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Radon in Atmospheric Studies: A Review

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The distribution of the isotopes of radon in space and time, their physical characteristics, and their behavior in the dynamics of the atmosphere have presented challenges for many decades. ^{220}Rn , ^{222}Rn and their daughters furnish a unique set of tracers for the study of transport and mixing processes in the atmosphere. Appropriate applications of turbulent diffusion theory yield general agreement with measured profiles. Diurnal and seasonal variations follow patterns set by consideration of atmospheric stability. ^{222}Rn has been used successfully in recent studies of nocturnal drainage winds and cumulus convection. Good results have been obtained using ^{222}Rn and its long-lived ^{210}Pb daughter as tracers in the study of continent-to-ocean and ocean-to-continent air mass trajectories, ^{220}Rn (thoron) because of its short half-life of only 55 seconds has been used to measure turbulent diffusion within the first few meters of the earth's surface and to study the influence of meteorological variables on the rate of exhalation from the ground.

Radon daughters attach readily to atmospheric particulate matter which makes it possible to study these aerosols with respect to size spectra, attachment characteristics, removal by gravitation and precipitation, and residence times in the troposphere. The importance of ionization by radon and its daughters in the lower atmosphere and its effect on atmospheric electrical parameters is well known. Knowledge of the mobility and other characteristics of radon daughter ions has led to applications in the study of atmospheric electrical environments under fair weather and thunderstorm conditions and in the formation of condensation nuclei. The availability of increasingly sophisticated analytical tools and atmospheric measurement systems can be expected to add much to our understanding of radon and its daughters as trace components of the atmospheric environment in the years ahead.

Introduction

The occurrence of radon in the atmosphere has been known since soon after its original discovery in 1900. Yet, its distribution in space and time, its physical state and its mode of participation in the dynamics of the atmosphere are still not entirely understood. In many cases, however, the isotopes of radon furnish a unique set of tracers for the study of transport and mixing processes in the atmosphere. Nuclides belonging to the three naturally occurring radioactive series are found throughout the earth's crust in minute amounts. The radon isotopes are listed in Table 1 with their principal characteristics. ^{219}Rn is of minor importance in the atmosphere both because of its short half-life and the relatively low abundance of its long-

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lived parent, ^{232}Th is more abundant than ^{238}U , but the 55.6 s half-life of ^{220}Rn allows only a small fraction of that produced in the soil to escape into the atmosphere. Since radon atoms are chemically inert, they diffuse into the soil gas capillaries from mineral grains where they are formed. Molecular diffusion and atmospheric pressure changes are the major mechanisms in the escape of radon across the earth-air interface. Once in the free atmosphere, turbulent diffusion, convection, and wind motion are the chief transport modes.

Table 1. Isotopes of radon found in nature

Series	Long-lived Parent	Crustal Abundance* (ppm)	Radon Isotope	Common Name	Half-life	Particle and Energy (MeV)
Uranium	^{238}U	2.7	^{222}Rn	radon	3,824 day	α 5.49
Thorium	^{232}Th	8.5	^{220}Rn	thoron	55.6 sec	α 6.29
Actinium	^{228}U	0.02	^{219}Rn	actinon	3.96 sec	α 6.82

*Krauskopf, K. [1], other data from Walker et al. [2].

Because of the rapid decay of ^{220}Rn , it is found mainly within the first few meters of the earth's surface. The second daughter in the ^{220}Rn decay series is ^{212}Pb (10.6 hr). This nuclide because of its relatively long half-life is distributed much more uniformly with height, but at activity levels only a few percent of that of ^{222}Rn . The daughter nuclides of ^{222}Rn can be divided into a short-lived group which consists of isotopes of polonium, lead and bismuth all having half-lives of one-half hour or less and a long-lived group made up of ^{210}Pb (21 y) and its two immediate daughters, ^{210}Bi (5.0 d) and ^{210}Po (138 d).

The isotopes of radon exist as free atoms in the air. The daughter products may appear briefly as ions, and then become attached to aerosols. Their behavior in each case has been the subject of intensive study, yielding information both about the atmospheric part of the natural radiation environment and about the behavior of the atmosphere through a variety of tracer-type studies.

An earlier review of radon isotopes and their daughters in the atmosphere has been provided by Schumann [3].

Transport Processes

Any analytic treatment of the time and space distribution of radon and its daughters must take into account radioactive decay, transport by the winds, turbulent diffusion, sedimentation, precipitation scavenging, attachment, recombination, and ion migration due to natural electric fields. The resultant time-dependent differential equation has been treated by a number of investigators. Jacobi and Andre [4] were the first to provide numerical solutions for the vertical distribution of ^{220}Rn , ^{222}Rn and their daughter products. They used turbulent diffusion coefficients that varied with altitude and included a term for removal processes.

