RADIOACTIVITY IN LAND, WATER AND ATMOSPHERE

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Summary

The Earth has always been radioactive and life has evolved in and partly because of the radioactive environment. Anthropogenic radioactivity has existed for less than a century but has revolutionized warfare and international diplomacy and made major contributions to science, medicine, industry and energy.

As a result of planned chronic releases and a few high profile accidents, man's activities have added marginally to the radioactive background dose received by all living things and more significantly by a few highly exposed individuals and hence to the associated health risks.

This article describes the nature of radioactivity and radiation, the rates and magnitudes of the mass and energy changes involved, the instruments used for detection and measurement and the biological effects of exposure.

The principal sources of natural radioactivity are set out including: primordial radionuclides which have their genesis in supernova explosions, cosmogenic radionuclides created by bombardment of the Earth by high energy particles from space and the radioactive decay products of both these primary sources.

The history and development of radioactivity is traced from its discovery in 1895 through its many medical, scientific, military and industrial applications in the twentieth century. The nature, magnitude and distribution of radioactive pollution from man's activities, principally nuclear weapons testing, nuclear power production and radioactive waste management are described. The natural reactor at Oklo and its significance for radioactive waste disposal are discussed

Finally, the current state of knowledge of the chemical and physical behavior of radioactivity, both natural and anthropogenic, in land, water and atmosphere is outlined. The hope is expressed that through an understanding of the real benefits and risks of applied radioactivity, a balanced judgment may be reached that will inform future related decisions affecting man's life support system, planet Earth.

1. Introduction

The world has always been radioactive. Formed from the dust of exploded stars (supernovae) four and a half billion years ago, some of the very long-lived radionuclides are still present in the Earth's core, mantle and crust today. Indeed without the input of heat, which accompanies radioactive decay, the core and mantle would long since have cooled and solidified, all tectonic plate movement, earthquakes and volcanism would have ceased and the Earth would be geologically as dead as the Moon.

Life on the planet has evolved in a radioactive environment. Almost certainly no human being in our present form would have been present today without it. Radiation is one of the causes of genetic mutation to which life is exposed. It is surely inconceivable, therefore, that radiation did not cause a significant number of the billions of mutations through which natural selection has proceeded over the last three billion years of evolution to produce all of today's living world including ourselves.

Since its discovery in the last decade of the nineteenth century, radioactivity has revolutionized international diplomacy through the creation of weapons of mass destruction. These were both the means of ending one world war and the determining factor in international power politics which has led to the avoidance of further global conflict thereafter.

Radioactivity has made a major impact on industry, science and medicine providing exploratory tools and diagnostics, therapy for cancer and alternative means for the mass production of that most fundamental of all raw materials, energy.

Through these activities humankind has added albeit very marginally to the natural background radiation. Nevertheless, perhaps because of its connection in people's minds with nuclear weapons, cancer and genetic mutation, radioactivity is perceived to carry a high risk. Radioactive contamination of the environment has become one of the most contentious of modern pollution issues.

It is not generally disputed that ionizing radiation is a hazard with the potential to inflict significant damage on biological material. Both the beneficial effects of controlled exposure in cancer therapy and the harmful effects of uncontrolled accidental exposure to large doses of radiation as at Chernobyl are well understood. It is the magnitude of the risk, real rather than perceived, that is less well understood.

For all the above reasons it is important to gain an appreciation of what radioactivity is, its discovery and early applications, its sources, both natural and man-made, its behavior in the environment and its biological effects. In this way a balanced judgment of the benefits and hazards, costs and risks of radioactive materials may better be made.

2. Radioactivity and Radiation

2.1. Discovery

Radioactive substances were used by humankind long before it was known what radioactivity was. The incandescent properties of thorium-cerium oxide were exploited

to increase the luminosity of the gas mantle in the 1880's and uranium oxide has been used to provide orange-yellow glass and ceramic glazes since Roman times.

Discovery came in an intense burst of scientific activity beginning in 1895 when Wilhelm Röntgen discovered an agent capable of penetrating paper, wood and metal and lighting up a fluorescent screen. He called this agent x-rays and produced the first x-ray photograph of the bones of the human hand.

