RADON: PAST, PRESENT AND FUTURE*

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Received November 15, 2012

Following its discovery by Dorn in 1900 [1] studies of radon and its progeny have contributed to many scientific fields but it is best known for being a significant cause of lung cancer. From the work of Paracelsus [2] and Agricola [3] it is clear that radon was responsible for the fatal lung disease of silver miners in Saxony and Bohemia in the 16th century. In the 20th century the causal role of radon and its short-lived progeny in lung cancers in underground uranium miners was established in the 1950s. From the mid 20th century, stimulated initially by the work of Holmqvist in Sweden [4], there has been an ever-growing interest in the health effects of exposure of the general public to radon in their homes. Over recent decades a large body of radon epidemiological studies both of underground miners and the general population has quantified the radon lung cancer risks to both of these groups. Radon was classified in 1988 by IARC [5] as a human carcinogen mainly on the basis of the miner epidemiology. More recently the World Health Organisation International Radon Project in 2009 identified radon as the second cause of lung cancer after smoking [6]. Many governments now have established radon control strategies to reduce the risk to public health from radon. Apart from the health effects of radon it has also played a role in many scientific areas such as radiotherapy, meteorology and geophysics. Starting with the NASA Apollo lunar missions the detection and behaviour of radon and its progeny on the Moon have made contributions to our understanding of the geology of the Moon. There is currently a growing interest in radon behaviour on Mars which it is hoped will add to our knowledge of its volcanology and the availability of water on that planet.

1. INTRODUCTION

In 1900 following on the work of the Curies in 1899 Ernst Friedr ich Dorn, a German physicist established that radium emitted a radioactive gas which we now call radon [1]. Radon (\(^{222}\text{Rn}\)) was accepted in 1912 as a new element by the International Commission for Atomic Weights. The International Committee for Chemical Elements and the International Union of Pure and Applied Chemistry (IUPAC) in 1923 approved the use of the names radon (Rn), thoron (Tn), and


actinon (An) for the three isotopes of this element with the longest half-lives. Their respective half-lives are 3.823 d (\(^{222}\)Rn), 55.6 s (\(^{220}\)Rn) and 3.96 s (\(^{219}\)Rn). Radon for many years continued in some scientific circles to be called radium emanation or simply “emanation”. As early as 1901 the presence of radioactive substances in the air was established by collecting activity on a wire suspended in outdoor air at a high negative potential [7]. The atmospheric radioactivity so collected was found to be mainly positively charged radon progeny produced by radon in the air. Due mainly to their alpha particle emissions the radon progeny were found to be significant contributors to the production of atmospheric small ions in the air near the ground. Radon and its progeny were thus found to be an important source term in atmospheric electricity in the troposphere. It is of interest to note that the discovery of cosmic radiation by Victor Hess arose as a result of his balloon flights, up to 5.3 km, in the period 1911-12 investigating the fall off with altitude of the ionisation of the lower atmosphere [8]. He found that the ionisation of the air, up to about 1 km height decreased but above that height the ionisation started increasing again. He correctly postulated that this was due to radiation penetrating the atmosphere from the cosmos. His discovery of cosmic radiation was recognised when he was awarded the Nobel Prize in 1936. His balloon investigations which were related to radon in the atmosphere can justifiably be seen as having an important role in the discovery of cosmic radiation. This in turn led to the development of nuclear physics.

