

RADIOACTIVITY IN THE MARINE ENVIRONMENT: COSMOGENIC AND ANTHROPOGENIC RADIONUCLIDES

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ABSTRACT

Natural and anthropogenic radionuclides are used to study a suite of environmental processes. Yet their applications in aquatic systems are hindered by a general lack of knowledge regarding the underlying concepts of radioactivity, the occurrence of radionuclides in ecosystems, and the equations used to describe their decay mechanisms in environmentally applicable ways. The goal of this lecture is to provide upper level undergraduate and graduate students with a basic understanding of how cosmogenic and anthropogenically produced radionuclides can be used to address a range of environmentally relevant questions in marine systems. The lecture begins with a brief introduction to cosmogenic and anthropogenic radionuclide production and sources to the marine environment. The remaining lecture focuses on specific case studies using these radionuclides in a range of applications including: ocean circulation and mixing, particle scavenging, and sedimentation/age dating. This lecture is the third of a four-part lecture series on radionuclides in the marine environment.

SLIDES

- 1) The overarching goal of this lecture is to provide a basic understanding of how cosmogenic and anthropogenic radionuclides are used in the marine environment to investigate a variety of processes. The lecture begins with a brief introduction to cosmogenic and anthropogenic radionuclides and their distribution in the marine environment. The remaining lecture focuses on specific case studies using these radionuclides in a range of applications including: Ocean circulation, particle scavenging, and sedimentation/age dating.
- 2) Why study radionuclides? Radionuclides can be used as clocks to study key processes and rates in the ocean. They are ubiquitous, relatively easy to measure, and have a wide range of half-lives spanning seconds to billions of years. One of the classic introductory texts to this topic is *Tracers in the Sea* by Broecker and Peng (1982) (Also available online at: http://www.ldeo.columbia.edu/~broecker/Home_files/TracersInTheSea_searchable.pdf). This text describes a general introduction to marine radioactivity and a suite of basic applications. While dated, the underlying approaches remain the same.
- 3) There are three main sources of radionuclides to the environment and they are loosely classified as the naturally occurring U-Th series radionuclides, naturally occurring cosmogenic radionuclides, and radionuclides produced by anthropogenic processes. Some radionuclides have both cosmogenic and artificial sources (e.g., ^3H and ^{14}C). Note that other primordial radionuclides exist as well, such as ^{40}K , but are less commonly used in marine applications. These radionuclides enter the ocean via both point and non-point sources and their distributions are controlled by their biological and chemical reactivity and half lives. The goal of this lecture is to introduce students to using cosmogenic radionuclides and radionuclides produced by anthropogenic activities to gauge marine processes.
- 4) In contrast to U-Th series radionuclides, the major sources of oceanic relevant cosmogenic radionuclides occurs via atmospheric deposition. Anthropogenic radionuclides enter the oceans via a variety of pathways, but on the global scale, the flux is dominated by atmospheric deposition as well.
- 5) There are a number of naturally occurring radioactive elements produced from cosmogenic processes. This table presents a general list of the cosmogenic radionuclides found in marine systems, their source, and globally averaged *in situ* atmospheric and oceanic production rates.

The most common radionuclides used in marine systems are ^{14}C and ^7Be . For more information, see Lal (1999).