The following year Antoine Henri Becquerel discovered rays emanating from uranium salts which were different from x-rays but would still penetrate black paper and create an image on a photographic plate.

In 1898 Marie Curie found that 'Becquerel rays' were emitted by both uranium and thorium minerals and postulated a new radioactive element in the uranium ore pitchblende. Later the same year Marie and her husband Pierre published two papers announcing the discovery of the elements polonium (named for Marie's native Poland) and radium (from the Latin 'radius' meaning ray).

Ernest Rutherford in 1899 distinguished between the two types of ionizing radiation emitted by uranium, the nature of which was not yet understood. He named the less penetrating rays '*alpha*' and the more penetrating '*beta*'. The following year he introduced the concept of radioactive half-life and measured this property for 'thorium emanation', which is now known to be an isotope of the gaseous element radon. In 1902 with Frederic Soddy, Rutherford concluded that 'radioactive elements must be undergoing spontaneous transformation'.

By 1909, this time working with T. Royds, Rutherford had identified the alpha particle (rather than ray) as doubly charged helium. In 1911, he proposed the existence of the atomic nucleus to explain the backscattering of bombarded alpha particles discovered by Hans Geiger and Ernest Marsden two years before. Meanwhile Soddy in 1913 introduced the term 'isotope' for atoms of the same nuclear charge but different mass.

The culmination of this phase of discovery came in 1919 when Rutherford proved that atoms were not indestructible by bombarding nitrogen with α particles to form oxygen and thereby realizing the alchemists' dream of the transmutation of the elements.

2.2. The Nature of Radiation

As Rutherford and Soddy discovered, radiation arises from the spontaneous transformation of the nucleus of the atom. Not all nuclei undergo this change. Thankfully most naturally occurring atoms are stable but those that are not, and the many more which are now artificially produced, lose mass or energy and emit radiation.

An element is defined by its atomic number (Z). This is the number of protons in every atom of this element. However, for every element there are atoms of different mass. These have different numbers of neutrons (N). Each individual form of atom having a

certain number of protons and neutrons is called a nuclide. All the nuclides of $Z = 6$ are carbon and are called isotopes of carbon. Those containing five, six, seven and eight neutrons are written thus: $^{11}_{6}C$, $^{12}_{6}C$, $^{13}_{6}C$, $^{14}_{6}C$. The mass number in the upper left is the sum of the protons and neutrons in the isotope. The charge on the nucleus is shown at the bottom left and equates to the number of protons. This may be omitted in an abbreviated form thus ^{14}C as it is implied by the chemical symbol of the element. $^{12}_{6}C$ and $^{13}_{6}C$ are stable but $^{11}_{6}C$ and $^{14}_{6}C$ are unstable and decay radioactively.

