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## **RADIOACTIVITY IN THE MARINE ENVIRONMENT: UNDERSTANDING THE BASICS OF MARINE RADIOACTIVITY**

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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 the fundamentals of radiochemistry, including the origin and stability of elements, radioactive decay mechanisms, and the fundamental equations that govern radioactive decay. This lecture is the first of a four-part lecture series on radionuclides in the marine environment.

## SLIDES

- 1) Title Slide
- 2) The overarching goal of this lecture is to provide upper division undergraduate and beginning graduate students with a basic understanding of radioactivity. The lecture is split into three parts: 1) Understanding how elements form and why radionuclides occur in nature, 2) How radioactive elements release their energy and the more common modes of decay, and 3) The classic equations used to describe the radioactive decay process.
- 3) This slide provides some examples of how radiation is used in everyday life. Our focus will be on applications in marine science.
- 4) In order to understand radioactive decay, one must first remember the basic components of an atom and their properties. It is important to understand the difference between neutrons (no charge) and protons (positive (+) charge) and that they have significantly more mass than an electron (negative (-) charge). These properties define the various elements and contribute to the stability of that element. Isotopes have identical chemical properties and identical proton (Z) numbers, but differ in the number of neutrons (N), and thus, the atomic mass (A).
- 5) This periodic table of the elements contains a pie chart of the relative abundance of naturally occurring isotopes on Earth. This chart includes both stable and long-lived unstable elements. Note that this chart does NOT include relatively short-lived unstable elements (e.g., such as  $^{14}\text{C}$ ). The open circles are elements with no stable isotopes.
- 6) This Chart of the Nuclides shows all of the possible nuclei (each square) that exist or can be artificially created. The color coding depicts the half-life of each nuclide (the time it takes for 50% of the initial isotope to decay away). There are many more nuclides than what was shown in the previous figure, which focused on the dark red stable elements located in the center of the nuclide plot. Note that at higher atomic numbers, elements have more neutrons than protons. Radiation is an unstable element's mechanism of removing excess energy and allows the nucleus to change to a more stable configuration.
- 7) There are many types of radiation. Radiation is energy that occurs in the form of high speed particles or electromagnetic waves. This is based on the concept of wave-particle duality developed through the work of Max Planck, Albert Einstein, Louis de Broglie and others that states that every particle has both particle and wave like characteristics (see Slide 20). Not all radiation is the same. It can have both high and low energy and is characterized by a specific

wave length, which is inversely related to frequency (where wavelength ( $\lambda$ ) \* frequency ( $\nu$ ) = speed of light ( $c$ )).

- 8) Radiation can be ionizing (interacts with atoms and can cause the emission of an electron) or non-ionizing (there is not enough energy to cause electron emission). When discussing radioactive decay, we focus on the higher energy, ionizing radiation, which has enough energy to change normal cellular functioning. It is these particles that are emitted from an unstable or radioactive nuclide (see Slides 24-27).
- 9) It took a while to discover radioactivity and indeed, its discovery is often cited as one of the “Great Failures” in science that has led to a major discovery (another being the discovery of penicillin). Here, the photographic plate reflects a copper cross that was placed between the uranium ore and the photographic plate.
- 10) Now that we understand radioactivity, we need to learn more about why elements exist in nature in the first place and why one element is stable and another radioactive. This periodic table of the elements highlights elements in red that are most commonly used in the aquatic sciences.
- 11) Nuclear synthesis and the creation of elements began with the Big Bang and the formation of our universe. The first atoms did not form until well into the process.
- 12) Elements are formed in 2 major ways: Fusion and neutron capture. Fusion is in essence the crashing together of various protons, electrons, and other elements to make bigger elements. This process releases energy and is the source of heat in stars. As individual pieces get larger and larger, it takes more energy for those pieces, once they have collided, to form a new atom. Not all of these reactions release the same amount of energy, and the number of reactions that occurs decreases the larger the size of the piece (as there are simply fewer larger pieces to interact with one another).
- 13) This slide shows examples of hydrogen and carbon/nitrogen burning. The key is that each of these reactions release energy – but the energy is NOT the same for each reaction.
- 14) This slide shows the results of oxygen and silica burning. Note that P = proton here and not phosphorus ( $^{31}\text{P}$ ). Under silicon burning (2), the seven  $^4\text{He}$  (alpha particles) do not need to simultaneously combine with  $^{28}\text{Si}$  to form  $^{56}\text{Ni}$ .
- 15) Fusion eventually becomes increasingly difficult and there is not enough energy to keep the large scale reactions going. The probability of larger elements slamming together decreases. As such, the next process that occurs is neutron capture. This is a slower, more gradual process. Neutrons that are rapidly captured create isotopes of the initial element. Neutrons that are captured more slowly will decay into a proton prior to capture by the initial element and will result in the formation of a new element.
- 16) When all of these processes are put together, we can create a new periodic table that shows where each element is predominantly created. The colors denote how and where each element was formed. Cosmic Rays are particles that continuously bombard our planet and originate from the sun as well as originate from supernovas (see Lecture 3, Radioactivity in the Marine Environment: Cosmogenic and Anthropogenic Radionuclides). It is important to note that ALL elements have likely been created in one of these pathways, but the ones we now call “artificial” have decayed away since solar system formation and no longer naturally occur on Earth. This periodic table of the origin of the solar system elements is continuously being improved with new discoveries. For example, a recent study has found that gravitational waves

