



Conference of Radiation Control Program Directors, Inc.

November 2007

17th National Radon Conference

By Curt Hopkins, Radon Program Manager, CRCPD Office of Executive Director (OED)

on Sunday

Additional activities The 17th National Radon Conference was held September 9-12, in Jacksonville, Florida, with 128 attendees registered for the conference. There was an additional training session Sunday morning, State Radon Program Development Training, to help radon coordinators develop a quality radon program. Also, there was an ASTM E 1465-07a Comment Session in the afternoon.

organizations were represented

Many different → The categories of attendees are as follows:

- State radon programs had 46 representatives from 35 different programs
- State partners—26
- Tribes—7
- EPA—22
- Industry-11
- Other-16

a joint day with AARST

The first day was. The first day of the conference was a joint day with the American Association of Radon Scientists and Technologists (AARST), and the presentations were subjects of interest to both groups. On day two and three, separate sessions were held, with the ability to attend either session.

Zoo!

We went to the The night out activity was a visit to the Jacksonville Zoo and no reports have been received to date regarding anyone that was kept at the zoo! I suppose that is progress!

of critiqued items rated good or excellent

Greater than 90%

Responses on the critique sheets showed greater than a 90% rating of good to excellent on each of the meeting parameters. You can make next year's Radon Conference better by presenting a paper.

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National Radon Conference (Continued)

Vegas!

We are going to

Next year's meeting, the 18th National Radon Conference, will be held September 14-17, 2008, in Las Vegas, Nevada, at the Tuscany Suites Hotel, so pack your suits (bathing or otherwise) and join us!



Radon Consensus Dialogue Group Meets

Curt Hopkins (OED)

site of third Radon Consensus Dialogue group meeting

Jacksonville The third Radon Consensus Building Dialogue was held September 13th in Jacksonville, Florida, with representatives from State radon programs, CRCPD, the U.S. Environmental Protection Agency, the American Association of Radon Scientists and Technologists (AARST), the AARST Consortium on National Radon Standards, the National Environmental Health Association/National Radon Proficiency Program, and American Society for Testing and Materials. The Radon Consensus Building Dialogue was formed in January to improve understanding and reach agreements between the different segments of the radon community. The dialogues were facilitated by the Consensus Building Institute, Washington, D.C. The complete Jacksonville meeting summary can be viewed at <www.crcpd.org/ radon.asp>.



World History of Radon Research and Measurement from the Early 1900's

Submitted by A.C. George, Radon Testing Corporation of America

discovered in 1900, but effects had been noted for 300 years

Radon was → In 1900, Friedrich Dorn discovered the emanation in the uranium series that eventually became the well-known gas 222Rn. From 1900 through 1908, it was demonstrated that 222Rn is a radioactive gas found in tap water, highly condensable at low temperatures with a half-life of approximately 3.7 days, and can be collected on charcoal by adsorption. Although radon was discovered in 1900, the effects of prolonged exposure had been suspected and noted 300 years earlier among underground miners who developed lung cancer. During the period from 1924-1932, it was suggested that radon was the cause of high lung cancer incidence. In 1951, researchers at the University of Rochester New York pointed out that the lung cancer health hazard was from the alpha radiation dose delivered by the radon decay products that deposited in the respiratory tract. The findings of the BEIR VI Report, which was based

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World History of Radon Research and Measurement (Continued)

have changed from crude to fairly sophisticated measurement instruments

Detection methods

on epidemiological studies in different groups of mines in the 1950's and 1960's and on laboratory studies, showed that from 60,000 miners over 2,600 developed lung cancer where only 750 were expected.

> The epidemiological study conducted in Iowa in 1998 showed beyond any reasonable doubt that radon decay products cause lung cancer among women who lived at least 20 years in their homes. This paper will cover early radon measurements in soil, building material, ground water and in different air environments such as in the atmosphere, caves, spas, underground mines, and in residential indoor air environment.

> Radon measurements were conducted in many areas for diagnostic purposes. Radon was used as natural tracer to study air masses, vertical diffusion, and atmospheric studies, in earthquake prediction, and as a geological indicator for radium and uranium. In the early radon measurements, electroscopes, electrometers, and primitive ionization chambers were used for many years.

> In the 1940's fast pulse ionization chambers replaced total ionization chambers. From the mid 1950's onwards, a variety of radon measuring instruments were developed to assess the radon and radon decay product exposure to underground miners, workers at contaminated sites with uranium and radium tailings, and to the general public in residential buildings. In the last 20 years, new instruments and methods were developed to measure radon by using grab, integrating, and continuous modes of sampling. The most common are scintillation cell monitors, activated carbon collectors, electret ionization chambers, alpha track detectors, pulse and current ionization chambers, and solid-state alpha detectors.

The complete paper is provided as an insert at the end of this Bulletin, and it is on the CRCPD Website: <www.crcpd.org/radon.asp>.



<u>Seasonal Variations Found To Be Significant in Alabama Study</u>

By James L. McNees, CHP, and Susan H. Roberts, Alabama Radon Program

variations in short-term radon measurements are far more significant than implied in existing guidance documents

Seasonal → The results of an Alabama study published in the July 2007 edition of The Health Physics Journal indicate that seasonal variations in shortterm radon measurements are far more significant than implied in existing guidance documents. The Alabama Radon Program provided free test kits during January of 2006 to homeowners from Alabama's high radon areas that in prior summertime radon tests had tested below 4.0 pCi/l. The results revealed that 41% had wintertime test results greater than 4.0 pCi/l, 20% had wintertime test results greater than

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Seasonal Variations Significant in Alabama Study (Continued)

8.0 pCi/l, and 10% had wintertime test results of greater than 20 pCi/l. A summary of the study was displayed as a poster at the Jacksonville National Radon meeting.