It is thus clearly important to have the correct ratio of protons to neutrons as both too high and too low a ratio lead to instability. A balance is necessary between the electrostatic repulsive effect of the protons and the attractive strong nuclear force between all nucleons. For the larger nuclei a surface effect also becomes significant.
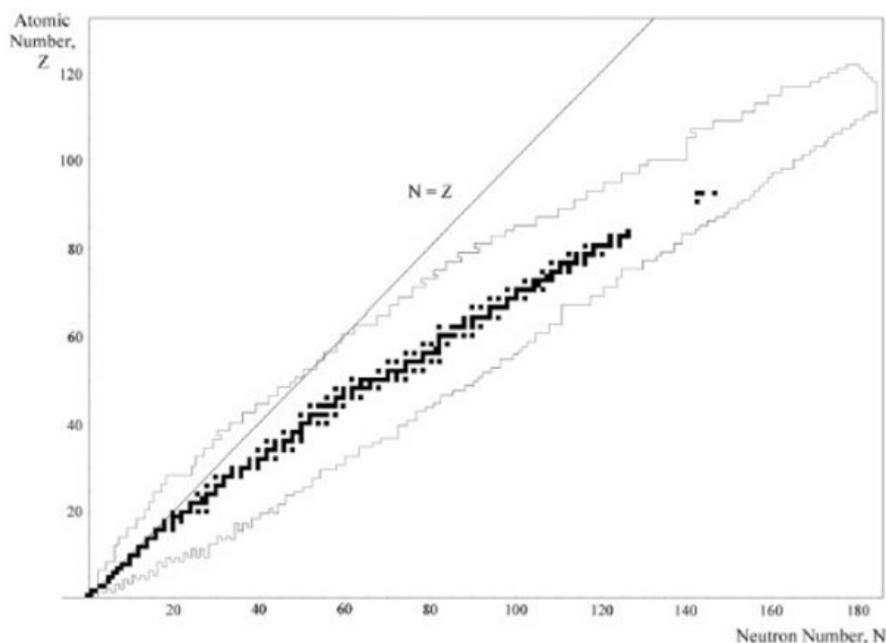
Figure 1. The Segrè chart of isotope neutron number (N) as a function of atomic number (Z). Adapted from Cottingham, W.N. and Greenwood, D.A. (2001), *An Introduction to Nuclear Physics*, Cambridge University Press, which uses data taken from *Chart of the Nuclides* (1977), General Electric Company, Schenectady.

A plot of neutron number against atomic number (see Figure 1.) for the known nuclei reveals the trend. Filled squares denote stable and long-lived naturally occurring nuclei. Neighboring nuclei are unstable: those for which data on masses and decay times are known fill the area bounded by the irregular lines.

A clear zone of stability is revealed. Below $Z = 20$ the zone follows the $N = Z$ line. At higher Z numbers a ratio of N to Z increasingly greater than unity is required for nuclides to be stable or to have lifetimes long enough to be detected. Above $^{209}_{83}Bi$ there are no stable isotopes, only long-lived radioisotopes such as $^{238}_{92}U$ and $^{232}_{90}Th$.

Even numbers of protons and neutrons convey high stability and elements with even numbers of protons often have many stable isotopes. Elements with odd numbers of protons e.g. $^{23}_{11}\text{Na}$ and $^{27}_{13}\text{Al}$ have only one stable isotope and two light elements $_{43}\text{Tc}$ and $_{61}\text{Pm}$ have no stable isotopes at all. Nuclides not in the zone of stability generally decay in such a way as to move towards the zone.

The most common emissions from radionuclides are alpha particles, beta particles and gamma rays. Yet other radionuclides decay through the capture by the nucleus of an electron from an inner shell followed by a cascade of in-falling electrons from outer shells accompanied by the emission of characteristic x-rays. Rarer decay modes include the emission of a positive electron (positron), and spontaneous splitting (fission) of the nucleus into two or three large fragments plus a small number of neutrons. On very rare occasions single neutrons or protons may be emitted alone or accompanying other modes of decay. After each radioactive decay the new nucleus may possess an excess of energy, this is usually released by emitting one or more gamma rays, characteristic of the particular radioactive nucleus. One or more X-rays may also be emitted as the orbiting_electrons adjust to the new charge on the nucleus; these electromagnetic radiations are not regarded as coming from the nuclear event itself.

Alpha (α) particles are identical to helium nuclei comprising two protons and two neutrons. They have a mass number of four and a charge of plus two. They are normally emitted by nuclei too heavy for the zone of stability. Their relatively large mass restricts their velocity to a maximum of about 10 percent of the speed of light and their high charge dislodges electrons from the atoms in the media through which they pass causing ionization. As a result the alpha particle quickly loses energy and is halted within a short distance. An example of alpha decay is:

$$^{226}_{88}\text{Ra} \quad \rightarrow \quad ^{222}_{86}\text{Rn} \quad + \quad ^{4}_{2}\text{He} \tag{1}$$

