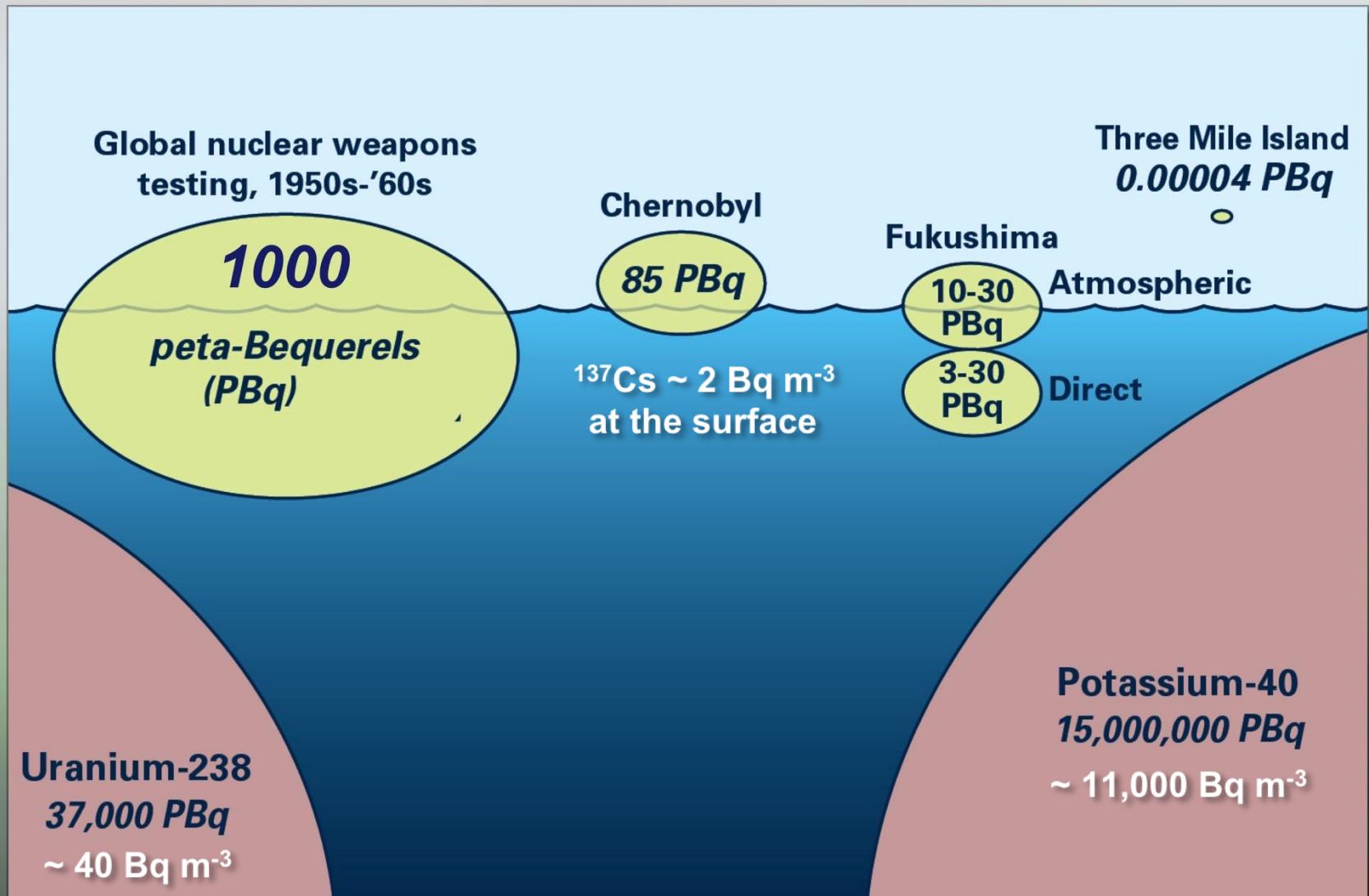


Modified from Buesseler (2014)

Total cesium-137: What are the largest sources?



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



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

Tracer applications of Anthropogenic Radionuclides in the ocean

1

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

^{90}Sr

^{14}C

^3H

^{129}I

^{36}Cl

$^{134,137}\text{Cs}$

2

Particle scavenging & transport
rates in the water column:

^{241}Am

^{144}Ce

^{106}Ru

$^{239,240}\text{Pu}$

3

Biological uptake: ^{14}C

(see slide 16)

4

Sediment accumulation & mixing:

^{134}Cs

^{137}Cs

^{241}Am

$^{239,240}\text{Pu}$

5

Age dating:

^{14}C

^{134}Cs

^{137}Cs

$^{239,240}\text{Pu}$

Case studies

Ocean Circulation

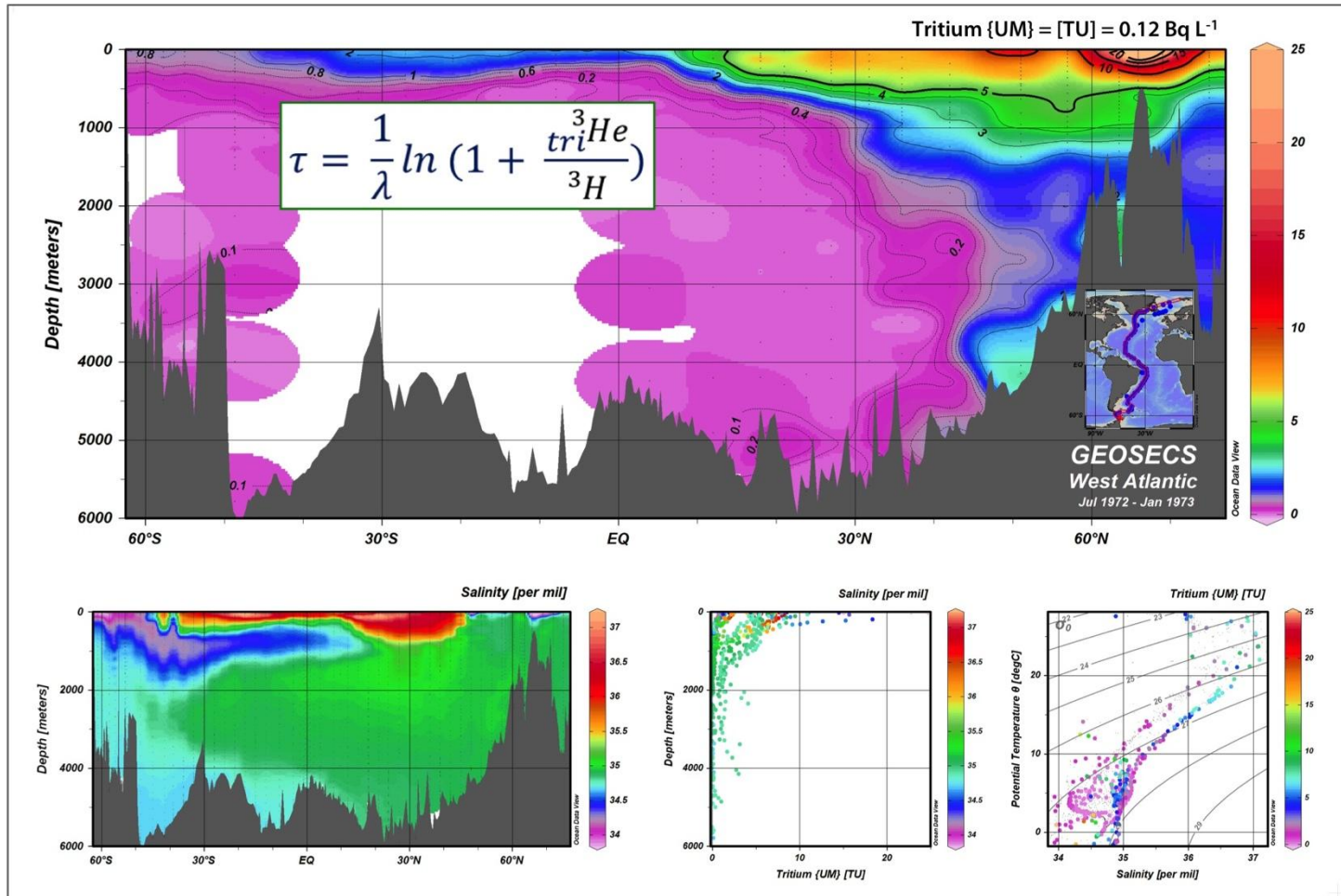
Anthropogenic radionuclides derived from (1) **atmospheric fallout (weapons tests)**, (2) **point sources from nuclear facilities (Sellafield & La Hague)** and (3) **nuclear accidents (Fukushima)** used to trace ocean circulation.

Particle Transport and Burial

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

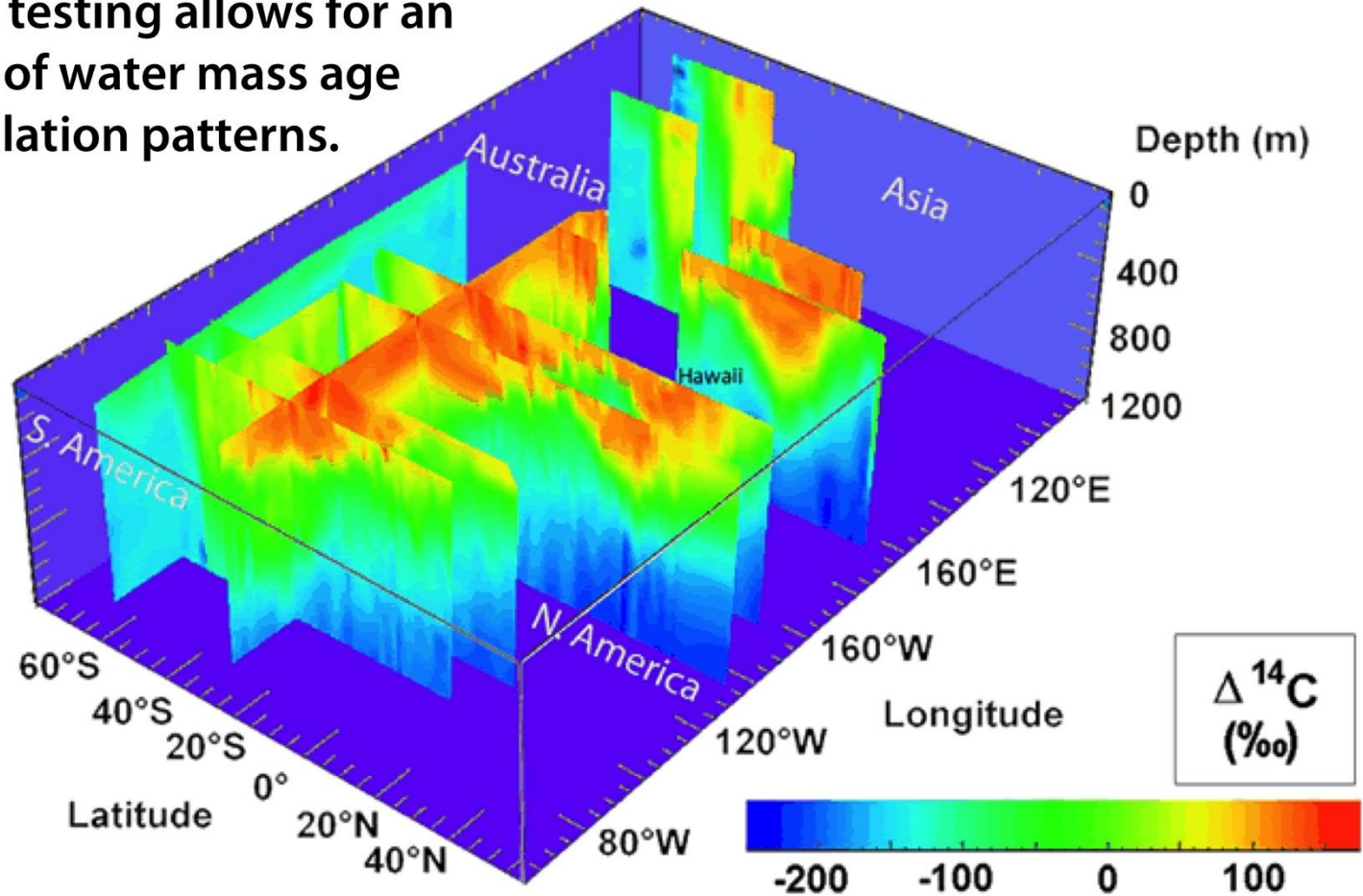
1) Circulation from atmospheric fallout - weapons tests: ^3H ($t_{1/2} = 12.3 \text{ y}$)