Radon Program recommends that the testing protocols be revised

The Alabama → The Alabama Radon Program estimates that if a homeowner purchases a property during the summer air conditioning season in one of Alabama's high radon areas, having a properly conducted radon test as part of the transaction with results of less than 4.0 pCi/l, then later sells the property in the winter heating season to a buyer who requests a new radon test, that there will be at least a one in three chance that the wintertime test will have an unacceptable radon test result, thus requiring the seller to install a radon removal system to complete the sale. With the publication of these findings, the Alabama Radon Program recommends that the testing protocols be revised such that the report of short-term summertime radon test results of less than 4.0 pCi/l contain a recommendation that the radon test be repeated during the winter heating season.

Retesting was done in the same house in different seasons

This study utilized retesting in the same house in different seasons, which the authors believe is the only way to provide a full characterization of the incidence and magnitude of seasonal effects, something that a statistical study of one result per house from many different houses in different seasons cannot provide.

programs should retest their known summertime negative radon tests

Other State radon → On page 3-15 of EPA 400-R-92-0011, Technical Support Document for the 1992 Citizen's Guide to Radon, EPA makes the statement that "seasonal variability is greater in areas with long heating seasons." Because there are many areas of our country with longer heating seasons than Alabama, the Alabama Radon Program suggests that other State radon programs should retest their known summertime negative radon tests during subsequent wintertime heating seasons to determine the extent of this problem in their State.

2006 results were confirmed

The validity of the

→ To further validate our conclusions: during January of 2007 the Alabama Radon Program repeated the study by offering free radon tests to owners of houses that had tested less than 4.0 pCi/l during the 2006 summer air conditioning season. The yet to be published January 2007 results confirmed the validity of the published January 2006 results.

radon levels in the winter and low radon levels in the summer

Alabama has high → Based upon the findings of the 2004 Alabama Radon Variability Study, presented at the San Diego National Radon Meeting, the predominating pattern of seasonal variability in Alabama is high radon levels in the winter and low radon levels in the summer. To verify this, in July of this year, Alabama provided free radon test kits to a limited number of homeowners from Alabama's high radon areas that had wintertime radon results of less that 4.0 pCi/l. Those results will be available later this year.



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Changes on the E-25 Committee on Radon

By James McNees (AL), Environmental Nuclear Council Chairperson

working group that serves as the representative of the State radon programs

E-25 is the

The CRCPD has two working groups or committees that deal with radon and radon related issues. SR-R Radon Regulations, chaired by Bob Stilwell (ME) is a part of the Suggested State Regulations Council and is responsible for Part R: Radon of the Suggested State Regulations for Control of Radiation. The Committee on Radon (E-25), chaired by Bill Bell (MA), is the continuous working group that serves as the representative of the State radon programs as described by the charges given to the committee. Those charges are:

- 1. Review and comment on draft and existing publications.
- 2. To provide input to EPA on the National Advertising Campaign and other public awareness activities, as requested.
- 3. Provide a liaison to EPA to assist in the establishment of model national construction standards and techniques, as requested.
- 4. Assist EPA in the establishment of standards and policy on radon in water by:
 - a. Attending AWWA workshops/meetings;
 - b. Recommending to CRCPD that the AWWA's Proposed Questions for Consideration by the Radon Technical Group be distributed to radon coordinators as a "heads up" to the nature of the AWWA's activities:
 - c. Provide a committee member to a permanent position on the EPA committee to set guidance for implementation of the SDWAA standard: and
 - d. Meet with EPA decision makers to investigate issues related to important polices related SDWAA.
- 5. Assist EPA in developing guidance for States on the requirements of the SDWAA through our activities in Charge 4.
- 6. Plan the agenda and assist in the coordination of the National Radon Meeting.
- 7. Assist EPA with the development of Uniform Results Reporting Language, as requested.
- 8. To provide liaison for CRCPD with the American Association of Radon Scientists and Technologists (AARST).

Chairperson; Sarah Morgan (NE) and Clark Eldridge (FL) added to committee

In accordance with the CRCPD Board Policies and Procedures, Adrian Howe (NV), who had already graciously and excellently served well beyond the term limits for committee chairperson, was recently rotated off and replaced by Bill Bell (MA) as chairperson of E-25. Because there had been no membership rotation in 2006, Sara Morgan (NE) and Clark Eldredge (FL) were rotated from advisor status to full membership. This is done

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Changes on the Committee Radon (Continued)

both to provide opportunities for service to members wishing to serve and to provide a break to those who have provided years of faithful service.

Become an

advisor!

CRCPD members wishing to serve as members of these or any other CRCPD working groups or committees should first request to become an advisor on the committee. Advisors receive all committee related communications and are allowed to comment by e-mail or by letter on all business before the committee. Advisors are welcome to attend committee meetings but must do so at their own expense. Each year there is an annual rotation of committee membership, wherein one of the longest serving members is replace by one of the active advisors.