Beta (β) particles are electrons emitted from the nucleus as a product of the conversion of neutrons into protons. $^{1}_{0}n \quad \rightarrow \quad ^{1}_{1}p \quad + \quad ^{0}_{-1}e$ This allows nuclides with too high an N to Z ratio to move towards stability. Beta particles have minimal mass (1.36×10^{-4} of an alpha particle), a velocity around 40 percent of the speed of light and a charge of minus one. They are also ionizing but less so than alpha particles, are less easily stopped and are therefore more penetrating. An example of beta decay is:

$$^{90}_{38}\text{Sr} \quad \rightarrow \quad ^{90}_{39}\text{Y} \quad + \quad ^{0}_{-1}e \tag{2}$$

Gamma (γ) radiation is very short wavelength electromagnetic radiation and is highly energetic. It is massless, uncharged and travels at the speed of light. It is ionizing because of its energy, which can be transferred to electrons in atoms of the medium through which it passes. However, this process is less efficient than alpha and beta ionization and consequently gamma radiation is highly penetrating. Gamma radiation may occur on its own from an electronically excited nucleus

$$^{198}_{80}\text{Hg}* \quad \rightarrow \quad ^{198}_{80}\text{Hg} \quad + \quad \gamma \tag{3}$$

where * denotes the excited state of the nucleus or may accompany other types of radioactive decay

$$^{137}_{55}\text{Cs} \quad \rightarrow \quad ^{137}_{56}\text{Ba} \quad + \quad ^{0}_{-1}e \quad + \quad \gamma \tag{4}$$
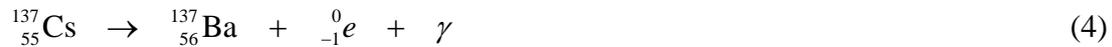
Electron Capture results in the conversion of a proton into a neutron $^{1}_{1}p + ^{0}_{-1}e \rightarrow ^{1}_{0}n$ and allows nuclides with low N to Z ratios to move towards stability. The nuclear decay event is normally accompanied by x-ray emissions which are less energetic and therefore less ionizing and less penetrating than γ rays. An example of Electron Capture is:

$$^{37}_{18}\text{Ar} \quad + \quad ^{0}_{-1}e \quad \rightarrow \quad ^{37}_{17}\text{Cl} \quad + \quad \text{x-rays} \tag{5}$$

In this case only x-rays are observed. Examples also exist in which both gamma rays and x-rays are produced.

Raising the N to Z ratio may also be achieved by the emission of a positive electron or positron (Positron Decay):

$$^{1}_{1}p \quad \rightarrow \quad ^{1}_{0}n \quad + \quad ^{0}_{1}e \tag{6}$$

This is an antiparticle with the same mass as the electron but opposite charge. An example of positron emission is:

$$^{8}_{5}\text{B} \quad \rightarrow \quad ^{8}_{4}\text{Be} \quad + \quad ^{0}_{1}e \tag{7}$$

It should be noted that in *β decay*, *Positron decay* and *Electron Capture*, a neutrino *v*, which escapes into space, is also involved in the reactions.

Some super heavy elements achieve accelerated size reduction by splitting (undergoing fission) spontaneously into two and occasionally three main fragments plus a small number of neutrons. The products of this violent process are not constant and a spectrum of isotopes of about sixty different elements is formed.

$$^{252}_{98}\text{Cf} \quad \rightarrow \quad \text{fission products and neutrons} \tag{8}$$
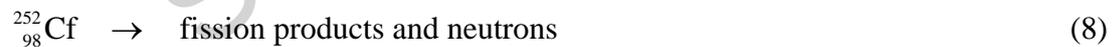
The spectrum has two peaks at around 100 and 150 mass numbers with a trough around 125. The products have very high N to Z ratios reflecting the high N to Z ratio in the parent atom and so decay rapidly and repeatedly by beta emission until stability is reached.