from colliding neutron stars are a significant source of heavy elements, such as gold, platinum, and uranium (Abbott et al., 2017).

- 17) Now that we understand why and how elements form, why are some elements stable and more abundant than others? Why are other elements unstable and emit radiation? Here we use the Liquid Drop Model of the nucleus first proposed by George Gamow and later further developed by Niels Bohr and John Wheeler. In this model, the nucleus of an atom is comprised of neutrons and protons that are held together by the nuclear force with interactions that are analogous to the structure of a spherical liquid drop, hence the name. While this model does not explain all of the interactions that occur within in a nucleus, it does enable a rough conceptual framework of the four main forces that contribute to a specific element's stability. The first is Spin Pairing. This is a positive force and uses the principle that all neutrons and protons have positive (+½) and negative (-½) spins that like to be paired within specific orbitals. The number of stable isotopes increases when both the number of protons and neutrons are even (e.g., fully paired). Indeed, there are only five elements (H, Li, B, N, and Ta) that exist with odd numbers of protons and neutrons.
- 18) The second force is Shell Binding or "Magic Numbers". As many students have learned in their introductory chemistry courses, electrons fill electronic shells (i.e., S, P, and D) in a specific order, with elements becoming increasingly inert once all the orbitals are filled (e.g., the noble gasses). The same is true for protons and neutrons. In particular, there are Magic Numbers of neutrons and protons which seem to be particularly stable: 2, 8, 20, 28, 50, 82, 126, etc. Nuclides that have both neutron and proton numbers equal to one of these magic numbers are called "Doubly Magic," and are even more stable.

The last two forces are negative forces in that they reduce nuclei stability. Surface Tension includes surface area and volume and is related to the number of either internal or surface nucleon (i.e., neutron or proton) interactions. In essence, the larger the volume, the larger the distance between neutrons and protons and the less they interact to maintain structure.

- 19) The last process is Coulomb Repulsion. Like charges repel, which means that the more protons that are forced into the nucleus, the greater the repulsive forces. This is why the neutron to proton ratio increases from  $\sim 1$  to  $\sim 1.6$  with increasing atomic number.
- 20) The Binding Energy reflects these four processes (Spin Pairing, Shell Binding, Surface Tension, and Coulomb Repulsion) and is the energy that would be required to dissemble the nucleus of an atom into its component parts. Remember back to the discussion of the formation of elements and the energy released during element formation? The Binding Energy is the energy released when you put together all of a nuclide's component parts. Ever notice that when you add the number of protons and neutrons together in an atom and compare it to what it actually weighs, you have excess mass? This change in mass, or mass defect ( $\Delta m$ ), is how one determines the energy released; using Einstein's famous equation  $E = \Delta mc^2$  and the concept of wave-particle duality (See Slide 7). Energy may also be released by combining two elements, and determined using the difference in mass between the initial and final elements. For example combining two lithium atoms to form boron will release energy.
- 21) A curve of Binding Energy per nucleon versus mass number shows the results of the four forces discussed in the previous slides. Note that He, C, O, and N are more abundant than their