An excellent recent treatment of the general theory and a review of experimental results is given by Reiter [5]. He notes that the scales of motion appropriate to a given radioactive tracer, e.g., ^{220}Rn or ^{222}Rn , depend upon both a physical time scale determined by the radioactive decay constant and by a meteorological scale dependent upon the magnitude of the turbulent diffusion coefficient. The one-dimensional, steady-state solution of the diffusion equation shows that the concentration of uncharged radioactive particles at a given point depends only upon (1) the concentration at ground level, (2) the height above ground, (3) the decay constant of the radioactive tracer, (4) the turbulent diffusion coefficient, and (5) the rate of change of the latter with height.

A sinusoidal, diurnal time dependence was assumed by Staley [6] in deriving solutions for concentrations of ^{220}Rn , ^{222}Rn and their first three decay products. He found that ^{222}Rn concentrations are influenced mainly by turbulent diffusion in a manner similar to the diurnal oscillation of potential temperature. Malakhov et al. [7] show the effects of changes in radon exhalation, diurnal differences in turbulent diffusion and precipitation on disequilibrium among radon and its short-lived daughters. Birot et al. [8] applied the diffusion model to an infinite plane source, a half plane, and a line source and compared results with a radon source of these dimensions and measured meteorological parameters. Karol's [9] development is applied to large, synoptic-scale air mass transport within the atmospheric boundary layer, the upper troposphere and the stratosphere. Attention will now be directed to the results of the use of radon in atmospheric studies.

^{220}Rn (*Thoron*). An intensive study of ^{220}Rn in air near the ground was carried out by the Israels at Aachen [10, 11]. An ionization-type instrument was used in their work which was capable of continuously recording ^{220}Rn concentrations at four different heights. At the same time wind speeds and other meteorological information were obtained. A theoretical base is derived including analytical and numerical solutions to the diffusion equation and its boundary value problems. From simultaneous wind and thoron profiles in the interval from 1 to 7.5 m above ground and ^{220}Rn exhalation measurements at the surface, mass and momentum transfer in the first few metres above ground level were obtained. Their measurements also showed effects of wind speed and precipitation on ^{220}Rn exhalation.

Additional work with ^{220}Rn has been done by Crozier and Biles [12] in the United States; Ikebe and Shimo [13] in Japan; Bruilhet and Fontan [14] in France; Bakulin et al. [15], and Filistovich et al. [16] in the Soviet Union; and Israelsson et al. [17] in Sweden. Guedalia et al. [18] measured temperature, wind, ^{212}Pb and ^{222}Rn concentration profiles in the lower atmosphere (100 to 2000 m) and found reasonable agreement with an "advection" diffusion model.

Most of the use of ^{220}Rn has been in the study of turbulent diffusion and atmospheric electrical parameters within the first few metres of the earth's surface and in examination of meteorological influences on exhalation at

ground level. The unique combination of the short-lived ^{222}Rn which is found near the surface and the 10.6 h ^{212}Pb which controls the remainder of the thorium series provides a challenging opportunity for tracer applications.

^{222}Rn . This isotope has a half-life which when coupled with diffusion coefficients characteristic of ordinary thermally-induced turbulent diffusion, make it useful for study of a variety of diurnal mixing patterns and meso-scale systems. Some excellent early work on time-height variations using towers was done by Moses et al. [19], at the Argonne National Laboratory, by Fontan et al. [20], near Toulouse and by Cohen et al. [21] near Philadelphia. These observers found that atmospheric stability, as characterized by the potential temperature difference between the ground and a given level, was the single-most reliable indicator of radon concentration at a given height.

According to the theoretical treatments, ^{222}Rn concentration should decrease with an increase in altitude in approximately exponential fashion. This has been supported experimentally by Moore et al. [22] using aircraft sampling. A scale height of about 3 km over continental regions is typical. Guedalia et al. [23] have introduced acoustic sounder (sodar) techniques for measuring inversion levels which, when combined with ^{222}Rn concentrations at ground level, allow them to infer variations in mixing heights, vertical diffusion coefficients and the radon flux from the ground. This work shows excellent promise for the analysis of atmospheric stability patterns.

For purposes of this review further details concerning turbulent diffusion and vertical distribution will be omitted in favor of certain more recent studies.