ELI Summary of Green Cleaning in Schools

Submitted by Tobi Bernstein, Environmental Law Institute

ELI document The Environmental Law Institute (ELI) has prepared a summary of selected written policies that promote green cleaning in schools. The document includes summaries of selected:

- State laws
- Unenacted (proposed) State legislation, and
- School district policies.

The document is available at: http://www2.eli.org/research/iaq.htm



Environmental Day at the Legislature

By John Stewart, Arizona Radiation Regulatory Agency

State lawmakers in Arizona were able to ask questions about radon and peruse the latest radon information publications when the legislators took their lunch break during the annual "Environmental Day at the Legislature" in Phoenix. Information booths and displays lined the sidewalk leading to the statehouse legislative chambers, providing easy access to reference materials and to experts who answered lawmakers' questions during one-on-one conversations. The radon information display was prepared and staffed by the non-profit Arizona Consumers Council, a radon coalition partner of the Arizona SIRG Program.

The Arizona Consumers Council also helps staff the radon information booth at the annual Arizona Home & Building Expo and sells home radon test kits priced at \$10, upon request http://azconsumerscouncil.org>.



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WORLD HISTORY OF RADON RESEARCH AND MEASUREMENT FROM THE EARLY 1900'S TO TODAY

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Abstract:

In 1900, Dorn discovered the emanation in the uranium series that eventually became the well-known gas ²²²Rn. From 1900 through 1908, it was demonstrated that ²²²Rn is a radioactive gas found in tap water, highly condensable at low temperatures with a half-life of approximately 3.7 days and can be collected on charcoal by adsorption. Although, radon was discovered in 1900, the effects of prolonged exposure had been suspected and noted 300 years earlier among underground miners who developed lung cancer. During the period from 1924-1932, it was suggested that radon was the cause of high lung cancer incidence. In 1951, researchers at the university of Rochester N.Y. pointed out that the lung cancer health hazard was from the alpha radiation dose delivered by the radon decay products that deposited in the respiratory tract. The findings of the BEIR Committee Report VI, which was based on epidemiological studies in different groups of mines in the 1950's and 1960's and on laboratory studies, showed that from 60,000 miners over 2,600 developed lung cancer where only 750 were expected.

Since 1998, the epidemiological study conducted in Iowa US, showed beyond any reasonable doubt that radon decay products cause lung cancer among women who lived at least twenty years in their homes. This paper will cover early radon measurements in soil, building material, ground water and in different air environments such as in the atmosphere, caves spas, underground mines and in residential indoor air environment. Radon measurements were conducted in many areas for diagnostic purposes. Radon was used as natural tracer to study air masses, vertical diffusion, and atmospheric studies, in earthquake prediction, and as a geological indicator for radium and uranium. In the early radon measurements, electroscopes, electrometers and primitive ionization chambers were used for many years. In the 1940's fast pulse ionization chambers replaced total ionization chambers. From the mid 1950's onwards a variety of radon measuring instruments were developed to assess the radon and radon decay product exposure to underground miners, workers at contaminated sites with uranium and radium tailings and to the general public in residential buildings. In the last twenty years, new instruments and methods were developed to measure radon by using grab, integrating and continuous modes of sampling. The most common are scintillation cell monitors, activated carbon collectors, electret ionization chambers, alpha track detectors, pulse and current ionization chambers and solid-state alpha detectors.

1. Introduction

Radon is a radioactive noble gas, having three naturally – occurring radioactive isotopes ²¹⁹Rn (Actinon, a by-product of the Actinium Series), ²²⁰Rn (Thoron, a by-product of the Thorium Series) and ²²²Rn (Radon, a by-product of the Uranium Series). Radon-222, being the most important radon isotope in terms of radiation exposure, is measured in different environments to determine its contribution to human radiation exposure. Rn-222 contributes about 55% of the annual radiation dose to the general population from all sources and 70% from natural radiation sources.

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In 1900, Frederick Dorn a German scientist found that radium-226 was giving off a gas which he called "radium emanation" known today as Rn-222. In 1908, Ramsey and Whytlow-Gray, isolated enough of the gas to study its physical properties and named it Niton (the shining one). In 1904, Mache [1,2], introduced the first unit for radon activity concentration and called it Mache-Enheit that was replaced in 1910 with the Curie. Mache used a galvanometer similar to the one used by Curie to measure the ionization current. One Mache (ME), corresponds to 13.5 Bq/L (364 pCi/L) and 37 Bq/M³ or (1 pCi/L) = 0.0028 ME

In 1901, Rutherford and Brooks [3] demonstrated that radon is a radioactive gas. Rutherford, in the same year discovered the active deposits of thorium and the Curies discovered the active deposits of radium. In 1902, Rutheford and Soddy characterized alpha particles when they worked on thorium compounds [4,5].

The Radon Health Effects of Underground Workers:

Although, radon was discovered in 1900, the effects of prolonged exposure had been noted about 300 years earlier. Georgius Agricola, a German physician and geologist (1494-1555), noted a high frequency of fatal lung diseases occurring among local miners. Paracelsus a Swiss physician and scientist (1493-1541) studied the diseases of underground miners in the Erz Mountains of Eastern Europe and found that many miners died from lung disease, concluding that dust and gases present in the mines were the cause of it. In 1879, two physicians Harting and Hesse, found that the mortality rate of uranium miners who worked in Germany and Czechoslovakia to be near 75% [6]. The German miners who worked more than 10 years developed the Erz Mountain disease which was later identified as lung cancer. In 1921, Margaret Uhlig, was the first to suggest that the radium emanation might well be the cause of lung cancer [7]. Three years later in 1924, Ludewig and Lorenser suggested that the lung cancers of these miners might be caused by the radon gas in the mines [8]. During 1924-1932, the finding of high concentrations of radon in the region and in the nearby mines of Joachimstal in Chechoslovakia and in radium mines of Schneeberg Germany, suggested that radon was the cause of the high lung cancer incidence. In 1932, Pirchan and Sikl, concluded that the radium emanation causes lung tumors among the miners at Jachymov. Over half the deaths were from lung cancer and most occurred among miners before they reached the age of 50.

