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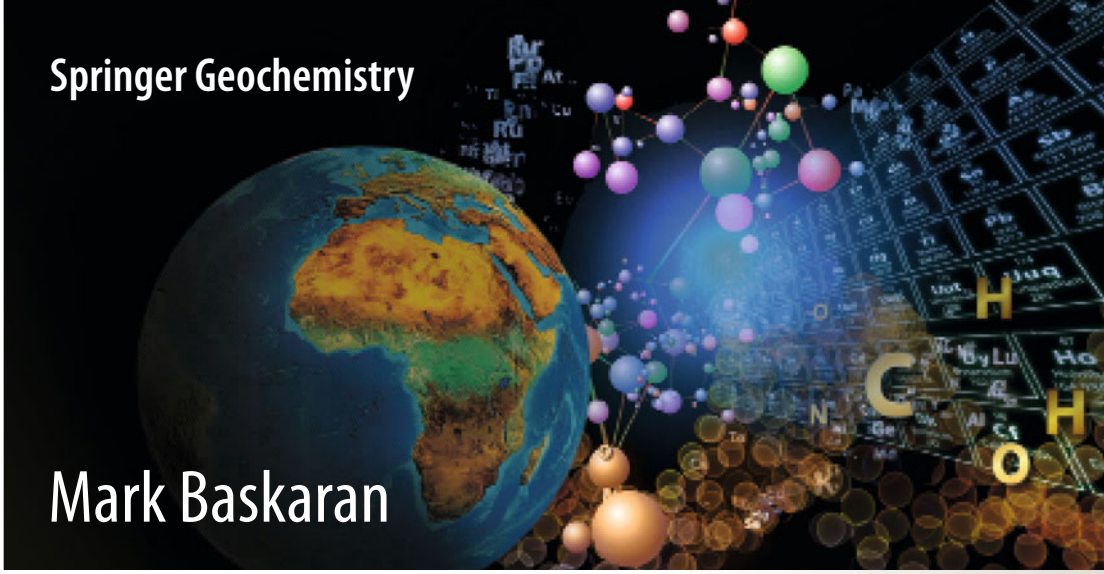

Mark Baskaran

Radon: A Tracer for Geological, Geophysical and Geochemical Studies

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To my beloved children:
Angelin, Justin and Gracelin
who always motivated me to work harder
and to carry on a passion for learning
for the generations to come

Foreword

A volume covering many roles of radon in the environment is a welcome addition to the geochemical literature. Radon is the maverick element of the noble gas clan. While there are shared familial characteristics, radon is governed by its own rules. It retains no memory of its history beyond a few times the meager half-lives of its natural isotopes, of a few days for ^{222}Rn and under a minute for the more fleeting ^{219}Rn and ^{220}Rn. These have concentrations that are typically maintained by their parent Ra isotopes, whose concentrations are in turn subservient to those of their U and Th progenitors. Radon will sometimes set off from the support of parental secular equilibrium, but this unsupported break is always short-lived. A radon atom can be set free from a solid when brought into existence during the decay that produces it along with a ^{4}He atom sibling, though these recoil from one another at birth, a violent process that projects them in opposite directions, sometimes right out of the solid. It is the subsequent travel that provides the opportunities for using Rn isotopes as a geochemical and geophysical tool, since the rate of decay of these liberated atoms provides a clock on their travel away from the site of production. The modes of transport reflect the chemical group trademarks. As a gas, it will prefer to be in the more mobile volatile, rather than liquid, phase. Being noble, it also generally disdains chemical interactions that can complicate travel. Consequently, radon escaping from minerals can be followed in groundwaters, into surface waters, and into the atmosphere, and following its path is how evidence is collected about radon sources and the flow of transporting waters and gases.

Professor Baskaran has made many valuable contributions to the field of U-series nuclides, and here he has assembled a systematic and comprehensive survey of the behaviour of radon in the environment. Of course, the factors that control harmful human exposure to radon have been extensively studied and reviewed elsewhere, but here the broader environmental occurrences of Rn are on display together. Those with interests in such disparate fields as hydrology, atmospheric processes, and geochemical exploration will discover the unique

insights that can be provided by radon and related short-lived nuclides. These will continue to constitute an essential part of the isotope geochemist's toolkit. This volume will serve both as a reference handbook and as an inspiration to new investigations for professionals as well as graduate students.