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



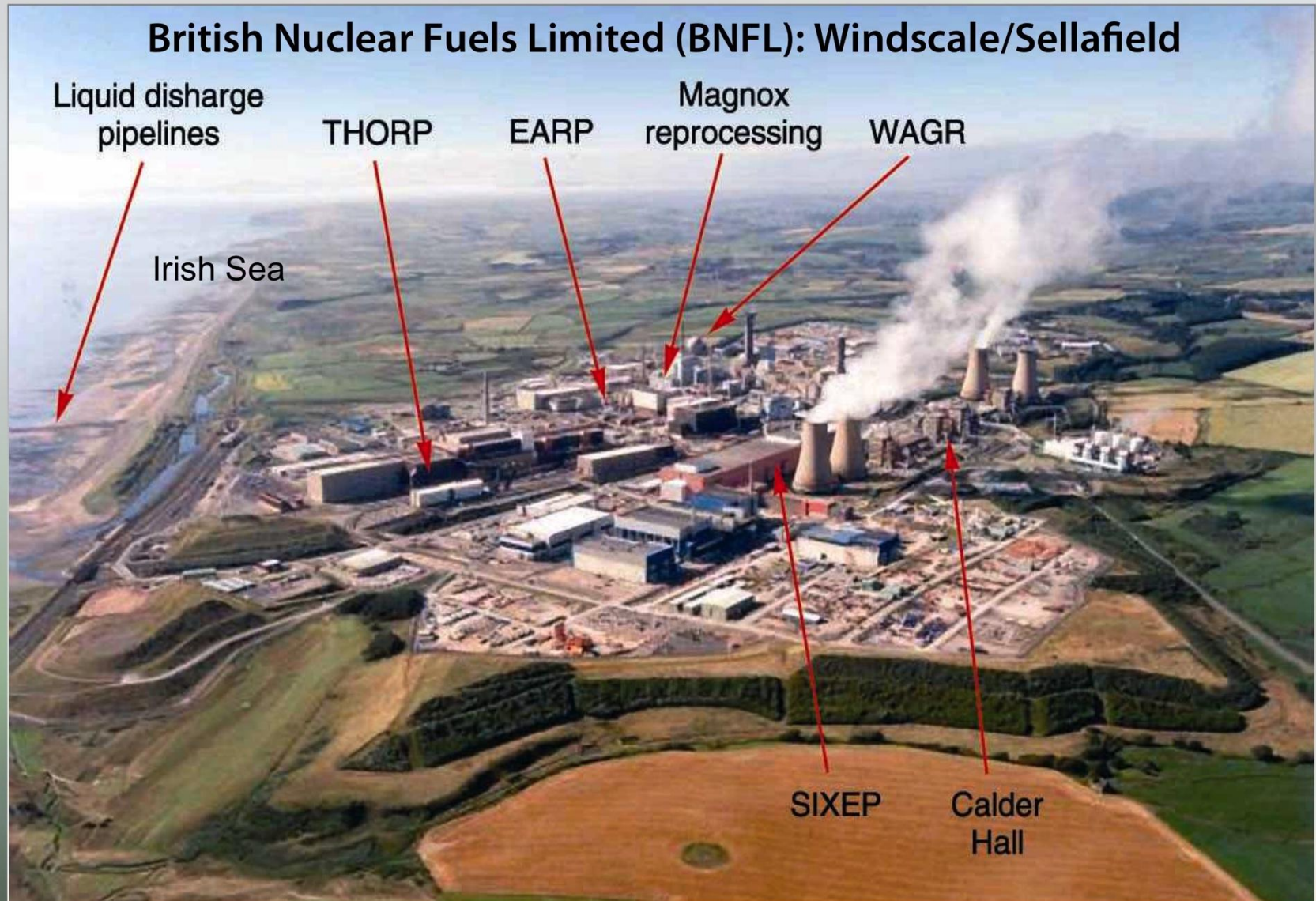
1) Circulation from atmospheric fallout - weapons tests: ^{14}C ($t_{1/2} = 5730 \text{ y}$)

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



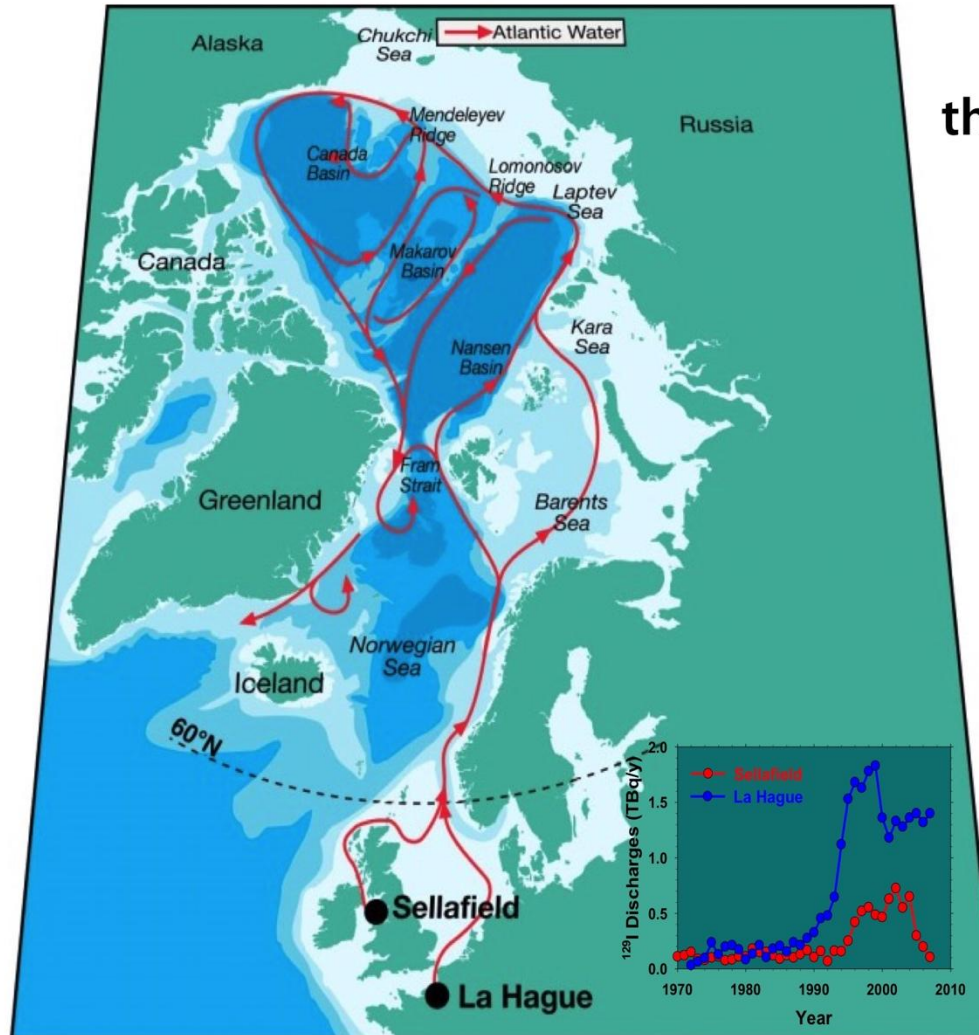
Povinec et al. (2009)