1.1. RADON INDOOR

For most people the majority of their lifetime exposure to radon and its progeny takes place in indoor environments such as their homes, schools and places of work. Indoor exposure to radon is much greater than that received outdoors not only because people spend about 80% of their time in indoor spaces, such as homes, schools and places of work, but also because indoor concentrations of radon are generally greater than outdoors. The principal source of radon to which humans may be exposed to in indoor air is \(^{226}\)Ra present in the rocks and soils beneath buildings. While some building materials with high concentrations of \(^{226}\)Ra are known to exist generally the contribution of building materials to indoor radon is much less than that from the radon in soil gas which can enter most buildings. Radon production in rocks and soil and the dynamics of its entry into a building are quite complex. Factors controlling its entry include soil permeability and water content, the integrity of the building foundations and meteorological variables. The main driver of soil gas flow into a building is the positive pressure differential that usually exists between soil gas and indoor air. As a result of many national and regional surveys of indoor radon it has been found that the average indoor radon concentration in most countries is typically less than 100 Bq/m\(^3\) [6]. The distribution of indoor radon concentrations in extensive surveys is usually
found to be log-normal and thus even in a number of countries with mean radon levels of less than 100 Bq/m³ concentrations of some tens of thousands of Bq/m³ have been found in a small number of houses. The behaviour of radon and in particular its airborne short lived progeny in the indoor environment depends on such variables as the air exchange rate, the size distributions of indoor aerosols and the internal surface to volume ratios of rooms. From the second half of the 20th century onwards models were developed to describe the behaviour of indoor radon and its progeny [9]. Airborne short-lived radon progeny will interact with aerosols in indoor air by attachment to them. Airborne radon progeny are present in two modes in indoor air. These are as unattached and attached radon progeny. The unattached mode are predominantly positively charged and are typically found in the 0.5 to 1.5 nm size range. The attached mode will have a size range and state of charge which reflects the characteristics of the aerosol particles to which they have become attached. The deposition characteristics of both these modes of radon progeny in the human respiratory tract is of considerable importance to the radiation doses and to the risk of inducing lung cancer in radiation sensitive lung tissue. While lung dosimetry modelling is quite complex it may be stated that irradiation of sensitive cells in the bronchial epithelium by the alpha particles emitted by the short-lived radon progeny ²¹⁸Po and ²¹⁴Po contribute significantly to the lung dose and the consequent risk of lung cancer [10].

1.2. HEALTH EFFECTS

The high mortality due to respiratory disease of silver miners in Saxony and Bohemia was reported by Paracelsus in the 16th century [2]. Agricola also gave an account of a fatal respiratory disease which was quite common in these miners [3]. We now know from the description given in these early accounts that this disease was lung cancer. Prior to extensive smoking in the general population lung cancer was very rare and its occurrence in these medieval miners was not formally recognised until the late 19th century [11]. While a number of substances in mine air such as ore dust and diesel fumes were also considered as contributing to lung cancer in miners radon first became suspected of being the primary cause of the lung cancer towards the middle of the 20th century. By the 1950s based on cohort epidemiological studies radon or more correctly its short-lived progeny became scientifically established as a primary cause of lung cancer in underground uranium and other hard rock miners. An elevated risk of lung cancer in underground miners exposed to high concentrations of radon and its progeny has been consistently demonstrated in a number of large miner epidemiological studies [12]. The International Agency for Research on Cancer largely on the basis of these miner studies in 1988 classified radon as a Group 1 human carcinogen [5]. A joint analysis of 60000 miners in 11 cohorts, in Europe, North America, Australia and Asia, in whom a total of 2600 lung cancers had occurred showed that the average
Excess Relative Risk (ERR) per WLM was estimated to be 0.44% (95% C.I.: 0.20% to 1.00%). It should be noted that in the cohorts there were differences in regard to quality of exposure data, the presence of other occupational risk factors etc. This limited to some extent the findings of the joint analysis. Here the WLM (Working Level Month) is a unit of exposure to radon progeny. For historical reasons it is generally used as the unit of exposure in miner studies. (1 WLM is defined as the cumulative exposure over 170 hours to a radon progeny concentration in any combination in one litre of air that been ultimately will release 1.3 x 10^5 of potential alpha particle energy.)

Following on the discovery of radon in Swedish dwellings by Hultqvist in the 1950s it became obvious that radon exposure in homes might be a cause of radiological concern for public health and was not just a problem for occupationally exposed underground miners [4]. High radon concentrations had been found during the 1970s in many countries. As a consequence of this a number of national radiological protection agencies began in the 1980s to establish national radon programmes. Since then there has been established an extensive data base of indoor radon mainly in Europe and North America as a result of both national and regional indoor radon surveys that have been carried out [13]. This data has been used in a number of residential radon epidemiological studies aimed at determining the relationship between lung cancer incidence in the general population and their exposure to indoor radon and its progeny. Case-control residential radon epidemiological studies have been carried out in a number of countries in Europe, North America and also in China. Most of such individual studies carried out had a low statistical power due to their small size. The results of studies in Europe, North America and China, where individual data on such important factors as smoking habits and radon concentrations in present and where possible in previous homes of the subjects was obtained, were therefore pooled and re-analysed. The results of these poolings were published in the last decade [14]. All the pooling studies yielded strong evidence that exposure to radon in combination with smoking is responsible for a not-insignificant fraction of lung cancers in the general population. They also provided direct estimates of the size of the risk whereas previously risks from radon exposure of the general population were estimated by extrapolation from the miner studies.