Don Porcelli
Department of Earth Sciences, Oxford University

Preface

Radon is the heaviest of all naturally occurring noble gases and it has a total of 36 isotopes, all of which are radioactive (ranging from ^{193}Rn to ^{228}Rn). Only three of these isotopes occur naturally and are constantly produced at a known rate in the ^{238}U-(^{222}Rn, radon), ^{235}U-(^{219}Rn, also known as actinon), and ^{232}Th-(^{220}Rn, also known as thoron) series. Radon has received the most attention among all the noble gases over the last four decades, largely due to the fact that it is the single major contributor to the ionizing radiation dose received by the general population. Radon is the second most frequent cause of lung cancer after smoking, and is classified as a human carcinogen. While the noble gas applications extend from cosmochemistry to different branches of geoscience, applications of radon are generally confined to near-surface Earth processes, due to its short mean life (5.53 days). Note that the presence of mantle ^{3}He in the oceans is indicative of diffusion of helium from much deeper sources inside the Earth, but sources of radon to Earth systems is much more confined to shallower depths. Most review articles and books such as *Noble Gases* by Minoru Ozima and Frank Podosek and the articles in the edited volume *Noble Gases in Geochemistry and Cosmochemistry* by Don Porcelli, Chris Ballentine and R. Wieler in Reviews in Mineralogy and Geochemistry serve as key reference works for the researchers working in noble gas geochemistry, but most of these publications have discounted the utility of radon as a tool to investigate several Earth surface processes. The sensitivity of radon measurement is the highest among all noble gases. In terms of molar concentration, 1 pico Curie (pCi) can be easily measured (1pCi of ^{222}Rn activity contains 1.76×10^{4} atoms which corresponds to 2.9×10^{-20} mole of radon). While there are a large number of scientific reports from national and international agencies (e.g., US Environmental Protection Agency, US National Academy of Science, World Health Organization) that describe findings on indoor radon in peer-reviewed journal articles, and many scientific reports pertaining to indoor radon as a health hazard, there is no single book on the applications of radon as a tracer in Earth systems. This book is designed to address that deficiency.

This volume is structured to examine the current body of knowledge in regard to all major aspects of the applications of radon as an environmental tracer. Each chapter is written as a review article, providing a critical synthesis with sufficient details for those who are technically trained but not necessarily working in this or similar area of research. This book is divided into 11 chapters. Chapter 1 is concerned with the physical, chemical, and nuclear properties of radon and the relationship between radon and its progeny. Chapter 2 gives an in-depth review of existing instrumentation for the measurements of radon and its progeny in environmental samples (water, air, and soil gas). The mechanisms of radon emanation and the factors that control the variations of the radon emanation coefficient are presented in Chap. 3. Radon has a simple emanation function from the Earth's surface, and the gradient in radon concentrations in the atmosphere as a function of latitude and longitude serves as a valuable tool to quantify factors and processes in the atmosphere that redistribute the radon released from the Earth's surface (Chap. 4). Progeny of radon, in particular ^{210}Po and ^{210}Pb, have been widely utilized as tracers to quantify a number of atmospheric processes that include source tracking and transport time scale of air masses, residence time and removal rate constant of aerosols and flux to and exchange between environmental systems of other gas species such as CH_4 and Hg^0 (Chap. 5). Radon concentration gradients at key interfaces such as sediment–water and air–water in aqueous systems (oceans, rivers, and lakes) have been utilized to determine exchange rates of gases across these interfaces (Chap. 6). The most widely used chronometer among the U-Th-series radionuclides is ^{210}Pb, which is a progeny of radon, with over 2,300 published articles over the past four decades. Its utility as a tracer and chronometer is presented in Chap. 7. Radon has a high solubility in organic liquids, similar to other noble gases, and hence its usefulness as a tracer for investigating the partitioning of radon between organic solvents and groundwater along with its application in quantifying infiltration of meteoric water and as a dating tool (in conjunction with ^{4}He) are given in Chap. 8. Radon as a tracer in geochemical exploration studies are presented in Chap. 9. In earthquake studies, radon is widely used as a precursor to predict earthquakes and several case studies are presented in Chap. 10. Finally, in Chap. 11, a summary of factors that affect radon entry indoor and potential techniques for radon entry prevention and mitigation are summarized. At the end of each chapter, future research direction is also included.

This book will prove to be a valuable resource for researchers in atmospheric science, marine and environmental sciences, earthquake studies, geochemical exploration studies (including uranium and hydrocarbon), organic pollutant studies in groundwater, noble gas geochemistry, and radon as a health hazard. Sections of the book are expected to be useful as a supplementary text to a number of graduate courses that include Environmental Isotope Geochemistry, Geochronology and Tracer Studies in Groundwater, Atmosphere and Ocean system.