2) Circulation from point sources - nuclear facilities



2) Circulation from point sources - nuclear facilities

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



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

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

● ^{129}I ($t_{1/2} = 1.6 \times 10^7 \text{ y}$)

Karcher et al. (2012)

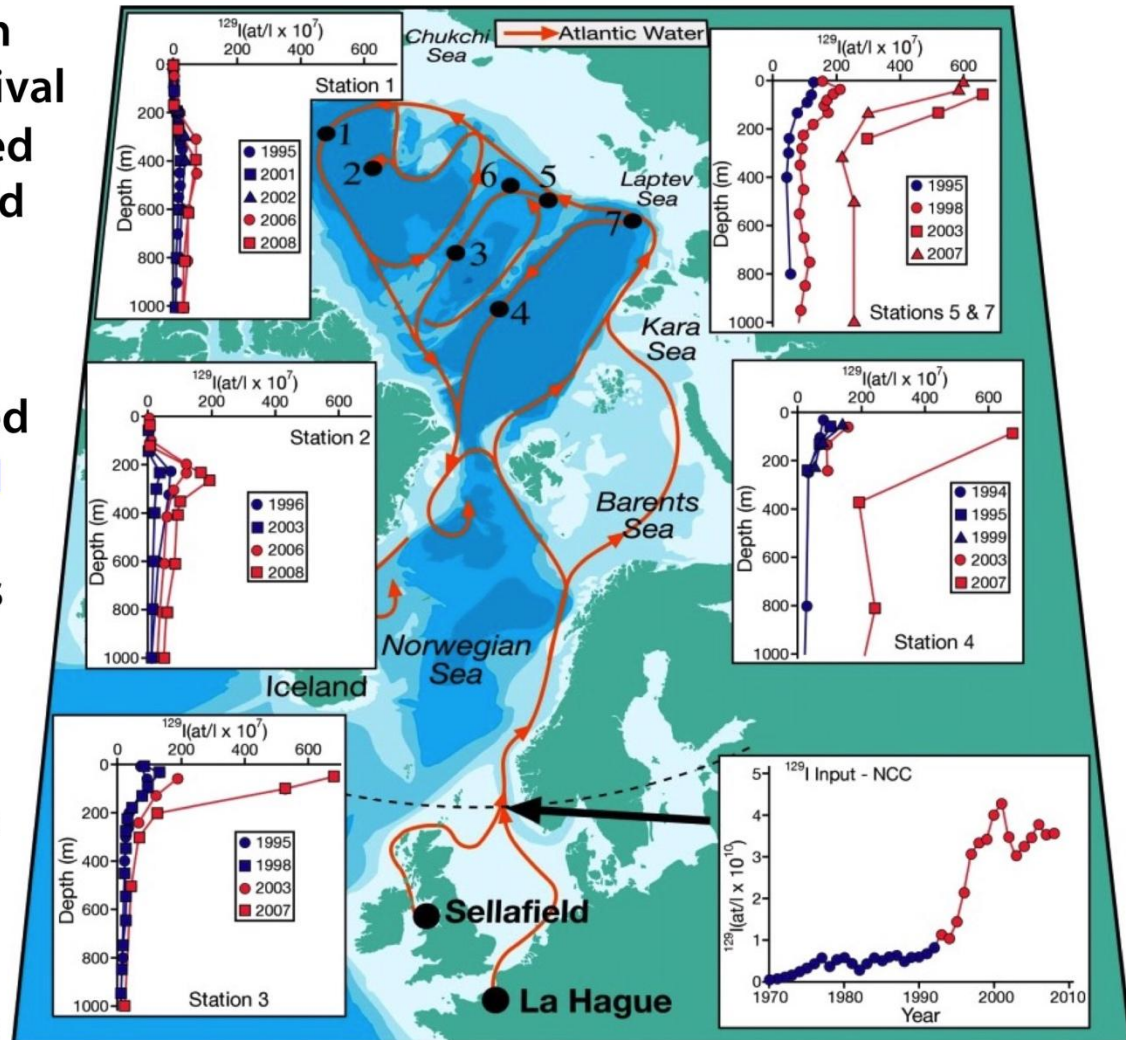
2) Circulation from point sources - nuclear facilities

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

^{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)