To date the largest of the pooled studies has been the European pooling and its findings were first published in 2005. In this study the data from 13 European case-control studies involving 7148 lung cancer cases and 14208 controls was re-analysed [14]. Detailed information was obtained for these subjects on their smoking histories and on the radon concentrations in the homes they had occupied during the previous 15 years or longer. Excess relative risks due to exposure to the long-term average radon concentration in their homes were calculated. This approach took into account the random year-to year variability in the measured radon in the dwellings of subjects. The analysis yielded an excess relative risk (not to be confused with absolute risk) of 16% (95% C.I.: 5%-31%) per 100 Bq/m³.
increase in the long-term average radon concentration. This excess relative risk was found to show no significant difference by study, sex, age or smoking status. It is interesting to note that the exposure-response relationship was approximately linear even down to below 200 Bq/m³ which is presently a typical action or reference level in many countries. To put the excess relative risk in context the estimated risks at 0, 100 and 400 Bq/m³ relative to lifelong smokers with no radon exposure are 1, 1.2 and 1.6 and 25.8, 29.9 and 42.3 for those continuing to smoke at 15-25 cigarettes per day. One of the most important results of this pooling, illustrated by these relative risk estimates, is the strong synergism between indoor radon exposure and smoking. This has implications for strategies to be used in national radon control programmes. It has been recently recommended, for example, that radon control and anti-smoking campaigns should be linked [15].

The WHO (World Health Organisation) has concluded that the risks from indoor radon estimated in these pooled studies and those of underground miners are in reasonable agreement. It is currently estimated by the WHO that globally the proportion of lung cancers which are attributable to radon exposure ranges from 3% to 14% [6]. The pooling studies outlined here can be considered a sound basis on which to base the development of national radon risk management policies.

In recent years a number of epidemiological studies have taken place to investigate the relation between radon exposure and a number of diseases other than lung cancer such as leukaemia and multiple sclerosis. For example in the period 1997-2001 thirty three studies took place to investigate a possible relationship between radon exposure and leukaemia in adults and children [12]. Eight of these studies were residential case-control studies, six were miner cohort studies and nineteen were ecological studies. Only in the case of the ecological studies was a positive correlation between leukaemia and radon exposure found. Recent residential studies have, however, reported a positive association between childhood leukaemia and radon [16].

2. SOME RADON APPLICATIONS

While radon is generally and correctly perceived as a major public health issue there has been since the early part of the 20th century a number of practical and beneficial applications of radon in such as areas as atmospheric physics, geophysics and radiotherapy. A small selection of these applications is now given here:

2.1. RADIOThERAPY

It was first suggested by Pierre Curie as early as 1901 that cancer the implantation of radioisotopes into malignant tumours could be used as a form of cancer therapy. Today this is a common form of radiotherapy called brachytherapy. From about 1915 onwards radon grown from radium solution and sealed in small
glass tubes was used for brachytherapy on thousands of patients in the U.S.A. This form of brachytherapy was improved and made more effective in the 1920s when Gino Failla at Memorial Hospital in New York began to encapsulate radon in gold tubes instead of glass [17]. The gold stopped not only radon progeny alpha particles but also the beta particles which caused necrosis of surrounding healthy tissue. The gamma radiation emitted by the radon series could on the other hand easily penetrate the gold to kill tumour tissue. The gold radon seeds typically contained between 2 and 200 MBq of radon and were about 5 mm in length and 1 mm in diameter. While radon seeds continued to be used from the 1920s to at least the 1960s their use in brachytherapy, in particular for prostate cancer, has now been supplanted by such radioisotopes as $^{215}$I and $^{103}$Pd.

2.2. ATMOSPHERIC PHYSICS

Measurements of the behaviour of radon and its progeny in the atmosphere have been used for many years to characterize the properties of the atmospheric boundary layer. Radon has been found to be well suited as a tracer for vertical mixing studies in the boundary layer mixing [18]. This is both because its 3.8 day half-life is long in comparison to turbulent timescales in the atmosphere and short enough to effectively restrict its concentration to the free troposphere. Radon has in recent years also been used as a transport tracer of forest canopy–atmosphere CO$_2$ exchange in tropical rain forests [19]. It has been demonstrated a radon based tracer technique is more reliable and accurate than those based on CO$_2$ at quantifying gas exchange rates between the atmosphere and forest canopies. The net exchange of trace gases and CO$_2$ between tropical forests and the atmosphere may be a process of importance to the global carbon cycle and a significant factor in the radiative budget of the earth. It is expected that the contribution of radon based tracer studies to our understanding of these processes will increase in the future.