Nocturnal Air Drainage. Inversion trapping of atmospheric trace gases and pollutants originating at or near the surface is a well-known phenomenon. This is especially noticeable in the case of ^{222}Rn originating from the soil. ^{222}Rn concentration increases during the night when the atmosphere is stable reaching a maximum at dawn. Less well known are the effects of mountain-valley air drainage on radon concentrations in the outflow system. Studies of this type have been made by Wilkening and Rust [24]. Measurements of ^{222}Rn concentrations near the mouth of a mountain canyon show diurnal changes consistent with the accepted model of canyon winds wherein surface air moving downslope at night accumulates radon, resulting in a steady increase in radon concentration and a decrease in air temperature due to radiation cooling of the slopes above. With the onset of up-canyon winds in the morning, the radon level near the mouth of the canyon drops rapidly to nearly one-tenth of the nocturnal maximum. The steady nighttime build-up can be interrupted by wind patterns that intercept the mountain at levels well below the mountaintop or by turbulent mixing induced by lee waves that increase temperature and decrease radon concentrations in the canyon air during the night.

A major effort to delineate flow patterns in mountainous terrain in the vicinity of extensive geothermal development is being carried out in the Geysers area of northern California [25]. In addition to point-source releases of tracers including sulfur hexafluoride, perfluorocarbon, heavy methane and fluorescent particles, ^{222}Rn is used as a dispersed-source tracer. Preliminary results indicate that ^{222}Rn can be used very effectively where tracing from an extended ground-level source is advantageous. Applications of this type will be of increasing interest as a wider range of energy sources is developed.

A unique case of nocturnal air drainage has been reported [26] where mountain gradient flow combined with land breezes interact with the trade winds on the windward side of the island of Hawaii. Downslope air in the land breeze has been traced 20 km upwind of the island by surface vessel and aircraft sampling.

Cumulus Convection. Radon is carried aloft by thermal updrafts associated with cumulus cloud development [27]. Radon sampling just below, above and around the perimeter of a developing cumulus congestus cloud demonstrates a positive input of ^{222}Rn into the cloud base and an excess within the cloud in contrast with ambient air outside. Further work is underway in New Mexico designed to include ^{222}Rn as a tracer in the study of entrainment, mixing, and outflow patterns during the growth of cumulus cloud systems.

Recent work along this same line has also been reported by Styro [28]. Radioactivity in cloud droplets as well as in cloud air was measured. Four different patterns of radioactivity versus height were observed. A change in distribution of cloud radioactivity with time was noted in their data also. A net flux of radon into the cloud bases was calculated from concentration measurements. With the increased sophistication of airborne instrumentation and radon sampling capability, more work in this area will be undertaken.

Continental and Marine Air Masses. It has been shown that relatively little radon escapes from ocean surfaces compared with the land areas of the world [29]. It is not surprising then to find that the diurnal and seasonal variations in radon and daughter concentrations are quite different in coastal regions from those occurring inland. Guedalia et al. [30] show that ^{222}Rn and ^{212}Pb concentrations increase with distance inland from the coast throughout the planetary boundary layer. Rama [31] used ^{222}Rn to study monsoon circulation and determine the continental component of monsoon air off the west coast of India. The work of Mishra et al. [32] using ^{222}Rn , its short-lived daughters ^{210}Pb and ^{212}Pb at both land and sea stations gives clear evidence of changes in seasonal cycles depending upon the degree of convective mixing and types of air masses present.

Measurements over the oceans have been used to identify radon rich air masses that have originated from continental areas. "Radonic storms"

were identified by Lambert et al. [33] as air masses over the Antarctic Ocean that exhibit "pulses" of higher than normal radon activity. Wilkniss et al. [34] have also measured radon and continental dust in the Antarctic. Larson and Bressan [35] used ^{222}Rn as an indicator of continental air masses over the North Atlantic, and were able to determine air mass boundaries over ocean areas as well. Prospero and Carlson [36] found ^{222}Rn and dust coming from the Sahara in a flight over the North Atlantic. A good correlation has been shown between ^{222}Rn and dust by a number of observers.

Moore et al. [37] found from a study of ^{222}Rn , ^{210}Pb , ^{210}Bi , ^{210}Po and their ratios that from two to three weeks has elapsed since radon in the trade winds at Hawaii had been over land, presumably the Asian continent. Tracer experiments of this type may have good potential for the identification of air mass trajectories over oceans.

Aerosols

Since ^{220}Rn and ^{222}Rn daughter products are isotopes of the heavy metals including bismuth, lead and polonium; they, unlike the parent atoms, attach readily to airborne particulate matter. These aerosols provide an important mechanism for the transport of radioactivity to man. The physical characteristics of the aerosol including settling velocity, diffusivity, inertia, and activity-size relationships are all of great interest. An important treatment of aerosols and radioactivity has been provided by Junge [38]; and a recent publication of the IAEA [39] gives a comprehensive review of particle-size distribution, gravitational and inertial separation of particles, electrostatic sizing devices, diffusion batteries, optional sizing techniques, and autoradiography. An important application of these naturally radioactive aerosols is in determining how long these particles remain in the atmosphere.