The Findings of the Radon Health Effects of Underground Miners in the Last Fifty Years:

Research on radium and radon exposure began to shift to the other side of the Atlantic. Work on factory workers, medical patients, radium dial painters and the military. In 1944, Lorenz pointed out that radon itself, contributed a small percentage of the radiation Bale in (1951) [9] and Harley in (1952), were the first to point out that the lung cancer hazard was from the alpha dose delivered by the radon decay products when deposited in the respiratory tract. In 1956, Bale and Shapiro calculated the dose to the lungs from inhaled and deposited radon decay products making radon decay product concentration Measurements very important.

The hypothesis that radon decay products caused lung cancer was not accepted by some, until the findings of epidemiological studies of underground miners in the 1950's and 1960's. The findings of the BEIR Report VI, which was based on underground mine data and laboratory studies showed that from 60,000 miners who worked between 1941-1990, in different groups of mines in 8 countries, over 2,600 developed lung cancer where only 750 were expected [10]. From 20 epidemiological studies of underground miners, eleven studies provided quantitative information on the exposure response relationship between radon and lung cancer risk (Lubin et al.) [11-13]. In the past 5 years, the epidemiological study in Iowa US in particular, showed a high incidence of lung cancer among women exposed to high radon levels in residential buildings [14]. As a result of these studies, both in the underground mines and in residential studies support very strongly the characterization of radon concentration levels and the assessment of the radiation dose from radon found in different environments.

To calculate the radiation dose from radon and radon decay products, we must have knowledge of their concentration, particle size, respiratory deposition and clearance of the radon decay products and other factors such as lung morphometry and breathing characteristics. The exposure of underground miners was estimated using data of radon and radon decay product concentrations obtained in several US mines in Colorado, New Mexico. (Breslin et al.[15], George [16], George et.al.[17], and Knutson and George [18].and BEIR VI [10]).

Knowing the concentration of radon in most cases is adequate to estimate an upper limit for radiation exposure. From thousands of simultaneous measurements of radon and radon decay products in residential buildings the average equilibrium ratios were found to range between 0.4 to 0.5 a value that can be used to estimate exposure to radon and radon decay products. In the past 25 years, emphasis is on radon concentration measurements because of the simplicity, convenience and the cost effectiveness of radon measuring instruments and methods. More measurements of radon were done than of any other radioactive pollutant because measurements are easy and simple and there is a health concern and a market for it. In the past 20 years in the U.S alone, about 1 million indoor measurements of radon concentration are conducted on an annual basis

Before I begin to describe the radon measurement methods and techniques used in different applications and environments starting from 1900 to the present, I will highlight the history of radon in some chronological sequence. Some of the best research in natural radiation, was conducted in the early 1900's, by eminent scientific investigators who were worthy of a Nobel Prize in Physics or Chemistry. I consider that time, the classical era of scientific research with limited available resources.

Chronological History, Related to Radon Research and Measurement:

- 1597 Agricola first noted the high incidence of lung disease
- 1879. Harting and Hess. Described the autopsy findings on underground miners.
- 1898 The Curies and Schmidt discover radioactivity of thorium and the elements radium and polonium
- 1898 Rutherford discovers alpha and beta particles.
- 1899 Thomson and Rutherford demonstrate that radioactivity causes ionization.
- 1900 Dorn discovers the emanation (Radon), in the uranium series
- 1901 Rutherford and Brooks demonstrate that radon is a radioactive gas.
- 1902 Rutherford and Soddy discover transmutation.
- 1902 Elster and Geitel, made qualitative measurements of radon decay products on charged wire screens
- 1902 Thomson, discovers radon in tap water
- 1903 Rutherford and Soddy, noted that radon is highly condensable at low temperatures.
- 1903 Rutherford and Soddy measured the half-life of the radium emanation as 3.7 days, not too far from the 3.82 days, used today
- 1906 Rutherford, suggested the adsorption of radioactive emanations by charcoal.
- 1907 Eve, used charcoal to collect radon to measure in an electroscope.
- 1908 William Ramsey and R.W, Whytlaw-Gray, isolated enough radium emanation to study its properties and called "Nitens", meaning "the shining one".
- 1909 Satterly, used charcoal to collect and measure the concentration of radium emanation in the atmosphere.
- 1908 Ashman, used charcoal to determine the quantitative radium emanation in the atmosphere
- 1909 Regener, built a scintillation system and counted ²¹⁰Po
- 1913 Arnstein, identified squamous carcinoma of the lung in autopsy of a miner
- 1913 Rutherford, described the properties of the emanations and that they only emit alpha particles, they condense around –150°C and are strongly adsorbed by coconut charcoal.
- 1914 First use of radon for medical purposes
- 1925 First mention of radon in the literature. The name radon and symbol "Rn" was adapted for all isotopes of element 86.
- 1932 Evans, determined small quantities of radon and thoron using two ionization chambers
- 1943 Hess, wrote a paper on the radon content of the atmosphere and the radium content of river-water
- 1940's Casual link shown between radon and lung cancer.