- 6) Cosmogenic radionuclides are produced via spallation (from the Middle English verb 'to ship or fragment'): A high energy nuclear reaction that results in the emission of two or more fragments from the nucleus.
- 7) High energy cosmic rays (protons and neutrons) continually bombard the Earth's surface. These rays interact with atmospheric gases (e.g., N, O, and Ar) and create secondary neutrons and protons of lower energy (as well as a number of other particles) that also interact with atmospheric gases, causing a "Nuclear Cascade." As a result, cosmogenic radionuclide production varies with height in the atmosphere. Most cosmic rays are galactic and originate from supernovas, but our sun also produces significant cosmic rays. Cosmic rays are about 89% hydrogen atoms (protons), 10% helium, and 1% heavier elements.
- 8) Cosmogenic radionuclide production also varies with latitude due to the Earth's magnetic field, which essentially acts as a shield from the high energy particles that constantly bombard the Earth. The Earth's magnetic field is strongest at the Equator, but decreases in strength at the poles.
- 9) This is one of the reasons why we have the "Northern Lights" or Aurora Borealis. High energy particles enter the atmosphere at the north and south poles as they are not blocked by a strong magnetic field in those regions.
- 10) Combined, cosmogenic radionuclide production increases with both atmospheric height and at increasing latitudes on Earth. Here is an example of atmospheric ^7Be production from cosmic rays. Note that the tropopause (the interface between the troposphere and the stratosphere) follows the contour line of $10 \text{ atoms min}^{-1} \text{ m}^{-3}$ air production. The stratosphere is stratified and gradients in cosmogenic radionuclide production are readily apparent. The troposphere, in contrast, is relatively well mixed due to convection (largest change in temperature). Mt. Everest is shown for scale (Height $\sim 8.5 \text{ km}$).
- 11) As cosmic rays are produced via solar activity, variations in solar activity over seasonal to decadal time scales also influence the production of cosmogenic radionuclides (increased cosmic rays = increased cosmogenic radionuclide production). Here is a figure showing sunspot activity over the past 400 years. Note that sunspot numbers vary from < 10 to more than 200 sunspots per year. Closer examination of the sunspot activity reveals a fairly regular 11 y cycle. Why this cycle occurs is poorly understood, but may be a result of variations in the sun's global magnetic field or "solar dynamo". From 1645-1715, there was a sudden and unexplained reduction in sunspot activity that is referred to as the Maunder Minimum and coincides with what we now know as a colder period in Earth's climatic history called the "Little Ice Age." For more information on sun spots, see the Sunspot Index and Long term Solar Observation (SILSO) website: <http://www.sidc.be/silso/home>.
- 12) Combined, measurements of longer lived cosmogenic radionuclides, such as ^{14}C require careful correction factors to capture the variability associated with seasonal to decadal production rates and changes in reservoir size. These include anthropogenically produced ^{14}C and the production of fossil fuels (Suess Effect). In this figure, the 1:1 ^{14}C conventional age (red dashed line) versus calibrated age is shown. In order to obtain a calibrated ^{14}C age, ages must be corrected using other archives with known ages, such as marine and lake sediments, corals, Bahamian speleothems, and even tree rings. Note that if conventional ages were used, samples would

appear younger than their true age. For more information on how ^{14}C ages are calibrated, see <https://c14.arch.ox.ac.uk/oxcal/OxCal.html>.

- 13) What are cosmogenic radionuclides used for? Many are used to better understand ocean mixing and age dating. Most tend to focus on ^7Be and ^{14}C given their well-defined source to the ocean. See Lal (1999) for a more comprehensive list of historical applications.
- 14) One major example for the application of cosmogenic radionuclides is the age and circulation patterns of the ocean. Our initial understanding of thermohaline circulation is derived from measurements of naturally occurring ^{14}C .
- 15) This now classic figure shows the age of waters at 3500 m as derived from ^{14}C abundance. In the broadest sense, ^{14}C enters the ocean via gas exchange in the form of CO_2 . Cold salty waters of the North Atlantic sink and form deep waters that then travel to the South Atlantic before moving through the Indian Ocean and into the Pacific Ocean. Once $^{14}\text{CO}_2$ is removed from the surface ocean and gas exchange ceases, the ^{14}C begins to decay. The relative age of the water mass since removal from the atmosphere is subsequently determined, providing information not only on the age of the water mass, but the pathway of travel.
- 16) In this second example, dissolved organic carbon (DOC), the humic component of DOC, dissolved inorganic carbon (DIC) and sinking and suspended particulate organic carbon (POC) were measured for ^{14}C activities with depth in the Sargasso Sea. This information provided some of the first evidence that DOC is comprised of organic matter that has cycled through the oceans several times prior to being removed (e.g., > 2000 years old). It further provided evidence that POC is young (contemporary) and turns over at a much faster rate. This pioneering work by Druffel et al. (1992) revolutionized our view of oceanic carbon cycling and is the basis for a multitude of studies that now use ^{14}C to date the age of specific biomarkers and compound classes. See Druffel et al. (2016) for more information.
- 17) ^7Be can be used to examine upper ocean mixing rates on short timescales. This information is critical for understanding how nutrients are injected into the sunlit upper waters of the water column. This process can vary significantly over space and time. This schematic depicts a simple model used to describe ^7Be profiles in the water column and to determine vertical mixing (defined as J in the equation, i.e., eddy diffusivity and upwelling) rates. Also see Kadko and Johns (2011) for more information.
- 18) In this example, ^7Be profiles were measured at stations in the eastern tropical South Pacific. ^7Be profiles were fit to an exponential below the base of the mixed layer using the previous model and vertical mixing (eddy diffusivity) estimates were made. The red line shows constant ^7Be activity in the mixed layer, according to the assumption made in the box model. Shown on each plot are upwelling velocity (W_H), eddy diffusivity (K_z) and uncertainty in K_z (σK_z). Uncertainties were propagated from the uncertainty in W_H and the depth attenuation coefficient (α) from the fit. Points in parentheses were not used for the fit. For more information on the model and application, see Haskell et al. (2015). Note that another emerging application of ^7Be that relies on ^7Be ocean inventories is as a tracer of atmospheric trace metal deposition to the ocean. In this application ^7Be ocean inventories are used in combination with the trace element to ^7Be ratio in bulk aerosols. See Kadko et al. (2015) for more information on this emerging technique.
- 19) The third class of radionuclides used in marine systems to explore a variety of processes is the anthropogenic radionuclides. Anthropogenic radionuclides are those radioactive nuclides that do not naturally occur on Earth in the present day and are created by humans. By far the largest