Detroit, USA Mark Baskaran
July 2016

Acknowledgments

A significant portion of the writing of this book was done while on leave from Wayne State University, at East China Normal University as a Distinguished Visiting Professor in 2014–2016 and during a three-month visit in 2015 to Ege University as a Senior Fulbright Scholar. I thank the taxpayers of the United States, China, and Turkey, few of whom will ever know of this book but they contributed in indirect ways for this book to take this present form. I am grateful for long-term collaborations and friendships with several researchers that include Per Andersson (Sweden), Tom Bianchi (USA), Tom Church (USA), Jinzhou Du (China), Gi-Hoon Hong (S. Korea), Narekundi Karunakara (India), Michael Ketterer (USA), Don Porcelli (UK), Peter Santschi (USA), and Peter Swarzenski (USA). I am especially grateful for the inspiration and motivation over the years by senior researchers (late) Devendra Lal and Gerald J. Wasserburg and their constant support in my career. I am grateful for the efforts of the following scientists in reviewing one or more chapters of this book: Bill Burnett (Florida State University, USA), Doug Hammond (University of Southern California, USA), Scott Chambers (Australian Nuclear Science and Technology Organization, Australia), Y.S. Mayya (Indian Institute of Technology - Bombay, India), Guebuem Kim (Seoul National University, South Korea), and Peter Swarzenski (U.S. Geological Survey, USA). I thank Angelin Baskaran and Katie Krupp for outstanding editorial help with the manuscript. Ultimate responsibility for the contents, errors, and inaccurate statements lie with the author. Finally, I thank my wife Inthumathi Baskaran of our life journey together over the past three decades.

Contents

About the Author

Dr. Mark Baskaran is a tenured Full Professor in the Department of Geology at Wayne State University (Detroit, Michigan). He received his Ph.D. in Physics from Physical Research Laboratory (PRL), a premier research institution in India. After his Ph.D., he spent 2 years at PRL as a postdoctoral fellow before he moved to the Institute of Marine Science at the University of Alaska (Fairbanks, Alaska). After a year, he joined Texas A&M University (Galveston, Texas) where he taught introductory Physics and Geology courses at the Department of Marine Sciences, while conducting research related to atmospheric fluxes of radionuclides, mobility of radionuclides in groundwater, scavenging and particle cycling in marine environment and dating of recent sediments and carbonates. After his eleven-year career as a teacher and a researcher in Texas, he joined Wayne State University where he became a tenured Full Professor in 2007. He teaches both introductory level courses in Oceanography, Meteorology, and Physical Geology as well as upper level courses including Chemical Fate and Transport in the Environment, Nuclear Geology, and Environmental Geochemistry.

Professor Mark Baskaran has published over 130 peer-reviewed articles (with over 5,300 Google Scholar cumulative citations, h-index 43 in July 2016), most of which are related to the applications of isotopes as tracers and chronometers in Earth systems. He edited a two-volume Handbook entitled *Handbook of Environmental Isotope Geochemistry* with 40 articles contributed by eminent scholars in the field in 2011, published by Springer. He spent 3 months as a Senior *Fulbright Scholar* at Ege University (Izmir, Turkey) in 2015. He has given invited and plenary talks/seminars at over 60 national/international conferences, workshops, universities, and research institutions around the world.

Dr. Baskaran's research work includes all subsystems of the Earth system. Most of his work involved collaboration with a large number of researchers from universities and institutions around the world. His work with marine systems (estuarine, coastal, shelf and open-ocean) on the investigations of particular organic carbon export, particle cycling and remineralization, and colloidal thorium scavenging in the Arctic Ocean, Gulf of Mexico, North Atlantic, and East Pacific were funded by several funding agencies in the U.S. that include the National Science

Foundation (NSF), National Oceanic and Atmospheric Administration (NOAA), and the Department of Energy (DOE). His currently funded ongoing research is to investigate sedimentation and sediment dynamics in dams and other freshwater systems. He has been funded by NSF as a part of the U.S. GEOTRACES group in all four phases so far (Intercalibration, North Atlantic, East Pacific, and Western Arctic Ocean sections). He has served as a Chief Scientist in six major oceanographic expeditions in the Gulf of Mexico and Arctic Ocean.

He convened a National Workshop entitled *Recent Changes in the Biogeochemistry of the Great Lakes System* in March 2013 at Wayne State University. He also had convened a number of sessions and meetings at both national and international conferences and workshops.

Dr. Baskaran currently resides in Troy, Michigan with his wife (of 33 years) Inthumathi Baskaran. He has three grown-up children.