A critical study of this research has been made by Martell and Moore [40] who showed that residence times of aerosols in the troposphere have been over estimated in much of the earlier work. Since mean residence times are of the order of one week, it is to be expected that ^{210}Pb (21 y) and its daughters ^{210}Bi and ^{210}Po have played an important role in atmospheric tracing on a continental and even global scale [9]. The reader is referred to the extensive literature on radioactive aerosols for further details.

Atmospheric Electrical Properties

^{220}Rn , ^{222}Rn , and their daughter products are important to the study of atmospheric electricity not only because of their significant role in production of ion pairs in the lower atmosphere, but also for their use as tracers in the study of certain aspects of charge transport in fair weather and thunderstorm environments.

Radon-Daughter Ions. The ions of interest are the residual nuclei formed at the instant of decay of a radon or daughter atom. After they reach the end of their recoil range their most probable charge state is positive. They may then form ion complexes or recombine to form neutral atoms either before or after attachment to aerosols. The characteristics of these ions

have been reviewed by Bricard and Pradel [41]. They exhibit mobilities comparable to ordinary atmospheric small ions.

The terms affecting the concentration of the daughter ions in the atmosphere are (1) *formation* by decay of parent; (2) *removal* by radioactive decay, by attachment to condensation nuclei, by recombination with negative ions, or by precipitation scavenging, and (3) *transport* processes including convection, advection, eddy diffusion, sedimentation, and ion migration under the influence of electric fields. A detailed expression of the differential equation for concentration of short-lived ^{222}Rn daughter ions including these terms has been given by Roffman [42] together with appropriate numerical solutions.

Radon daughter ions of positive sign have been found to account for only about 30 ppm of the total positive, small-ion concentration in clean outdoor air, but simultaneous measurements of concentrations of the ^{222}Rn daughter ions and the total atmospheric small ions of the same sign yield correlation coefficients of 0.8 or better for fair weather conditions [43]. Further experiments have shown that the electrical conductivity of the atmosphere is directly proportional to the concentration of the total small-ion concentration at both mountain and valley sites in New Mexico. Hence, the validity for the use of radon-daughter ions as tracers in atmospheric electrical studies is confirmed.

A study of 40 thunderstorms at the Langmuir Laboratory in New Mexico clearly showed that both radon-daughter ions and positively-charged, atmospheric small ions were depleted near ground level under the influence of storm-produced electric fields. It was concluded from this work that apart from the major transport mechanisms of eddy diffusion and the vertical wind component which apply in both fair-weather and thunderstorm environments, ion migration is the dominant mechanism for the depletion ^{222}Rn daughter positive small ions near the ground in the presence of electric fields associated with thunderstorms. Large increases in condensation nuclei concentrations of the order of $50 \times 10^9 \text{ m}^{-3}$ or more can affect the daughter ion concentration but these situations are observed less frequently [42, 44].

Ion-Pair Production. During the radioactive decay of radon and its daughter products, alpha, beta, and gamma rays are given off. These radiations give up their energy by ionization and excitation of atoms and molecules which they encounter. A single alpha particle from the decay of ^{222}Rn will produce about 150,000 ion pairs with an average expenditure of 34 eV per pair. The complicated processes that are involved in the transformation of these ion pairs, or those created by other ionizing agents, into atmospheric small ions have been described by Mohnen [45]. ^{220}Rn , ^{222}Rn and their daughters in the atmosphere are the chief ionizing agents in the lower layers of the atmosphere, exceeding that due to cosmic radiation and radioactive substances in the ground [46]. The part played by the decay of radon and thoron daughters in the formation of condensation nuclei and other ion complexes has been reviewed by Vohra [47].

The effects of elevated levels of radon concentration on atmospheric electrical parameters has been demonstrated by Wilkening and Romero [48] in their work in the Carlsbad Caverns. They found that ^{222}Rn concentrations averaging about 65 pCi/l during the summer result in an ion-pair production rate that is about 200 times that which occurs in the outdoor air. As a result, the measured electrical conductivity and the positive and negative ion densities in the cave were found to exceed the values of these parameters reported for the free atmosphere by from two to three orders of magnitude, respectively.

Atmospheric electrical parameters are influenced in a major way due to ionization by ^{220}Rn , ^{222}Rn and their daughter products as noted in these examples.

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