source of anthropogenic radioactivity was derived from atmospheric nuclear weapons testing that began in 1945 at the Trinity Test Site in New Mexico and ended in October, 1980 at the Lop Nor Chinese Test Site. The nuclear testing at Bikini Atoll encompassed 23 nuclear detonations between 1946 and 1958 (Richards et al., 2008). When the Paris fashion designer, Louis Réard, introduced his new women's swimsuit design in the summer of 1946, he called it the "bikini," and suggested that its revealing design was just as shocking as the Bikini Atoll atomic bomb tests that were capturing the public's attention that summer.

- 20) The two major sources of anthropogenic radionuclides to the environment are from nuclear weapons testing and nuclear facilities (e.g., nuclear reprocessing plants).
- 21) Why produce anthropogenic radionuclides? Nuclear Power. Anthropogenic radionuclides, particularly ^{233}U , ^{235}U , and ^{239}Pu produce energy during controlled fission chain reactions (See Lecture 1, Radioactivity in the Marine Environment: Understanding the Basics of Marine Radioactivity). Uranium enriched to concentrations above 0.7%, but less than 20% ^{235}U , is defined as low enriched Uranium (LEU).
- 22) Why produce anthropogenic radionuclides? Nuclear Weapons. Anthropogenic radionuclides, particularly ^{235}U also produce energy during uncontrolled fission chain reactions (See Lecture 1, Radioactivity in the Marine Environment: Understanding the Basics of Marine Radioactivity). Nuclear weapons use high enriched Uranium (HEU), which is comprised of more than 20% ^{235}U . Weapons-grade HEU is typically defined as 90% HEU or above. This minimizes weapons' size. Smaller and lighter nuclear weapons are much easier to deliver. The lower the enrichment level the greater the amount of material required to achieve a critical mass, i.e., enough mass to build a nuclear bomb. The figure shows the relationship between weight and yield. For more information, see: <http://tutorials.nti.org/nuclear-101/uranium-enrichment/#sthash.ZS4qbeWl.dpuf>.
- 23) Anthropogenic radionuclides are also useful for a wide variety of medical, scientific, and industrial applications, ranging from treatment of cancer to smoke detectors.
- 24) Major sources of *known* anthropogenic radionuclides to the ocean occur via atmospheric testing of nuclear weapons, planned liquid releases from nuclear fuel reprocessing plants, and accidents related to nuclear power production, satellite re-entry, and undetonated weapons. Data from IAEA (1999), IAEA (2001), and Hu et al. (2010).
- 25) Continuation of slide 24
- 26) Major sources of *potential* (poorly constrained) anthropogenic radionuclides to the ocean also include nuclear waste dumping and accidents/losses at sea. Data from IAEA (1999), IAEA (2001), and Hu et al. (2010).
- 27) This figure presents data on nuclear weapons testing from 1945-2007 by various countries as well as the location and size of the blast. Note that most of the tests occurred from 1945 – 1964 with large scale reductions in above ground testing occurring after the signing of the Partial Test Ban Treaty (PTBT) by the United States, the former Soviet Union, and the United Kingdom, on August 5, 1963. The PTBT banned detonations for military and civilian purposes underwater, in the atmosphere, and in outer space. Note that some nuclear tests continued to occur after the PTBT, with France conducting atmospheric tests until 1974 and China until 1980, however, the number and yield of these tests were significantly lower.
- 28) Atmospheric nuclear weapons testing led to temporal and spatial variability in global radioactive fallout or atmospheric deposition: 12% of the fallout was deposited near the test site ("close-