Chapter 1
Physical, Chemical and Nuclear Properties of Radon: An Introduction

1.1 Discovery of Radon

Radon is the heaviest of all noble gases, a class of chemically inert gases with low reactivity, and has a total of 36 isotopes ranging from ^{193}Rn to ^{228}Rn. All of the isotopes are radioactive, with three isotopes that are constantly produced from the ^{238}U (^{222}Rn), ^{235}U (^{219}Rn, also known as actinon, derived from its grandparent, ^{227}Ac) and ^{232}Th (^{220}Rn, also known as thoron, derived eventually from ^{232}Th). Only four of 36 radon isotopes have half-lives greater than 1 h. Radon was the last noble gas to be discovered by Friedrich Ernest Dorn, a German scientist in 1898, after the discoveries of four other radionuclides, uranium (U), thorium (Th), radium (Ra) and polonium (Po) by others. The first paper reporting the release of radioactive gas from the decay of radium was published in 1900 (Dorn 1900). One year prior to this publication, Rutherford placed thorium oxide inside a lead box and observed that the rate of leakage of the gold-leaf electroscope was much slower when the door was opened compared to when the door was closed. Based on this result, he concluded that thorium compounds continuously emit radioactive particles which retain their radioactive powers for several minutes and these results were published in 1900 (Rutherford 1900). At around this time, Pierre and Marie Pierre observed that the gas emitted from radium remained radioactive for a month, giving some indication on the half-life of ^{222}Rn (Curie and Curie 1899).

Radon belongs to group 18 (noble gases, p-block and period 6; [Xe] $4f^{14}$ $5d^{10}$ $6s^2$ $6p^2$) in the periodic table. Although noble gases are chemically inert, the interactions of these gases with other species are weaker (comparable to van der Waals type, with energy of bonding on the order of 1–3 eV, compared to ionic and

Throughout the book, three terms are synonymously used to represent the concentration of radon in environmental samples, as done in most published literature. Those are: concentration, activity, activity concentration, and specific activity. The units are: for fluids (liquid and gas): Bq L^{-1}/Bq m^{-3} or pCi L^{-1}/pCi m^{-3}; for solids: Bq g^{-1}/Bq kg^{-1} or pCi g^{-1}/pCi kg^{-1}

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Table 1.1 Mean atomic velocities (The mean velocity of a particle as a function of Temperature and mass are calculated from: $v_{mean} = (8KT/\pi m)^{1/2}$ where K is Boltzmann constant (1.3806×10^{-23} m^2 kg^{-1} s^{-2} K^{-1}), m is mass in kg, and T is temperature (absolute temperature scale)) at mean global temperature of 14 °C

Isotope	Velocity (m/s)
H	2,456
He	1,232
Ne	549
Ar	390
Kr	269
Xn	215
^{222}Rn	165

covalent bonding (on the order of 3–8 eV)) and the interactions are expected to be less complicated. There is a fair amount of literature that discusses the geochemistry of noble gases other than Rn (e.g., Ozima and Podosek 2002; Porcelli et al. 2002); however, data on the geochemistry of radon is limited. The three radon isotopes in the ^{238}U–^{235}U–^{232}Th series are commonly found in the environment. The molar abundance of these radon isotopes are the lowest among all the other noble gases, but there are several techniques available to measure such low abundance with counting instruments (Chap. 2). The geochemical factors that determine the distribution are similar to those of other noble gases and hence analogies with other noble gases are relevant to the discussion.

The average atomic or molecular velocities of noble gases for the average surface temperature on earth (14 °C or 287 °K) for radon and other noble gases are given in Table 1.1. The mean atomic velocity of radon, 165 m s^{-1}, is significantly lower than other noble gases, as expected.

1.2 Properties of Radon

1.2.1 Chemical Property

The atoms of radon and other noble gases have a closed-shell electronic structure and are extremely stable, as the ionization enthalpies are high. There are no ordinary electron-pair interactions among the noble gas atoms. The weak forces (van der Waals type or London type) are proportional to the polarizability and inversely proportional to the ionization of enthalpies of the atoms (Cotton et al. 1988). The first ionization energy (the energy needed to remove one electron from the outermost filled shell) for radon is 1,037 kJ/mol and it increases with decreasing atomic number for the other noble gases (Fig. 1.1a). The electronegativity in a column on the periodic table decreases with increasing atomic number and hence radon has a lower electronegativity (electronegativity = 2.2 Pauling scale, Table 1.2) compared