2.3. Rates of Decay

The radioactive decay process is spontaneous and random. It is impossible to determine at what time the disintegration of a particular nucleus will take place. It is possible,

however, to determine probabilistically how a population of nuclei will disintegrate with time. This rate of decay is independent of normal physical conditions such as temperature and pressure. It is first order kinetic and has activation energy of zero. If N is the number of atoms present at time t the rate of change of N is given by:

$$\frac{dN}{dt} = -\lambda N \tag{9}$$

where λ is the decay constant unique to the radionuclide concerned. Integrating this expression between times t_1 and t_2 gives:

$$N_2 = N_1 \exp\left[-\lambda\left(t_2 - t_1\right)\right] \tag{10}$$

where N_1 and N_2 are the numbers of atoms of the radionuclide present at times t_1 and t_2, respectively. Setting t_1 equal to zero then gives:

$$N = N_0 \exp\left(-\lambda t\right) \tag{11}$$

where N_0 is the number of atoms of the radionuclide at time zero and N the number at time t.

A useful parameter in charactering radionuclides is the half-life. This is the time after which only half of a population of N_0 nuclides remaining due to spontaneous disintegration, i.e. when $\dfrac{N}{N_0} = 0.5$, therefore from equation 11 above

$$0.5 = \exp\left(-\lambda t_{\frac{1}{2}}\right) \tag{12}$$

and therefore $t_{\frac{1}{2}} = 0.693/\lambda$ where $t_{\frac{1}{2}}$ is the half-life and substituting back into (11) gives

$$N = N_0 \exp\left(-\frac{0.693t}{t_{\frac{1}{2}}}\right) \tag{13}$$

which allows calculation of residual radioactivity at any elapsed time (t) knowing the original amount of radioactivity and the half-life. Many compilations of radionuclide half-lives exist. A few environmentally significant radionuclides with their half lives and modes of decay are listed in Table 1.

Radionuclide	Principal Decay Mode	Half-Life
^{3}H	β	12.3y
^{7}Be	Electron capture	53.3 d
^{14}C	β	5730y
^{40}K	β and γ	1.28×10^9 y

^{60}Co	β and γ	5.27 y
^{85}Kr	β and γ	10.8 y
^{87}Rb	β	4.75×10^{10} y
^{90}Sr	β	28.8y
^{99}Tc	β	2.11×10^{5} y
^{106}Ru	β	374 d
^{129}I	β and γ	1.57×10^{7} y
^{131}I	β and γ	8.04 d
^{137}Cs	β and γ	30.2y
^{220}Ru	α	55.6 s
^{222}Ru	α	3.82 d
^{226}Ra	α	1600y
^{232}Th	α	1.41×10^{10} y
^{235}U	α	7.04×10^{8} y
^{238}U	α	4.47×10^{9} y
^{239}Pu	α	2.41×10^{4} y
^{241}Am	α	432y

Table 1. Half lives and decay modes of some environmentally significant radionuclides

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Biographical Sketch

John V. Holder: During a successful career in environmental chemistry, John Holder has held senior posts in the British Scientific Civil service, the nuclear power industry and academia spanning 25 years and has held executive positions on British and European chemical society committees.

He was born in Manchester and obtained an Honors Degree in Chemistry (1969) and a Ph.D. in Minerals Engineering (1972) from the University of Birmingham. After a period on the faculty at Birmingham researching metals extraction, he moved in 1975 to the UK Atomic Energy Research Establishment at Harwell where he became a Senior Scientific Officer developing the Fast Breeder Reactor fuel cycle. In 1980 he joined British Nuclear Fuels where he led multidisciplinary teams in research and development of improved aerial, liquid and solid radioactive waste management technologies and decontamination and decommissioning of nuclear facilities. He was made a Fellow of the Royal Society of Chemistry in 1986 and chaired the RSC Environmental Chemistry Group from 1993 to 1995.

He was appointed Head of the Chemistry Department at the University of Central Lancashire in 1990 where he continued his research into radionuclide behavior in the environment and lectured in environmental chemistry, pollution and radioactive waste management. He held the post of Dean of Science in 1995.

Joining the Federation of European Chemical Societies Division of Chemistry and the Environment in 1995, he has been its Secretary since 1996.

He is currently Technical Director of the Centre for Environmental Research and Technology Limited and a Visiting Fellow at the Department of Forensic and Investigative Sciences, University of Central Lancaster, Preston, UK.

He has published and presented extensively in the field of environmental radioactivity.