Early Radon Measurements:

In general, in the early 1890's scientists had two basic instruments with which to detect the ions created from uranium and other radioactive materials. The earliest measurements of radon were made with various home-made electrometers which were nothing but ion chambers. They were the quadrant electrometer and the electroscope. They were primitive ion chambers that operated in a closed vessel filled with the sample to be tested. The next generation of ion chambers included electrometer tubes, dc amplifier and recorder. In 1902, Elster and Geitel, were the first to make measurements of radon decay products by collecting them on a charged wire [19]. By 1914 the quadrant electrometer became obsolete and the electroscope remained as the best instrument for many years. In 1908, Rutherford and Geiger using this type of instrument proved that the alpha particle was a helium nucleus. The method that Geiger developed for this work is the prelude to the so-called Geiger counter. Ionization chambers were commonly used until World War II.

In 1909, Regener built a counting system for the alpha particles emitted by Po-210. He used ZnS medium as the detector and counted the alpha particles through an optical microscope. Robley Evans [20] developed a two-chamber system that allowed subtraction of background by having one chamber filled with sealed air that responded to external gamma radiation

In 1943, Curtis and Davis [21], measured total ion current in a single chamber.. At the beginning of World War II, the most common methods available for the detection of alpha and beta radiation, were scintillation electroscopes and ionization chambers. At the start of the Manhattan Project, the standard for protecting workers against radium and radon were set at $0.1~\mu Ci$ and 370~Bq/M3, respectively.

By 1947, Davis [22] and the Health and Safety Laboratory (HASL) of the U.S. Atomic Energy Commission and later Fisenne and Keller [23], opted for fast pulse ionization chambers rather than total ionization. With the latest HASL version with a 2 liter electropolished stainless steel instead of brass chamber, the method could measure 2 Bq for an overnight count. The HASL, pulse ionization chambers were used extensively to measure very low concentrations of radon in different environments such as in the troposphere and stratosphere. From 1981-1996, the HASL pulse ionization chambers became the Standards Instruments for low concentrations of radon and served the calibration and intercomparison needs of the International community.

Since the mid 1950's many measurement methods and techniques for measuring radon were developed to meet the needs of research scientists and of those who made measurements in underground, occupational and indoor and outdoor environments. Today, there is a variety of instruments and techniques for making radon measurements using the grab sampling, integrating and continuous methods. Emphasis will be directed on the most commonly used instruments that serve today's needs.

Application of Radon and Radon Decay Product Measurements:

The major source of radon in the atmosphere is the normal emanation from the radium-226 in the earth's surface. The global emanation from soil is estimated at about 2 billion curies per year. The ground water potential is about 500 million curies per year (Harley and (NCRP Report No. 97) [24],[25] and [26]

Radon measurements were conducted in many areas and they were essential for diagnostic purposes such as sources of radon emanation rate from soil, building materials and ground water. In 1974, Clements and Wilkening [27] measured the effects of atmospheric pressure on radon transport across the earth air interface. Kojima [28], measured the exhalation of radon from soil to surface using a flow-through accumulator and an ionization chamber. Choubey [29], measured radon in soil and in spring water using scintillation cell detectors. The most popular method for measuring radon in water today is the liquid scintillation technique described by Pritchard [30] where the alpha particles from Po-218 and Po-214 interact with the scintillating material in an organic cocktail to produce photons that can be detected by a photomultiplier tube. Radon flux measurements were made by Wilkening [31] using the accumulation method and by Countess [32] using activated carbon accumulators. Hutter and Knuton [33], measured radon exhalation in soil gas by transferring the soil gas into scintillation cells. Radon in building materials was measured in many countries by placing intact or crushed building materials into accumulators and then transferring the emanated radon into scintillation cells for counting.

T. Gesell [34], did an extensive review of the background atmospheric radon concentrations outdoors and indoors by examining the sources, variation with time of the day, time of the year, altitude and geographic location. At areas with low concentrations of radon, usually radon was collected in chilled activated carbon collectors and subsequently the radon was transferred into scintillation cells or into very sensitive fast pulse ionization chambers such as the electropolished HASL chambers described by Fisenne and Keller [23]. Another method was to use high air flow through a large two-filter tube system and count continuously the alpha activity from Po-218 and Po-214 collected on the second (exit) filter [35]. Smaller two-filter tubes were used to obtain grab samples in uranium mines where the concentrations of radon were higher. The second filter was removed and was counted on a scintillation or solid-state alpha detector.

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Radon and radon decay product measurements were made near Geothermal plants using integrating devices employing thermoluminescence detectors [36]. The radiation doses from radon in natural gas from combustion during heating and cooking were assessed from measurements conducted in different countries with concentrations ranging from 40-54,000 Bq m⁻³. The highest concentration range was measured in Texas, US, [37].

6

Radon was measured in the prediction of earthquakes. Radon monitoring in groundwater, soil and atmosphere near seismic areas of the world are being used to predict earthquakes and active-fault sites [38]. Temporal anomalies were observed before and after the 1995 Kobe earthquake. The observed anomalies are presumably caused by changes due to micro fracturing in rock or migration of water from different sources. Also, co-seismic changes may be caused by seismic shaking and regional stress. The experience acquired in the past 25 years indicates that changes in ground water radon do occur prior to major earthquakes [39]. A continuous radon monitoring system with a solid-state detector is used to record data in the Chuko fault zone in Taiwan for earthquake prediction.