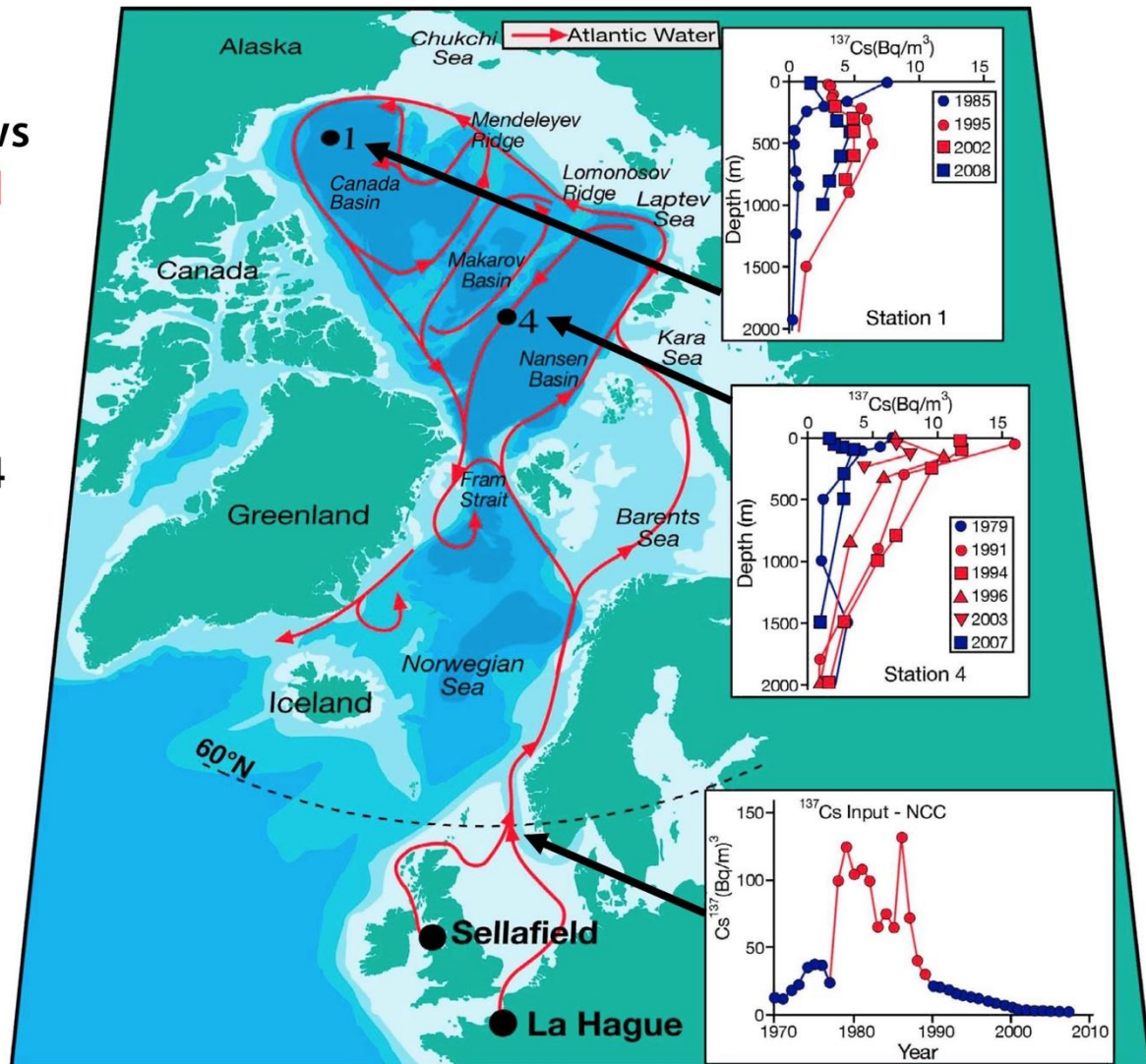


2) Circulation from point sources - nuclear facilities

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

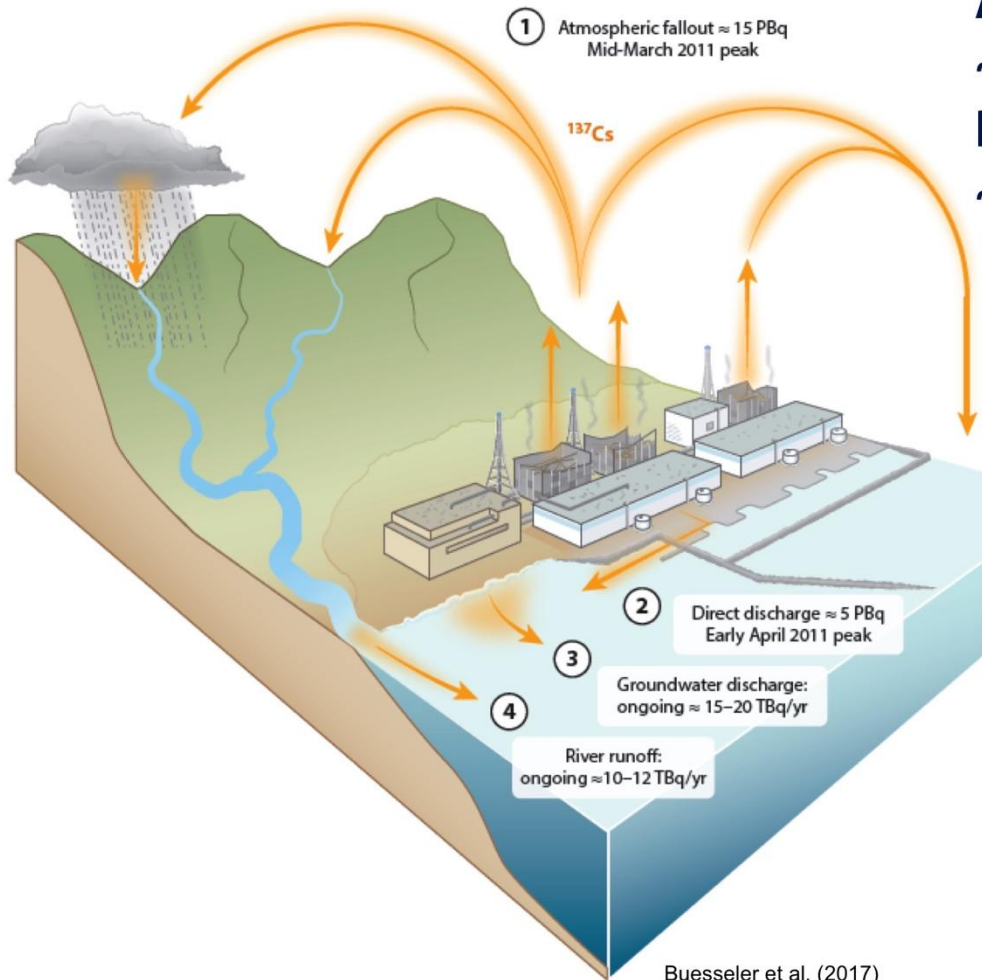
^{137}Cs input function (lower right inset) shows maximum values (red symbols) between 1978 and 1990.

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)

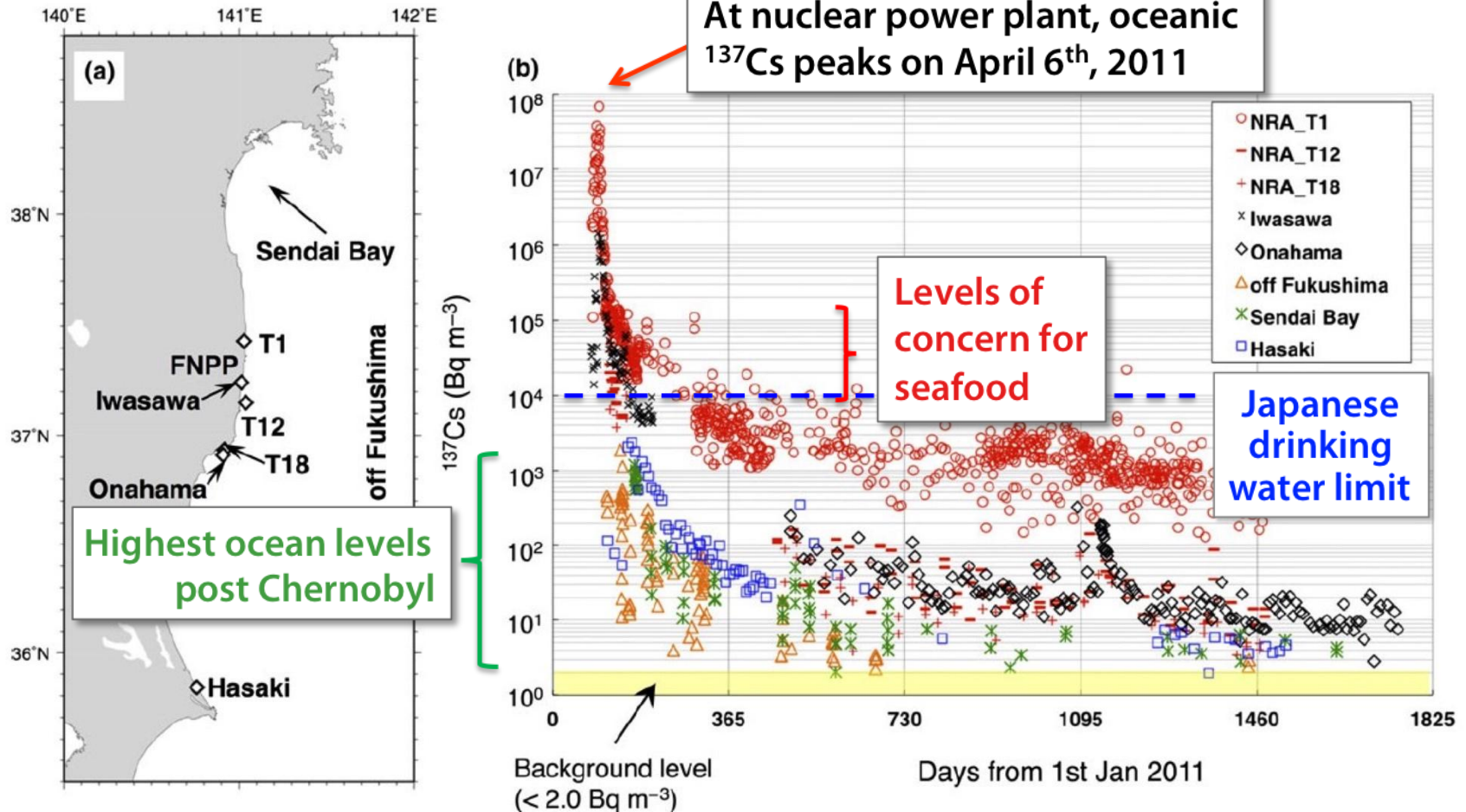
3) Circulation from nuclear accidents: Fukushima, March 2011