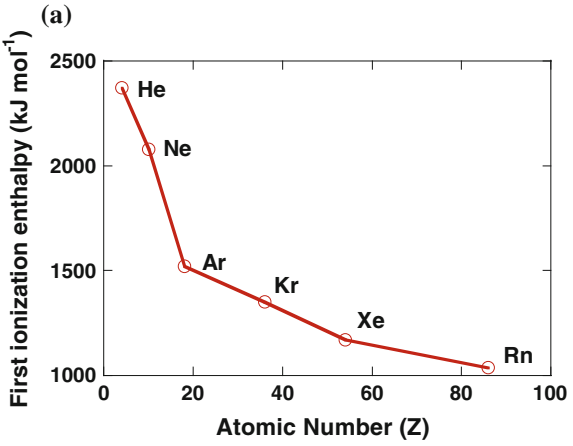

Fig. 1.1a First ionization energy (energy required to remove the most loosely held electron from one mole of noble gas ions each with a charge of +1) versus atomic number (data taken from Cotton et al. 1988)

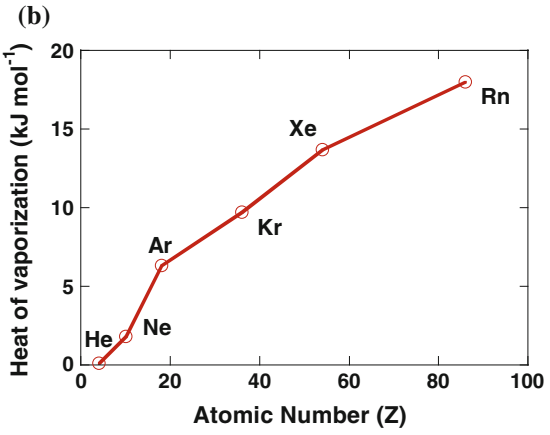

Fig. 1.1b Increase of heat vaporization with increase of the atomic number of noble gases—heat vaporization is the energy needed to overcome interatomic attractive forces (data taken from Cotton et al. 1988)

to the element one period before it, xenon (electronegativity = 2.60 Pauling scale) and thus radon is relatively more reactive than xenon. The solubility in a column also increases with increasing atomic number and hence radon is more soluble than xenon. Radon is also more soluble in organic liquids than in water. The heat of vaporization (measure of the work that must be done to overcome atomic attractive forces) also increases with atomic number (Fig. 1.1b).

Radon lies on the diagonal of the periodic table between the metals and non-metals and exhibits some of the characteristics of both groups, hence, it was suggested that radon can be classified as a metalloid element, along with boron, silicon, germanium, arsenic, antimony, tellurium, polonium and astatine (Stein 1987). Radon has been shown to react spontaneously at 25 °C or lower temperatures with fluorine, halogen fluorides (except IF_5) and a number of oxidizing salts (Stein 1987). Although there is no evidence for the existence of radon compounds or ions in aqueous solutions, solutions of cationic radon has been prepared in nonaqueous

Table 1.2 Atomic, physical and chemical properties of Radon

Property	Values
Atomic number	86
Standard atomic weight	222
Outer shell electron configuration	$6s^2 6p^6$
Density	9.73 kg m^{-3} (at 0 °C, 1.013×10^5 Pa)
Melting point (°K)	202
Normal boiling point (°K)	208.2
Heat of fusion (kJ mol^{-1})	3.247
Heat of vaporization (kJ mol^{-1})	18.0
First ionization enthalpy (kJ mol^{-1})	1037
Oxidation states	0, 2, 6
Electronegativity	2.2 (Pauling scale)
Covalent radius (nm)	0.150
van der Waals radius (nm)	0.220

Table 1.3 Nuclear and physical properties for ^{222}Rn, ^{220}Rn and ^{219}Rn

Parameter	Symbol	^{222}Rn	^{220}Rn	^{219}Rn
Half-life	$T_{1/2}$	3.823 d	55.83 s	3.983 s
Decay constant	λ	2.098×10^{-6} s^{-1}	1.242×10^{-2} s^{-1}	0.174 s^{-1}
Average recoil energy on formation	E_r	86 keV	103 keV	104 keV
Diffusion coefficient in air	D_a	1×10^{-5} m^2 s^{-1}	–	–
Diffusion coffficient in water	D_w	1×10^{-9} m^2 s^{-1}	–	–