Many measurements of radon were made using it as a natural tracer for the study of air masses and vertical diffusion as a function of altitude ranging from near the ground to hundreds of meters and several kilometers. Hess and Schmidt were the first to calculate the vertical distribution of radon assuming vertical diffusion coefficients [40]. In more recent studies radon was used as a tracer to study vertical diffusion in the lower atmosphere [41]. Fisenne and others [42] measured radon in the stratosphere (4-20 km) in three North American locations using chilled activated charcoal traps. The trapped radon was flushed with formic gas into the 2-liter fast pulse ionization chambers for counting. Iida, [43], measured the vertical distribution of radon up to 5 km in the atmosphere in central Japan using grab samples in a 32 liter cylindrical vessel. The positively charged ²¹⁸Po was collected electrostatically on the electrode of an aluminized Mylar coated with ZnS(Ag) phosphor. The scintillations are detected by a photomultiplier tube. Radon decay products were used in the study of small radioactive ions in the atmosphere and in other times were converted to radon concentrations assuming equilibrium conditions between them.

In other applications radon was measured in caves, spas, geothermal sites. Radon concentrations were also measured as a geological indicator for radium and uranium. To estimate the radium body burden, radon was measured in exhaled breath.

A few studies of radon and radon decay products were mainly conducted in indoor environments in different countries. Hultqvist, probably was the first to make radon daughter concentration measurements in Sweden[44]. Steinhausler et al., [45] and George [46] made indoor radon daughter measurements in 1975 in Austria and the U.S. respectively. George and Breslin, made indoor radon and radon daughter measurements in the Metropolitan area of New York City during a three year period from 1975-1978, [47]. Stranden [48] and Swedjemark [49] conducted radon daughter measurements in dwellings in Norway and Sweden respectively. These small surveys were conducted to estimate the radiation exposure of the general population to radon and radon decay products. Since 1984, when high radon levels were found in Pennsylvania, millions of short-term measurements were conducted throughout the US alone. In Europe and other countries usually long-term measurements with alpha track detectors were made to characterize the indoor air radon environment.

Besides the world radon programs administered by various government agencies, a radon measurement and mitigation industry was established in the U.S. to address the health risk from radon and radon decay products.

The US EPA, in a national radon survey found that more than 6% of the US homes have radon levels above 150 Bq m $^{-3}$. More recent measurements by radon inspectors and by the radon industry professionals indicate that perhaps 10% of the homes in the US, have radon greater than 150 Bq/M 3 .

Several methods for measuring radon decay products were developed to characterize underground, outdoor and indoor environments. All methods are based on the collection of radon decay products on high efficiency filter media. The radioactivity collected on the filters is determined by gross alpha counting or by alpha spectrometry. The Kusnetz method [50] and the Rolle method [51] were very popular in underground mines. They provide the potential alpha energy concentration (PAEC) or the working level(WL) only. The concentration of the individual radon decay products and the associated PAEC are measured with more sophisticated methods [52-58].

Methods for the Measurement of Radon Decay Product Size Distribution

The size of the radon decay products is important because it affects the amount and site of deposition in the respiratory tract. The activity size distribution of radon decay products is a complex and difficult measurement. Measurements are undertaken by research groups and the availability of data is scarce. Today, there are two basic types of size classifying instruments developed in universities and in some governmental laboratories.. They are the diffusion battery system and the micro-orifice uniform deposit impactor (MOUDI). Diffusion batteries consist of the collimated disc type [59] and the wire screen system [60]. Both systems cover the range between 0.5-500 nanometers. Their resolution above 500 nm, is poor. The MOUDI system [61] measures sizes between 50-5000 nm. Since it does not provide information on the ultra-fine particles that are <50 nm, a graded screen array system is simultaneously used to complement the MOUDI data and cover the whole range from 0.5-5000 nm.

Radon Measurements in the Last Twenty Five Years

Today, emphasis is on radon gas measurements rather than radon decay products. Radon measurements are simpler more convenient and practical, providing a good estimate of the radiation exposure of the general public in timely fashion. The special relationship between the parent radon and its decay products expressed by the equilibrium ratio (ER), makes radon gas measurements more desirable. With the development of more sensitive measurement techniques, short-term radon measurements (2-7 days long) have become very popular in the U.S. The US EPA found that about 85%-90% of the short-term radon measurements are in close agreement with the long-term measurements. Short-term measurements are very practical and have become very useful in identifying the areas with elevated radon levels.

According to US EPA radon survey, which was conducted in the early 1990s, about 7% of the homes in the US have radon levels >150 Bq/m³ (4.0 pCi/L). Today, US home inspectors and radon professionals estimate that the number of homes with elevated radon is probably close to 10%. Moreover, most of the radon measurements in the US are driven by real estate transactions and relocations making short-term radon measurements even more desirable. In Europe, Asia and other countries most tests for radon are conducted with long-term alpha track detectors for the assessment of the annual average radon concentration. The selection of the appropriate measurement method and technique depends on the sensitivity of the method, cost and how fast results can be obtained. The most commonly used instruments for the characterization of the indoor radon environment since the mid 1970's till today, consist of the following two categories.