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



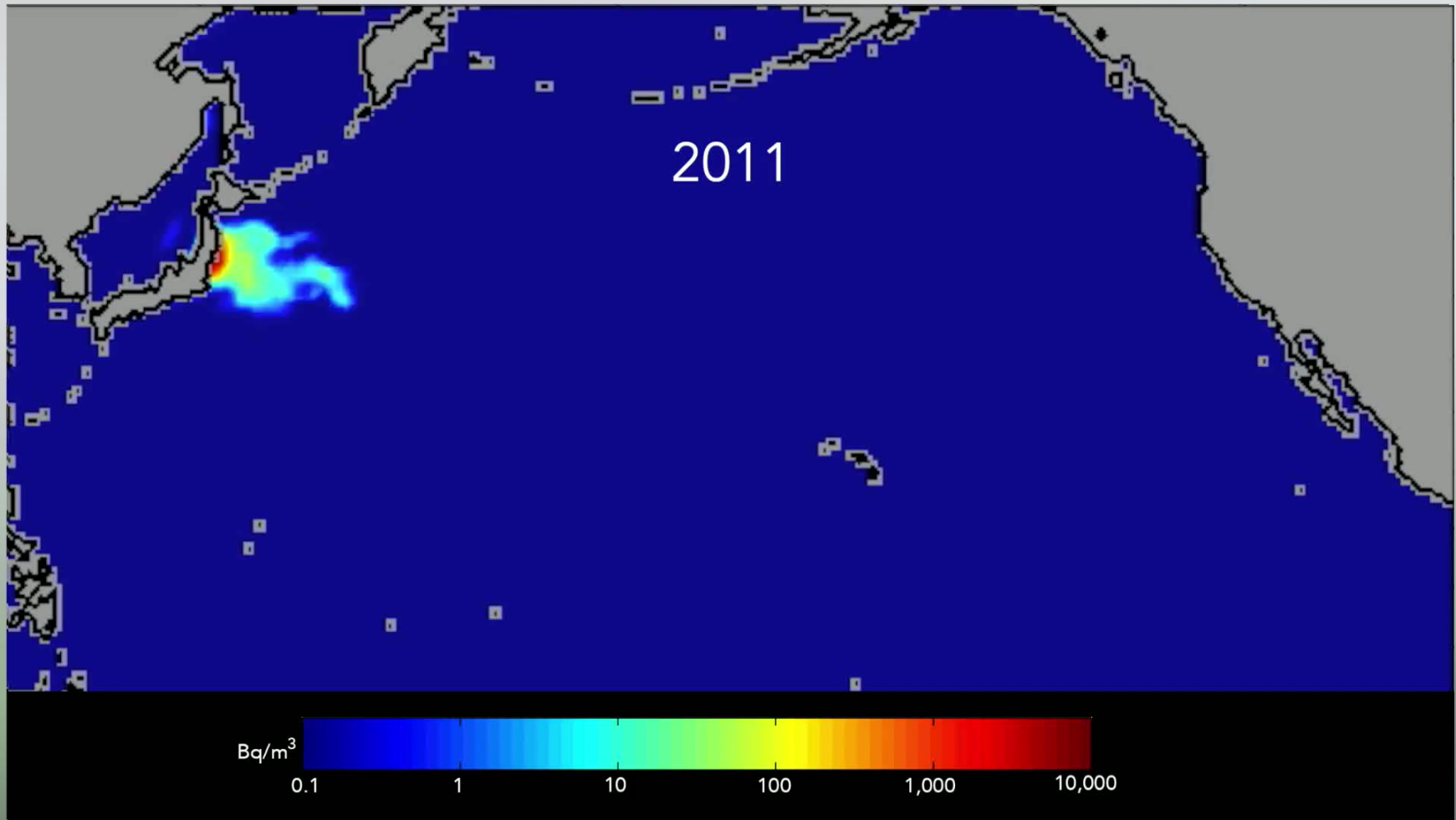
History of ^{137}Cs activity in ocean immediately off Fukushima



Levels prior to March 11, 2011

Kaeriyama (2016)

3) Circulation from nuclear accidents: Fukushima, March 2011



Rossi et al., 2013 & 2014

(Hover over image and click to play video)