- in”), 10% percent was deposited around the same latitude as the test site (“intermediate”), and the remaining 78%, “global” fallout, deposited in the same hemisphere as the original nuclear test. Again, note the peak in the deposition of anthropogenically produced ^{90}Sr in 1963 and the large difference in fallout between the northern and southern hemispheres. ^{90}Sr was of particular interest as strontium substitutes for calcium and there was concern regarding radioactive ^{90}Sr contamination in milk produced by cows feeding on contaminated food.
- 29) A second major point source of anthropogenic radionuclides is from spent nuclear fuel reprocessing plants. One such example was the planned release of liquid effluent from the Sellafield nuclear fuel reprocessing and nuclear decommissioning site, United Kingdom, to the Irish Sea.
 - 30) A third major point source of anthropogenic radionuclides to the ocean is from nuclear accidents at both land and sea. Note that while this map includes a list of worldwide accidents, not all have led to releases to the environment. The most well-known environmental releases are those that occurred on April 26, 1986 (Chernobyl, Ukraine) and on March 11, 2011 (Fukushima Accident, Japan), which is not shown.
 - 31) When investigating accidents and planned releases from nuclear power plants, ^{137}Cs ($t_{1/2} = 30.1$ y) is often used as a basis for tracking radiation in marine systems as it is soluble in seawater, has a high seawater abundance, and has weak particle reactivity. Many of the other radionuclides released have relatively short half-lives. The 2011 data reflects the initial activity and the 2015 data reflects the remaining activity. In the case of the Fukushima accident, ^{85}Kr is the only released nuclide with a higher activity than ^{137}Cs four years after the accident. With a half-life of 10.8 y, ^{85}Kr decay occurs relatively quickly. Furthermore, Kr, is a noble gas that is unreactive and is rapidly diluted in the atmosphere. Finally, there is significant naturally occurring ^{85}Kr (from cosmic ray production); the major nuclear accidents of Chernobyl and Fukushima contributed only marginally to global ^{85}Kr inventories.
 - 32) As a result of the above information and the short half life of ^{137}Cs , ^{137}Cs activity distributions in the ocean are spatially heterogeneous, with highest activities in regions where nuclear accidents or planned discharges have occurred, e.g., the Irish Sea, the Baltic Sea, and the Black Sea.
 - 33) It is important to note that atmospheric weapons tests are still the largest source of ^{137}Cs to the oceans. Specific activities are low (Bq m^{-3}) due to the large volume of the ocean (dilution).
 - 34) Indeed, anthropogenic radionuclides are relatively low compared to that which naturally occurs in seawater, by orders of magnitude. **For comparison, a banana typically contains 10 Bq and humans typically have ~ 8,000 Bq.** It is important to note, however, that different radionuclides may have very different fates and relative toxicity (See Lecture 4, Radioactivity in the Marine Environment: Understanding the Basics of Radioecology).
 - 35) Applications of anthropogenic radionuclides in the ocean span from investigations of water/atmospheric mixing rates to dating the age of sediments.
 - 36) The focus of the remaining lecture will be on applied case studies. The first set of case studies focuses broadly on ocean circulation, the second set of case studies focuses on particle transport and burial.
 - 37) Circulation from atmospheric fallout (weapons tests). This example focuses on ^3H produced during atmospheric weapons testing ($t_{1/2} = 12.43$ y, beta decays to stable ^3He). ^3H enters the oceans predominantly via precipitation and continental run-off that occurs almost entirely in the northern hemisphere. Note that the units in this figure are tritium units ($1 \text{ TU} = 0.12 \text{ Bq L}^{-1}$).