solvents, such as hydrogen fluoride and halogen fluorides (Stein 1970, 1984). From a comparison of the known properties of krypton and xenon fluorides with that of radon, it has been deduced that radon forms a difluoride, RnF_2, and derivatives of the difluoride, such as $RnF^+SbF_6^-$, $RnF^+TaF_6^-$ and $RnF^+BiF_6^-$. Similar to KrF_2 and XeF_2, RnF_2 is reduced to elemental radon by water (implying that radon exists in +2 oxidation state in RnF_2 and is reduced to elemental Rn (0 oxidation number)):

$$RnF_2 + H_2O \rightarrow Rn + 1/2\,O_2 + 2HF \tag{1.1}$$

When heated to 400 °C with elemental fluorine, non-volatile radon difluoride is formed:

$$Rn + F_2 \underset{400\,^\circ C}{\rightarrow} RnF_2 \tag{1.2}$$

When Rn exists in an ionic state, it is able to displace ions of the first group elements (H^+, Na^+, K^+, Cs^+) and some of the second group elements (Ca^{2+}, Ba^{2+}). Quantitative removal of radon has been observed on a column packed with either

Nafion resins or complex salts. The distribution coefficient of cationic radon, K_d, in dilute BrF_3—trichlorotrifluoroethane solution was found to range from ~90 to 4000 L/kg (Stein 1987). The degree of partitioning of radon is a function of the radon partition coefficient between water and air, amounting to $K_{W/Air} = 0.2395$ at room temperature (296 °K) (Clever 1979). Wong et al. (1992) reported organic-carbon normalized sediment-water partition coefficient values of 21–25 on a suite of sediments from Boston Harbor, Charles River and Buzzards Bay in the United States. The octanol-water partition coefficient (K_{OW}) for radon is given as (Wong et al. 1992):

$$K_{OW} \text{ of } {}^{222}Rn = 32.4 \pm 1.5 \quad (@20\,^{\circ}C) \tag{1.3}$$

1.2.2 Sorption Behavior of Radon

The general (and expected) chemical sorption behavior of ^{222}Rn is that it is inert and does not react with other elements or often with compounds. It was assumed that radon did not interact with the surfaces of soil grains. Schery and Whittlestone (1989) reported isolated sorption of radon on to soil and rock grains under certain cases. However, due to the pervasiveness of moisture in moist climate, the contribution from desorption is likely limited compared to other major mechanisms of release of radon to the atmospheric air over much of the Earth's surface. From a comparison of the environmental conditions that exist on the moon with that on the earth's surface, Tanner (1980) concluded that for the dry, low-pressure environment of lunar rocks, sorption seems to be important, but the evidence for significant sorption at temperatures and humidity common at the earth's surface is conflicting.

1.2.3 Partitioning of Radon Between Gas and Aqueous Phases

As radon is a gas, its equilibrium partitioning between the gas and aqueous phases can be described by Henry's law, which is expressed as:

$$C_w = K\,C_a \tag{1.4}$$

where C_a represents radon concentration in the gas phase (Bq per m^3 of pore air), C_w is radon concentration in water (Bq per m^3 of water) and K is the dimensional partition coefficient, which is a function of temperature. The reported partition coefficient for radon between distilled water and air are 0.245 at 20 °C and 0.193 at 31.6 °C (Boyle 1911; Nazaroff 1992).

1.2.4 Physical Properties of Radon

The physical parameters of radon are summarized in Table 1.2. Atomic, physical and chemical properties of radon including density, melting point, heat of fusion, heat of vaporization, oxidation states, electronegativity and first ionization potential (energy needed to remove the outermost (highest energy) electron from a neutral atom in the gaseous state) are given in Table 1.2. The ionization potential of radon is also compared with other noble gases (Table 1.4).

Table 1.4 Comparison of atomic radius and ionization potential in all noble gases

Isotope	Atomic wt.	Atomic radius (Å)	Ionization potential
Rn	222	–	–
Xe	131.29	2.2	12.13
Kr	83.80	2.0	14.00
Ar	39.948	1.9	15.76
Ne	20.1797	1.6	21.56
He	4.002602	1.8	24.59