- immediate neighbors due to the influence of Shell Binding and Spin Pairing. Larger atomic mass elements are impacted by increased Coulomb Repulsion and a reduction in Surface Tension.
- 22) The actual abundance of these elements on Earth is a function of both the Binding Energy *and* formation processes. Higher mass numbers are simply more difficult to form due to the reduced starting material and energy required to form them. This figure also shows the importance of some of these elements with regards to their use on Earth. Again note the impact of Shell Binding, Coulomb Repulsion, Spin pairing, etc.
  - 23) How do elements get rid of their “excess energy” to reach the most stable state and increase the Binding Energy per nucleon? They release energy in the form of radiation. The next several slides focus on the major forms of radiation or energy released.
  - 24) Alpha decay is the most energetic decay mechanism. An alpha particle consists of two protons and two neutrons and occurs in elements with Atomic Mass numbers  $> 82$ .
  - 25) Beta decay is the conversion of a neutron into a proton and the escape of an electron, which is also known as a beta particle. The examples listed are two common forms of beta decay.
  - 26) Unlike alpha particles, beta particles are emitted with a range of energies. This becomes important later for measurement techniques. Why is there a range? Beta decay also includes the emission of a neutrino. This results in the emission of several particle types with various energies that must be conserved. There is a maximum energy of emission,  $E_{\text{Max}}$ , which is typically used to describe the energy of the beta emission.
  - 27) Gamma emission is the most common form of energy release and is the conversion of nuclear energy to electromagnetic energy. Gamma emissions nearly always occur with alpha and beta decays and the energy of the gamma emission is specific to the radionuclide. Here,  $^7\text{Be}$  is an example of a radionuclide that is an excited state and emits its excess energy without changing atomic number.
  - 28) Each of these decay processes has different energies that influence their interaction with the surrounding environment. As such, we use a variety of materials as shielding to prevent those interactions from occurring. Understanding these interactions over the short and long term is critical for elucidating and describing radiation exposure by humans (see Lecture 4, Radioactivity in the Marine Environment: Understanding the Basics of Radioecology). In most environmental applications, shielding of various thicknesses is used to prevent external contamination from other naturally occurring radionuclides that may interfere with low level radioactivity analyses.
  - 29) While the focus is on alpha, beta, and gamma energies in this lecture, there are many forms of radiation that are emitted during radioactive decay.
  - 30) Unstable or radioactive elements will emit radiation in a variety of forms to reach a more stable state. Depending on the type of radiation emitted, there can be more than one resulting daughter element. Using nuclear physics and chemistry coupled with probability theory, we can describe these pathways of radioactive decay. In this example,  $^{40}\text{K}$  releases energy via two major pathways. The major decay pathway is via beta decay to  $^{40}\text{Ca}$  (89.3%). However,  $^{40}\text{K}$  will also decay to  $^{40}\text{Ar}$  via a combination of beta and gamma decays. Note that  $^{40}\text{Ca}$  contains even numbers of neutrons and protons (see above). Calcium and Ar have very different chemistries as highlighted by the animation. Argon is a gas, while Ca is biologically and chemically active and incorporated into bones.
  - 31) The decay schemes of radioactive elements are well described, including the modes, energies, and likelihood of decay patterns in the Chart of the Nuclides.

- 32) One last process to be considered in element formation and energy release is fission. The fission process (the nucleus of an atom splits into smaller, lighter components) produces a wide range of isotopes, including  $^{131}\text{I}$  and  $^{90}\text{Sr}$ . Note that while alpha decay may be defined as a fission process according to this definition, it is usually not treated as such. Recent work has shown that lightning bolts have enough energy that they can also ultimately create nuclear fission (Enoto et al., 2017)
- 33) The classic radioactive decay chains used in environmental science are the three U-Th series decay chains. This figure describes their decay patterns and chemical reactivity in marine systems. During element creation, there were other naturally occurring decay chains, such as the  $^{241}\text{Am}$ - $^{237}\text{Np}$  decay chain, that have since decayed away. The three series discussed in detail here only exist because the half lives of their parents are long enough to survive the age of the earth, although not much of  $^{235}\text{U}$  is left.
- 34) What are the timescales over which “radioactive decay” happens? Radioactive decay follows a Poisson distribution (discrete values), which mimics a Gaussian distribution (continuous distribution) for increasing numbers of decay events. With enough measurements, the probability that a decay event will occur can be predicted.
- 35) The rate of radioactive decay, or loss, can be described by the radioactive decay constant,  $\lambda$ . In essence, this constant is inversely related to the half life, which defines how much time it takes exactly 50% of the radioactive parent to decay to a daughter product.
- 36) As a result, radioactive decay can be used as a clock to trace the rate of a variety of processes.
- 37) The radioactive decay of an element, N, occurs as a linear loss rate described by the radioactive decay constant,  $\lambda (= \ln(2)/t_{1/2})$ .”
- 38) This example shows how this rate equation can be solved via integration to obtain the basic equation for radioactive decay.
- 39) This animation depicts how the radioactive decay process occurs for a parent decaying to a stable daughter. Note that the decay is instantaneous and random, but is still described by the radioactive decay equation shown in the previous slide
- 40) \*\*This slide could be skipped in the interest of time as this slide is just an algebraic reworking of initial equations. The accumulation of a stable daughter can be determined by rewriting the classic decay equation.
- 41) \*\*This slide could be skipped in the interest of time as this slide is just an algebraic reworking of initial equations. The time since the system was closed (no exchange between parent and daughter atoms with anything outside the system) can then be determined by measuring the number of parent and daughter atoms. Unfortunately, many of the closed systems that exist in environmental systems already contain daughter atoms that were incorporated prior to radioactive decay of the parent.
- 42) This slide describes what happens when the daughter atom is *also* radioactive, a common occurrence for radioactive elements such as the naturally occurring U-Th decay chain. For details on how to solve this equation, see Faure and Mensing (2004)
- 43) The following slides describe the evolution of parent and daughter products for the four cases that are encountered when the parent decays to a radioactive daughter.
- 44) Case 1:  $^{N1}t_{1/2} < ^{N2}t_{1/2}$ . The half life of the parent is shorter than that of the daughter.
- 45) Case 2:  $^{N1}t_{1/2} \sim ^{N2}t_{1/2}$ . The half lives of the parent and daughter are similar.