Repeated measurements allows for a more detailed examination of water mass circulation and mean age (τ). For more information on ^3H as a circulation tracer, see the World Ocean Circulation Experiment (WOCE) website.

(http://www.ldeo.columbia.edu/res/fac/etg.tmp/text/woce_method.html)

- 38) Circulation from atmospheric fallout (weapons tests). This example focuses on ^{14}C produced during atmospheric weapons testing ($t_{1/2} = 5730$ y, beta decays to stable ^{14}N). Similar to cosmogenic produced ^{14}C , anthropogenic ^{14}C enters the oceans via gas exchange. In contrast to ^3H , the longer half-life of ^{14}C coupled with the different chemical reaction pathways resulted in a broader and more even distribution of bomb ^{14}C in the atmosphere. Thus, anthropogenic ^{14}C , in the form of gaseous $^{14}\text{CO}_2$, has been used to examine rates of gas exchange and penetration depths throughout the surface ocean. In this example, ^{14}C units are given as $\Delta^{14}\text{C}$ notation in ‰. This includes a variety of correction factors that are beyond the scope of this lecture (See Slide 12). Here ~ -60 ‰ is considered to be “pre-bomb” ^{14}C . For more information on sampling see the WOCE website (<http://woceatlas.ucsd.edu/>).
- 39) Ocean circulation patterns from point sources. There are two major centers for nuclear fuel reprocessing in northwestern Europe – COGEMA-AREVA NC-ORANO La Hague in France, which discharged into the English Channel (or La Manche), and BNFL Sellafield in United Kingdom, which discharged into the Irish Sea. At La Hague & Sellafield, discharges in the release of radionuclides to the environment have varied as a function of treatment methods and in response to increases in the throughput of fuel and/or type of fuel being reprocessed. This figure is an aerial photograph of Sellafield and shows the extent of the site, the location of some of the main spent fuel reprocessing facilities, and the pipeline through which liquid waste is discharged into Irish Sea along the seabed about 2 km offshore.
- 40) Radionuclide tracers (^{129}I , $t_{1/2} = 1.57 \times 10^7$ y) discharged from Sellafield and La Hague are transported northward through Fram Strait and the Barents Sea into the Arctic Ocean where they circulate at different depth levels, marking water of Atlantic origin. Shown here is the schematic flow from Sellafield and La Hague into the Arctic Ocean, its continuation at mid-depth along the continental margin, and the return flow toward Fram Strait along the Lomonosov and Mendeleev Ridges. The inset shows the history of ^{129}I discharges (TBq y^{-1}) from Sellafield (red line) and La Hague (blue line).
- 41) ^{129}I water-depth profiles in Arctic Ocean show the arrival of the large pulse produced in the 1990s from Sellafield and La Hague. Inset (lower right) shows the ^{129}I input function at 60°N . The arrival of the lower, pre-1993 signal (blue color) is indicated at each station by blue ^{129}I profiles. The arrival of the higher, post-1993 signal is indicated by red profiles. For example, ^{129}I activities at Station 4 (North Pole) are constant (blue color from 1994 - 1999, but begin to increase (red color) in 2000 and then further increase by 2007. This shows that the 1990s pulse in ^{129}I takes about 10 years to reach Station 4. For details on how to determine transit times see Smith et al. (2011). More recent work uses ^{236}U in a similar manner, e.g., Castillejo et al. (2018).
- 42) A similar figure can be made for ^{137}Cs . Note the difference between input functions of ^{137}Cs and ^{129}I . Although a different radionuclide with a different chemical reactivity and input function, ^{137}Cs shows a similar 10 year transit time to ^{129}I .
- 43) Ocean circulation from nuclear accidents. On March 11, 2011, a magnitude 9.0 earthquake initiated a tsunami off the coast off Japan that damaged the Fukushima Nuclear Power Plant in

Fukushima, Japan, releasing significant radioactivity into the environment. The activities shown in this figure are converging estimates updated from Buesseler et al. (2017).