NM Not measured

Table 1.5 SI units and equivalents for traditional units

Parameter, SI unit	Conversion
Activity, Bq	1 Curie (Ci) = 3.7×10^{10} Bq
1 pico curie (pCi)	2.22 disintegration per minute, dpm
1 Bq	60 dpm (disintegrations per minute) or 1 disintegration per second, 1 dps
Concentration (Bq m^{-3})	1 pCi = 37 Bq m^{-3}
1 Potential alpha-energy concentration (PAEC, J m^{-3})	1 Working level (WL)[a] = 1.3×10^5 meV/L = 2.08×10^{-5} J m^{-3}
Equilibrium-equivalent decay product concentration (EEDC) for ^{222}Rn (Bq m^{-3})	1 Working level (PAEC) = 3,740 Bq m^{-3}
Exposure (J m^{-3} s)	1 Working level month (WLM) = 12.97 J m^{-3} s
Exposure (Bq m^{-3} y)	1 WLM = 73.9 Bq m^{-3} y^{-1}
Exposure rate (J m^{-3})	1 WLM/y = 4.11×10^{-7} J m^{-3}
Exposure rate (Bq m^{-3})	1 WLM/y = 73.9 Bq m^{-3}

The conversion factors for PAEC, EEDC, Exposure and Exposure rate are from Nero (1992f)

[a]In studies of radon-exposed miners radon progeny concentrations are generally expressed in terms of 'working levels' (WL). The working level is defined as any combination of the short-lived progeny in one liter of air that results in the ultimate release of 1.3×10^5 meV of alpha particle energy

1.2.5 Diffusion of Radon

From the published literature on radon transport, it is now established that transport of radon from soil to the atmosphere occurs predominantly by molecular diffusion (Schery et al. 1984; Nazaroff 1992). The activity flux density (J^d, Bq m^{-2} s^{-1}) resulting from random molecular motion is described by Fick's law which states that the radon flux density is linearly proportional to its concentration gradient:

$$J^d = -D_m \nabla C \tag{1.5}$$

where D_m is the molecular diffusion coefficient (m^2 s^{-1}) and ∇C is the gradient of radon activity concentration (Bq m^{-3}/m). The negative sign is to indicate that radon diffuses from higher to lower concentrations. The molecular diffusion coefficients in air (D_{ma}) and water (D_{mw}) are approximately 1×10^{-5} and 1×10^{-9} m^2 s^{-1}, respectively (Table 1.3).

The rate of radon movement in soil is expected to be slow compared to its movement in a homogenous medium such as pure air for two reasons: (i) the tortuous flow path around particles (tortuosity = τ) and (ii) relatively smaller fluid volume (porosity = n). Taking these two factors into consideration, the bulk radon flux density is given by:

$$J^d = -n_s \tau D_m \nabla C \tag{1.6}$$

where n_s is the porosity of the soil. Tortuosity (τ) is unity for a pure solution and is generally less than unity in soils. It was found to be 0.66 for closely packed uniform spheres. The terms D_m and τ are lumped together and are collectively known as the diffusion coefficient of the pore fluid such that

$$D = \tau D_m \tag{1.7}$$

Substituting for D from Eq. (1.7) in (1.6),

$$J^d = -n_s D \nabla C \tag{1.8}$$

The product of porosity and D can be represented by the notation D_e, the effective bulk diffusion coefficient of the soil. Note that D is directly related to the diffusion length of radon in the matrix and is a more fundamental parameter.

Radon diffusion coefficients in soil depend on the position of the ^{222}Rn location, which generally depends on the soil type, pore size distribution, water content and porosity. Since the molecular diffusion coefficient in air and water differ by a factor of 10^4, moisture effects dominate over other physical factors in the mobility of radon in soil. Earlier studies have shown that diffusion of radon is severely affected when soil moisture exceeds a certain threshold, which depends on the geometry of the soil pore space (Ishimori et al. 2013). The effective diffusion coefficient for radon in soil, comprised of mail tailings, silty sandy clay, compacted silt sands,

clayey sands, compacted inorganic clays, varved clays, mud, loams and alluvial-detrital deposits of granite, has been reported to vary between 2.2×10^{-10} m^2 s^{-1} (mud with 85 % water content) and 7.2×10^{-6} m^2 s^{-1} (mill tailings) (Nazaroff et al. 1992).

1.3 Decay Products of ^{222}Rn

Being an intermediate radioactive decay product in the ^{238}U (^{222}Rn), ^{235}U (^{219}Rn) and ^{232}Th (^{220}Rn) decay chain, radon undergoes radioactive decay (Fig. 1.2). The aggregate mean life of the daughter products of ^{222}Rn (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po) is 71.57 min while the corresponding aggregate mean-lives of the ^{220}Rn (^{216}Po, ^{212}Pb, ^{212}Bi, ^{212}Po) and ^{219}Rn (^{215}Po, ^{211}Pb, ^{211}Bi, ^{207}Tl) are 16.81 h and 1.03 h, respectively. The ^{238}U, ^{235}U and ^{232}Th series with the half-lives, decay mode, energy of emission (alpha, decay and gamma) and yield of the nuclear decay are given in Tables 1.6a, 1.6b and 1.6c.