I. Passive integrating radon measurements:

I.

- 1. Activated carbon collectors of the open face (OF) or diffusion barrier type (DB) are used for 2-7 day-exposures. The canister type uses the gamma counting technique. The liquid scintillation vials (L/S) type, uses alpha and beta counting. About 70% of the radon measurements in the US, use passive open face or diffusion barrier activated carbon collectors [62]-[64].
- 2. Electret ion chambers used for 2-7 day-exposures measure the voltage reduction on the electret being proportional to the radon concentration. About 10-15% of the radon measurements in the US, use this detector technique [65]
- 3. Alpha track detectors are more appropriate for long-term radon measurements. They are used infrequently in the US. However, they are very popular everywhere else [66]. They were used in national surveys throughout Europe and Asia.

II. Passive or Active Continuous radon measurements:

- 1. Scintillation cell monitors mostly of the flow through type.
- 2. Current and pulse ionization chambers (mostly passive).
- 3. Solid State, passive or active if they use a pump to move air through the sensitive volume of the monitor.

The sensitivity and accuracy of the most recent developed radon measuring instruments and methods is summarized in table 1. In this report, I tried to emphasize instruments that are portable and practical to use in the field. Instruments developed for research purposes and those that I had no information about them were not included. Along with the sensitivities of the different instruments listed are the principle of detection and cost.

The cost of continuous radon monitors ranges from \$1,000-\$8,000, with the most popular, from \$1,000-\$4,500. The cost of individual passive radon collectors or detectors analyzed by private laboratories is about \$25.00.

The sensitivities of the different activated carbon collectors was determined from a 2-day radon exposures at 150 Bq M⁻³ (4 pCi/L). Analysis was performed 3 days after exposure. Activated carbon canisters were analyzed using gamma counting and the Liquid Scintillation LS vials using the alpha counting method. The sensitivities of different continuous radon monitors, alpha track detectors and the calibration factors of electret ion chambers were given by the manufacturer or cited from published literature. Instruments or methods with high sensitivity can achieve results with greater certainty. Table 1, shows that activated carbon collectors are the most sensitive and precise devices for measuring environmental radon for periods from 2-7 days, even if the radon concentration varies by as much as a factor of 10, and the humidity ranges from 25%-80% during the measurement [62, [63] and [64]. These collectors must be calibrated initially at different conditions of exposure duration, temperature and humidity to obtain the calibration factor. At environmental levels of radon, activated carbon collectors when analyzed yield the highest net counting rate at 150 Bq M⁻³ (4 pCi/L). Activated carbon collectors come in different configurations and sizes. When properly calibrated under different conditions of exposure, they can become a powerful tool in short-term radon measurements. The sensitivity of activated carbon collectors that use the gamma counting technique is roughly proportional to the mass of carbon in the collector. Their high sensitivity is due to the accumulation of radon in the collector, which is decreasing by the decay of radon only until counted. In contrast, continuous radon monitors detect radon that enters the sensitive volume of the monitor without the advantage of accumulation. Table 1, shows that activated carbon collectors at radon levels of 150 Bg m⁻³ are 15-50 times more sensitive than the most frequently used continuous radon monitors. For this reason, continuous radon monitors use the cumulative hourly counts instead of the minute counting intervals to improve counting statistics.

TABLE 1. RADIOSENSITIVITY AND ACCURACY OF RADON MEASURING INSTRUMENTS AND METHODS

| | Principle of | Sensitiv | vity | Cost of Device |
|--------------------------|------------------|----------------|-------------|----------------|
| <u>Instruments</u> | Detection | dps/150 Bq m-3 | cpm/4 pCi/L | Cost/exposure |
| RTCA, 50g, DB canister | Gamma | 1.50 | 90.0 | \$25.00 * |
| RTCA, 90g, DB canister | Gamma | 2.40 | 145.0 | \$25.00 * |
| RTCA, 2g DB (LS) | Alpha | 0.90 | 54.0 | \$25.00 * |
| RTCA, 90g OF canister | Gamma | 4.20 | 250.0 | \$25.00 * |
| PA, DER, 75g DB canister | Gamma | 1.00 | 60.0 | \$25.00 * |
| US EPA, 75g DB canister | Gamma | 0.80 | 48.0 | \$25.00 * |
| | | | | |
| Sun Nuclear 1028, CRM | Solid-State | 0.0030 | 0.18 | (\$700-1,500) |
| Sun Nuclear 1029, CRM | Solid –State | 0.0060 | 0.36 | \$1,000.00 |
| Rad Star RS500, CRM | Solid-State | 0.0050 | 0.30 | \$2,000.00 |
| Radon Scout, CRM | Solid-State | 0.0055 | 0.33 | \$1,000.00 |
| Durridge Rad7, CRM | Solid State | 0.0470 | 2.80 | \$5,800.00 |
| Femto CRM-510 LP | Pulse Ioniz. | 0.0200 | 1.20 | \$4,500.00 |
| RTCA E-Smart CRM | Current Ioniz | z. 0.0200 | 1.20 | \$2,250.00 |
| Radalink Aircat | Pulse ioniz. | 0.0280 | 1.70 | |
| Alpha Guard, CRM | Pulse Ioniz. | 0.0470 | 2.80 | \$6,000.00 |
| Active, Atmos 12D | Pulse Ioniz. | 0.0500 | 3.00 | \$5,000.00 |
| | | | | |
| Active Pylon CRM, AB-5 | Scint. Cell | 0.0950 | 5.70 | \$3,500.00 |
| Active DOE, CRM | Scint. Cell | 0.1400 | 8.40 | \$3,000.00 |
| Active Eberline, CRM | Scint. Cell | 0.4000 | 24.00 | \$6,000.00 |