- 44) Dissolved seawater ^{137}Cs activities have been continuously measured since the accident at several coastal locations. Within the first year, ^{137}Cs activities decreased from $\sim 10 \times 10^8 \text{ Bq m}^{-3}$ to $\sim 10 \times 10^4 \text{ Bq m}^{-3}$. Current Japanese limits are shown for comparison.
- 45) This animation from Rossi et al. (2013, 2014) shows the migration of ^{137}Cs across the Pacific Ocean.
- 46) The second set of case studies focuses on the use of anthropogenic radionuclides to examine particle transport, burial, and sediment geochronology.
- 47) In this figure, $^{239+240}\text{Pu}$ activities derived from atmospheric fallout has been repeatedly measured in the Northwest Pacific Ocean. Note the surface peak at depth and how it moves downward over time. It has been suggested that this sub-surface maximum is due to the scavenging of Pu onto sinking particles and subsequent remineralization by biological activity. Enhanced $^{239+240}\text{Pu}$ concentrations in deep bottom waters are explained by transport of Pu isotopes attached to larger particles that more rapidly sink through the water column to deep ocean sediments and are resuspended by near-bottom currents. For more information, see Lindahl et al. (2010).
- 48) Once in the sediments, anthropogenic radionuclides may be used to examine temporal and spatial variability of sediment burial as well as bioturbation. In this example, ^{137}Cs inventories, derived from the Fukushima accident, are plotted off the coast of Japan. Symbols denote the data source (see Buesseler et al. (2017) for more information). At the time of the compilation, ^{137}Cs inventories, although less than 1% ($130 \pm 60 \text{ TBq}$) of the initial ^{137}Cs activity released from the accident, was 5 – 10 times larger than the total inventory of Cs in the overlying waters. Rapid downward mixing of Cs-contaminated sediments by bioturbation helps to reduce surface Cs sediment concentrations and subsequent biological uptake at the sediment-water interface. However, bioturbation can also return contaminated sediments to the surface on timescales of years to decades.
- 49) In sediment cores with minimal mixing, measurement of anthropogenic radionuclides, such as ^{137}Cs , can be used as markers of specific date horizons. In this case, two peaks in sediment ^{137}Cs occur: 1) From the peak in weapons testing in 1965, and 2) from the Chernobyl Nuclear Accident in 1986 - note the one year delay between the accident and material buried at the seafloor. In this example, ^{210}Pb and ^{137}Cs are used to determine the relative sedimentation rate (see Lecture 2, Radioactivity in the Marine Environment: Uranium-Thorium Decay Series) in Pettaquamscutt River Basin sediments, Rhode Island, U.S.A. For more information, see Lima et al. (2005).
- 50) For more information on anthropogenic radionuclides in the ocean, see the IAEA MARiS Database on Anthropogenic Radionuclides. This searchable database includes over 170,000 radionuclide measurements in seawater, sediment, biota, and suspended matter and is continuously being updated. Profiles of a variety of radioactive elements are also available from the Geotraces website (www.eGeotraces.org)
- 51) In summary, 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 years.
- 52) Acknowledgements
- 53) References

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Useful Websites

IAEA MARiS Database for Anthropogenic Radionuclides: <http://maris.iaea.org>

eGeotraces. An online Electronic Atlas of Artificial and Naturally Occurring Radionuclides measured during the GEOTRACES Program: <http://www.egeotraces.org/>

Nuclear 101. Informative website on Nuclear Energy and Nuclear Weapons and how they work:

<http://tutorials.nti.org/nuclear-101/uranium-enrichment/#sthash.ZS4qbeWI.dpuf>

Our world in data: <https://ourworldindata.org/nuclear-weapons/>

A selected bibliography of anthropogenic radionuclides in the Marine Environment published by NOAA

in 2011: http://www.lib.noaa.gov/researchtools/subjectguides/marine_radionuclides.pdf

Online Nuclear and Radiochemistry teaching material:

<https://nucwik.wikispaces.com/Nuclear+and+Radiochemistry+Textbook+and+Compendia>

Rio5 Cookbook. For more information on how to measure specific radionuclides in seawater:

<https://cmer.whoi.edu/cookbook>

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