Case studies

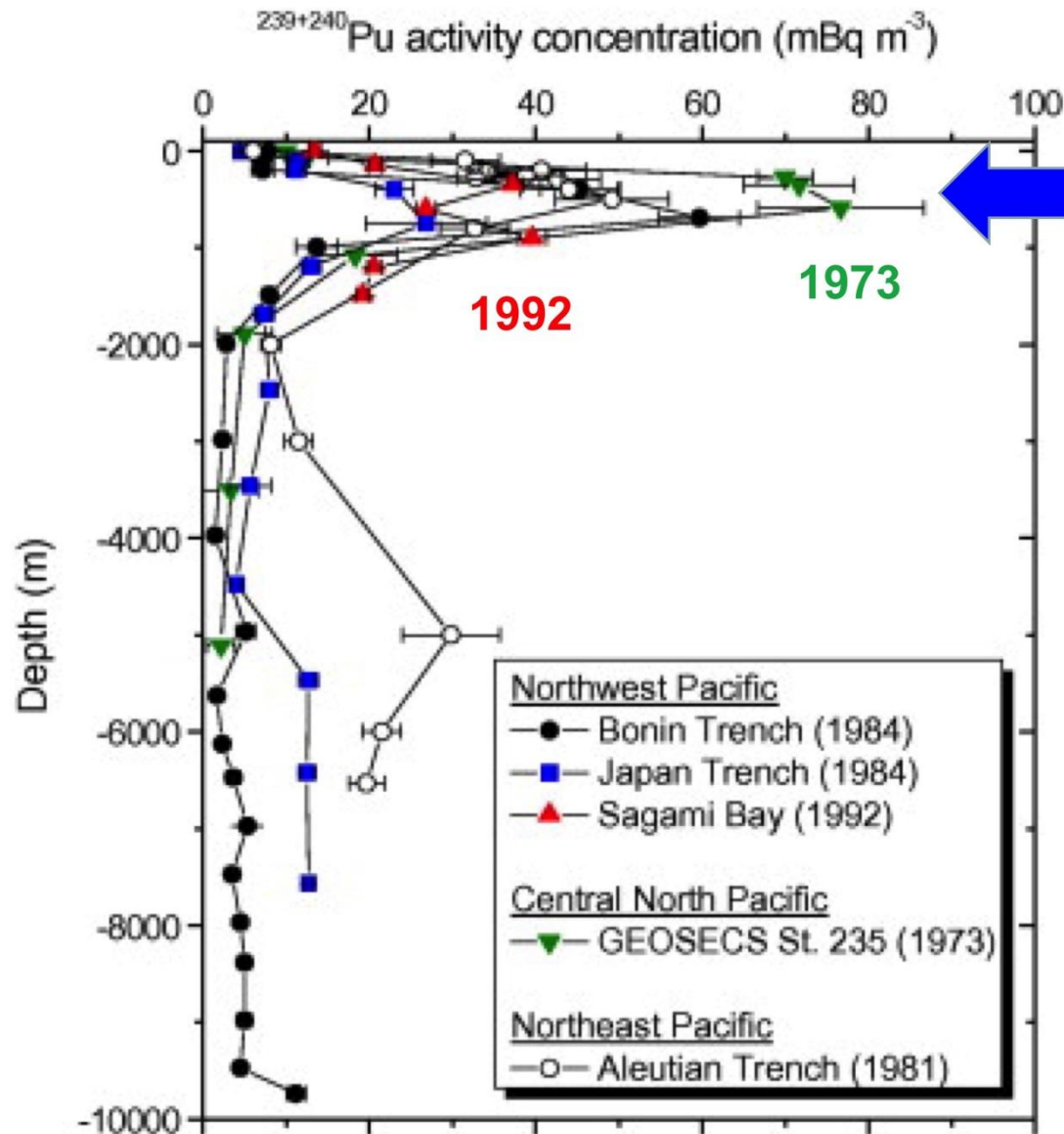
Ocean Circulation

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**.

4) Scavenging Tracers

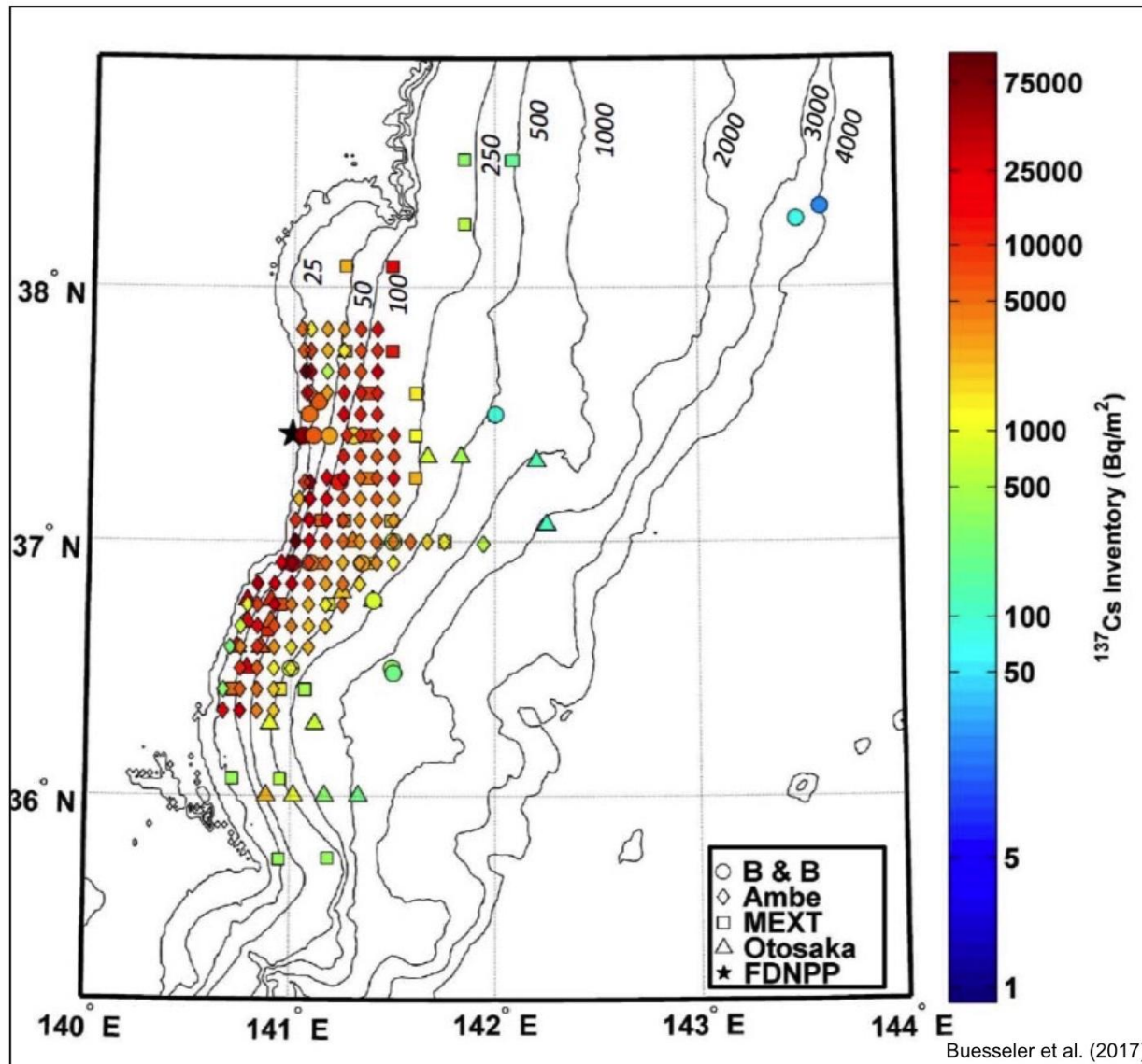


Particle scavenging leads to decrease in atmospherically derived $^{239+240}\text{Pu}$ over time.