^{*} Cost per test

In Table 2 and Table 3 listed are the sensitivities of alpha track detectors and the calibration factors for electret ion chambers respectively. Although these latter two types of radon detectors cannot be compared directly with the devices listed in Table 1 in terms of counts per150 Bq m⁻³, the number of tracks per cm² per unit time and the calibration factors are useful in selecting the appropriate device for a specific measurement application. Information on these techniques for radon testing was obtained from the literature and from private communications with the developers and users of some of these devices. Alpha track detectors with larger sensitive volumes exhibit the highest sensitivity.

At this time, I was able to locate two manufacturers of electret ion chambers (both in the US) for short-term and long-term radon exposures. The two types of electret ion chambers have similar sensitivities and can be used for short-term or long-term exposures depending on the thickness of the electret.

TABLE 2. SENSITIVITES OF ALPHA TRACK DETECTORS AND ELECTRET ION CHAMBERS

| | Sensitivity –Tracks cm ⁻² | | |
|----------------------------|--|---|--|
| Device Id. and Type | per 150 Bq m ⁻³ h ⁻¹ | per 4 pCi L ⁻¹ d ⁻¹ | |
| | | | |
| Landauer CR-39 (closed) | 0.07 | 1.70 | |
| Germany Makrofol (closed) | 0.15 | 3.60 | |
| Japanese Makrofol (closed) | 0.19 | 4.50 | |
| RSSI CR-39 (closed) | 0.23 | 5.60 | |
| Swedish Cr-39 (closed) | 0.39 | 9.40 | |
| NYU CR-39 (closed) | 0.48 | 11.50 | |
| French LR-115 (open) | (0.20 - 0.60) | (4.80-14.40) | |
| Gamma Data CR-39 (closed) | (0.33-0.66) | (7.90-15.80) | |
| Italian LR-115 (closed) | 0.92 | 22.00 | |
| Slovenia CR-39 (closed) | 0.97 | 23.40 | |

TABLE 3. THE SENSITIVITIES OF ELECTRET ION CHAMBERS

| Device Id | Volume (ml) | Calibration Factor 150 Bq M ⁻³ h ⁻¹ | or Volts per - <u>4 pCi L⁻¹ d⁻¹</u> |
|---------------------------|-------------|---|--|
| Rad Electret (short-term) | 200.0 | 0.330 | 8.0 |
| Rad Electret (long-term) | 200.0 | 0.029 | 0.7 |
| RTCA Radome (long-term) | 65.0 | 0.029 | 0.7 |

CONCLUSIONS:

Radon research and measurement from the early 1900's to the present has come a long way. Some of the best research in natural radiation and specifically of radon was conducted in the early 1900's in European research laboratories by eminent scientific investigators such as the Curies, Dorn, Rutherford, Soddy, Thomson, Elster and Geitel. I consider that time, as the classical era of scientific research with limited resources.

The suggestion in 1924-1932, that radon was the cause of high incidence of lung cancer in European underground miners intensified the research and study of radon throughout the world to be subsequently interrupted by the Second World War. In the meantime, in 1951 across the Atlantic at the university of Rochester NY, researchers pointed out that the lung cancer in underground miners was due to the alpha radiation dose delivered by the radon decay products. The findings of the epidemiological studies in different groups of mines (1950's-1960's) conducted and cited in the BEIR Committee Report VI, solidified the evidence that from 60,000 miners over 2,600 developed lung cancer where only 750 were expected. Since 1998, the epidemiological study conducted in the State of Iowa, US, showed beyond any reasonable doubt that radon and radon decay products cause lung cancer to the general public in residential buildings.

While studies were conducted to assess the health risk from radon and radon decay products, radon research and measurements were conducted to characterize different environments such as radon in soil, building materials, ground water, the atmosphere, caves, spas underground mines and in the residential environment. Radon measurements were used to study air masses vertical diffusion, atmospheric studies, earthquake prediction and in geological studies.

Measurements of radon in the early years were conducted with electrometers, electroscopes and ionization chambers. By 1947, fast pulse ionization chambers begun to replace total ionization chambers that could measure as low as 2 Bq for a 16-hour count. Beginning with the mid 1960's many measurement methods for radon and radon decay product concentrations their inhalation and particle size distribution were made in The US and Canada, Austria, France, Germany, Norway, Sweden, Australia and Japan. In the last twenty-five years, several instruments and methods for measuring radon rather than radon decay products were developed to characterize the indoor radon environment. Today's, most commonly used instruments for radon measurements are passive integrating devices such as activated carbon collectors and electret ionization chambers (for 2-7 day exposure) and alpha track detectors (for 90 days or longer). The measurement application of any of these devices is a function of sensitivity providing the average radon concentration during the measurement period.

In the last twenty years, several passive or active continuous electronic devices have been developed that produce hourly measurements and the average value during the test period. They are represented by scintillation cell, current and pulse ionization chambers and by solid-state detector monitors. Tables 1 and 2 compare the sensitivity and cost of each device making it easier to select the appropriate one for a specific measurement application.

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