- 46) Case 3:  $N^1 t_{1/2} > N^2 t_{1/2}$ . The half life of the parent is longer than that of the daughter.  $^{228}\text{Ra}$ : $^{228}\text{Th}$  disequilibrium is often used in studies of submarine groundwater discharge. Note that as time increases, the activity ratio of the parent to daughter approaches a constant described by their radioactive decay constants.
- 47) Case 4:  $N^1 t_{1/2} \gg N^2 t_{1/2}$ . The half life of the parent is much longer than that of the daughter. This case is one that we commonly observe in environmental systems. Note the large scale difference in the *number* of atoms between  $^{238}\text{U}$  and  $^{234}\text{Th}$ . Why?  $^{238}\text{U}$  is relatively stable compared to  $^{234}\text{Th}$ , which is decaying at a much faster rate.
- 48) The naturally occurring radioactive decay chains of the U-Th series are an example of Case 4 radioactive decay, all have initial parents with very long half lives relative to their daughter radionuclides.
- 49) If a closed system is maintained for a sufficient period of time ( $t \gg 1/\lambda_2$ ), Case 4 allows one to greatly simplify the Bateman Equation solution and use a term called Secular Equilibrium, where the Activity of the longer lived radioactive parent, defined by  $\lambda_1 N_1$ , equals the activity of the radioactive daughter,  $\lambda_2 N_2$ , etc.
- 50) Activity has the added benefit of being the unit that is actually measured by many radiation detection systems.
- 51) Let's look at how the number of atoms versus activity compares for radioactive isotopes with short versus long half lives. Here we use an example of a sample containing  $1 \text{ dpm kg}^{-1}$  of  $^{232}\text{Th}$  and assume that the entire decay chain down to short lived  $^{224}\text{Ra}$  is in secular equilibrium. We then compare the activity and atom abundance of the two isotopes. It is easier to measure longer lived radionuclides by measuring the number of atoms (such as with a mass spectrometer). It is easier to measure short lived radionuclides (with low atom abundance) using their activity or radioactive decay (such as with an alpha, beta, or gamma detector). With the improvement of mass spectrometric techniques, the decision between the measurement of activity or mass is shifting to ever shorter half lives. Here, we define specific activity as the amount of activity per unit of mass.
- 52) We can rewrite Case 4 moving from atoms to activity.
- 53) This is the same figure as previous but now in units of Activity. The time scale of the approach to secular equilibrium is given by the radioactive decay constant of the daughter.
- 54) Using these equations and knowledge about a radionuclide's behavior in the environment, we now have all of the tools necessary to apply radioactive nuclides to a variety of questions. This is a schematic diagram of radioactive decay (horizontal arrows) and typical transport processes in the ocean and atmosphere.
- 55) In summary, radioisotopes are produced and decay in known and measureable pathways. The equations that describe these processes can be simplified to address a variety of marine environmental questions that are detailed in the following lectures.
- 56) Acknowledgements
- 57) References/Reading List for more information.

## References and Reading List

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### Useful Websites

NDT Resource Center provides general introduction to radiation and ionization: <https://www.nde-ed.org/EducationResources/HighSchool/Radiography/radioactivity.htm>

Excellent introductory text on Nuclear Chemistry Free online:  
[http://preparatorychemistry.com/Bishop\\_Book\\_18\\_eBook.pdf](http://preparatorychemistry.com/Bishop_Book_18_eBook.pdf)

Table of Isotopes: <https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html>

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

### Sources for in class activities:

Excellent source for demonstrations regarding various forms of radiation: Johnson A. (2013) Radiation and Atomic Literacy for Nonscientists. Science 342: 436-437.

Excellent in class exercise to describe half-life, probability, and data analysis:  
<http://sciencenetlinks.com/lessons/radioactive-decay-a-sweet-simulation-of-a-half-life/>

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