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

Lindahl et al. (2010)

4) Scavenging Tracers and Age Dating



5) Age Dating: Event Markers

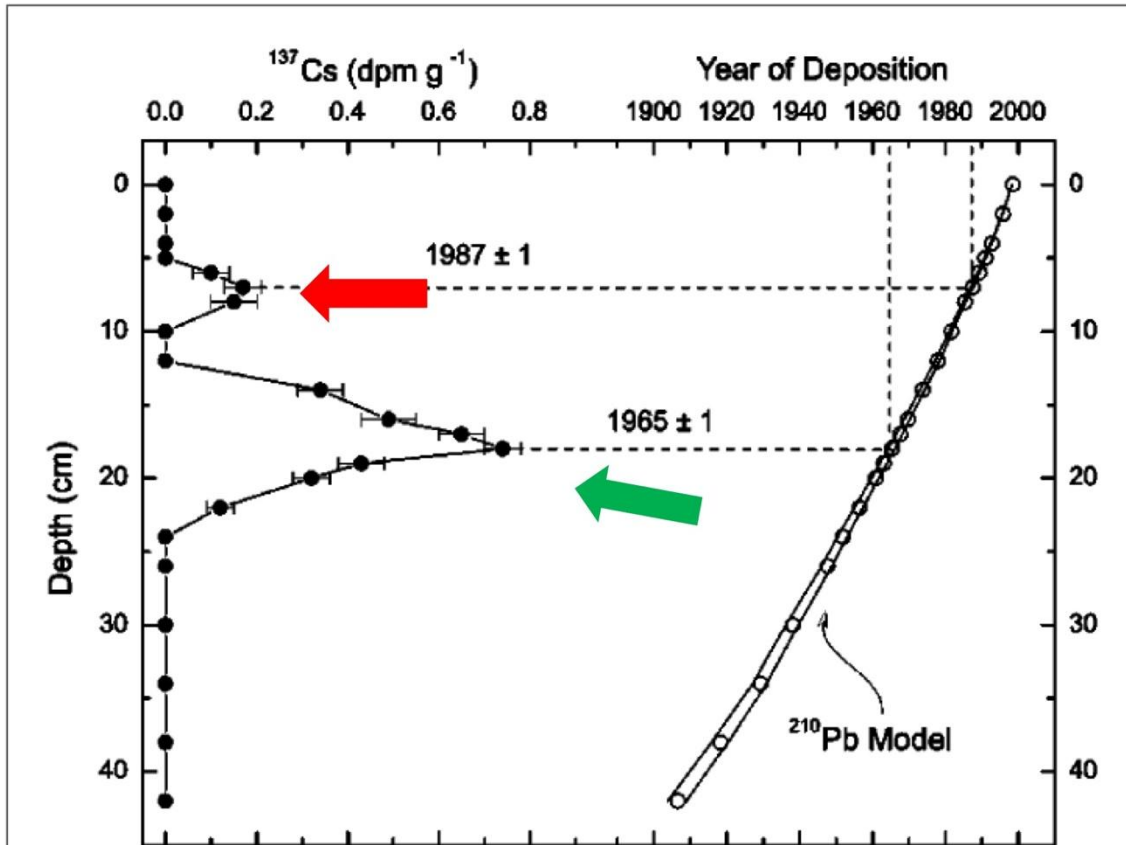


FIGURE 2. ^{137}Cs activity (closed circles) and year of deposition (calculated using unsupported ^{210}Pb , open circles) plotted versus sediment depth. Samples corresponds to 0.5-cm-thick sediment slices. Years of deposition are shown with an envelope of uncertainty ($\pm 1\sigma$).

Lima et al. (2005)

Chernobyl Accident

**Peak in Atmospheric
Nuclear Weapons
testing**

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

How to find marine radioactivity data?

e.g. IAEA MARiS Database for Anthropogenic Radionuclides

Search

Region:

Subregion:

Select a Region and/or Subregion to show samples on the map.

Data Provider:

Sample Type:

Tracer:

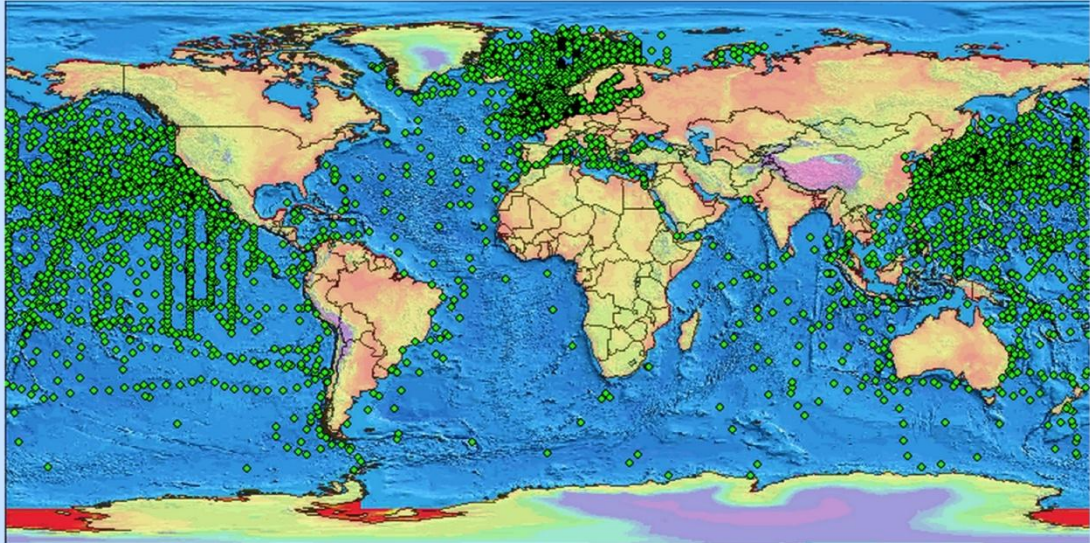
Select a Tracer to enable the Search button.

Depth:

From to

Year:

From to



<http://maris.iaea.org>

<http://maris.iaea.org>

Geotraces: <http://www.egeotraces.org/>

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

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