Within a decay series, the evolution of the abundance of a daughter radioactive isotope is dependent upon its decay rate as well as production from its radioactive parent. Since the abundance of the parent is in turn dependent upon that of its parent, and so on up the decay chain, the systematics can become complicated, although for most applications simplifying circumstances can be found. The evolution of the abundances of isotopes within a decay series is described by the Bateman equations (Porcelli and Baskaran 2011). For a decay series starting from the long-lived parent N_1 and ending with a stable isotope S,

$$N_1 \rightarrow N_2 \rightarrow N_3 \rightarrow N_4 \rightarrow \ldots S \tag{1.9}$$

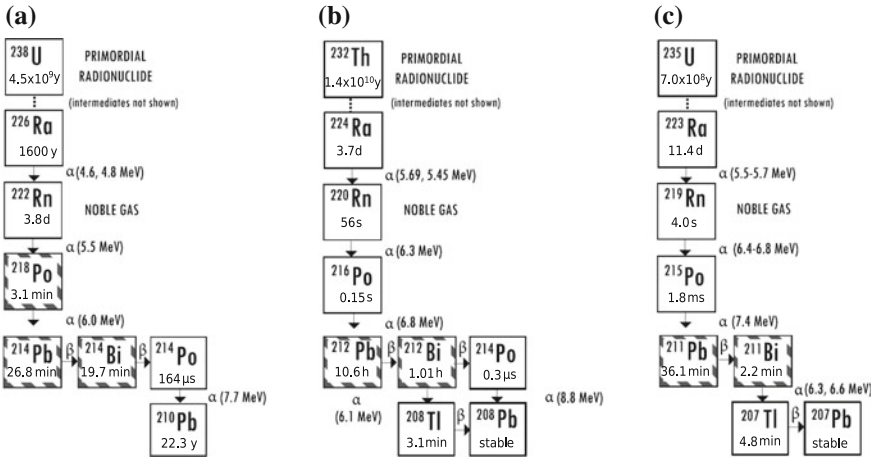

Fig. 1.2 ^{238}U, ^{232}Th and ^{235}U decay chains

Table 1.6a ^{238}U Decay Series—from ^{226}Ra (data taken from Firestone and Shirley 1999)

Isotope	Half-life	Decay	Emission energy (MeV)[a]	Yield (%)
^{226}Ra	1.6×10^3 a	α	4.784 (α_0)	94.55
–	–	–	4.601 (α_1)	5.45
–	–	γ	0.186	3.51
^{222}Rn	3.82 d	α	5.489 (α_0)	~100
^{218}Po	3.10 min	α	6.002 (α_0)	~100
^{214}Pb	26.8 min	β^-	1.023 (β_0)	~100
–	–	γ	0.3519	35.9
–	–	γ	0.2952	18.5
–	–	γ	0.2420	7.5
^{214}Bi	19.7 min	α	5.621 (α_0)	0.021
–	–	β^-	3.272	~100
–	–	γ	0.609	46.1
–	–	–	1.120	15.0
–	–	–	1.764	15.9
^{214}Po	164 μs	α	7.687 (α_0)	~100
^{210}Pb	22.3 y	β^-	0.0635 (β_0)	16
–	–	γ	0.0465	4.25
^{210}Bi	5.013 d	β^-	1.163 (β_0)	100
^{210}Po	138.38 d	α	5.304 (α_0)	100
^{206}Pb	Stable	–	–	–

[a]Energies of beta decay are maxima

Table 1.6b Isotopes of the ^{235}U Decay Series (from ^{223}Ra)—data taken from Firestone and Shirley (1999)

Isotope	Half-life	Decay	Emission energy (MeV)[a]	Yield (%)
^{223}Ra	11.435 d	α	5.539-5.747	96.8
–	–	γ	0.2695	13.7
^{219}Rn	3.96 s	α	6.819 (α_0)	79.4
–	–	–	6.553 (α_1)	12.9
–	–	–	6.425 (α_2)	7.6
^{215}Po	1.78 ms	α	7.386 (α_0)	100
^{211}Pb	36.1 min	β^-	1.373	100
^{211}Bi	2.14 min	α (99.72 %)	6.623	83.8
–	–	–	6.278	16.2
–	–	β^- (0.28 %)	0.579	100
–	–	γ	0.351	13.0
^{207}Tl	4.77 min	β^-	1..423	100
^{207}Pb	Stable	–	–	–

[a]Energies of beta decay are maxima