2.3. LUNAR AND MARTIAN RADON

Since the mid-20th century radon applications in geophysics such as earthquake prediction and mineral exploration became well established but since the 1970s there has been a growing interest in the behaviour of extra-terrestrial radon mainly on the Moon but more recently on Mars.

In the essentially vacuum conditions on the lunar surface radon migrates in a manner considerably different from that on the earth. At a typical daytime lunar surface temperature of 300 °K radon atoms will be emitted from the lunar regolith at thermal velocities of about 0.15 km/sec. This is well below the 2.4 km/sec escape velocity from the moon. Under the influence of lunar gravity the emitted radon atoms undergo a series of ballistic trajectories or “bounces” over the lunar surface until they decay [20]. During the bouncing process the radon atoms may
reach a maximum altitude of about 10 km. Depending on the lunar surface temperature and time of accommodation between bounces it is estimated that radon atoms may be displaced from their point of origin by between 700 and 800 km. It has even been predicted that there is a pile up of radon at the lunar sunrise terminator of the moon. On the other hand when a radon atom on or above the lunar surface decays its immediate decay product $^{218}$Po has a recoil energy of about 100 KeV corresponding to a velocity in vacuo of about 300 km/s which is well above the lunar escape velocity. Depending on the direction of recoil this implies that a significant fraction of lunar radon progeny formed from the decay of radon on or above the lunar surface may escape from the moon. Quantitative evidence of the existence of radon and its progeny on the moon was obtained in the early 1970s from the Apollo series of lunar missions. For example a significant increase in radon activity in the region of the crater Aristarchus was detected by the alpha spectrometer on the Apollo 15 command module suggesting possible internal activity at the site which could indicate the emission of other gases [21]. Large variations and localised increases in the radon progeny $^{210}$Po over many parts of the lunar surface were detected by the Apollo 16 alpha spectrometer. In addition the observed $^{210}$Po surface activity was found to be significantly greater than that of radon indicating that large temporal variations of radon emanation is occurring on the moon. This also indicated internal activity in the moon [22]. Alpha spectrometric measurements by the Lunar Prospector during 1998–99 confirmed these earlier findings and detected radon outgassing events in the vicinity of the Kepler and Aristarchus craters [23]. Such lunar radon studies have helped so far to contribute to our knowledge of aspects of lunar “geophysics”.

Two Exploration Rover vehicles successfully landed on Mars in 2003. They were equipped with alpha particle detectors. A statistical analysis of the high-energy end of the alpha spectrum obtained by one the vehicles in 2006 yielded evidence of $^{210}$Po on Martian dust at the Meridiani Planum location [24]. It has been inferred from this analysis that the radon exhalation rate on the surface of Mars may be significantly greater than on the moon. This might be due to the presence of water in Martian surface soil as radon exhalation from fluid filled pores should be greater than from vacuum filled pores. In 2016 the Martian Trace Gas Orbiter mission is scheduled to commence. This mission will be devoted to detect and characterise the trace gases in the Martian Atmosphere and is expected to yield more information on Martian radon.

3. CONCLUSIONS

Radon studies have a long and interesting history. Its role as the second cause of lung cancer means that indoor exposure to radon is a significant public health issue. An important aspect of indoor radon exposure, both in residential and
occupational settings, is that it is easily measured and controllable. Using modern building technology it is possible to construct buildings in which the indoor radon concentration will be at an acceptable level. It should be noted that it is also technically possible at relatively low cost to reduce high indoor radon levels in existing buildings to acceptable level. At present many EU Member States have a voluntary Reference Level of 200 Bq/m$^3$ for existing and future houses and in a few of them there is, in addition, a 100 Bq/m$^3$ Target Level for new houses. What is required at a national level to reduce the public health burden from radon is the establishment and implementation of a coherent and cost-effective radon control strategy. Guidance and advice on how to achieve this is now readily available [6, 15].

REFERENCES


