Radon and Its Decay Products in Caves

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Abstract:
Most radioactive substances are solids that possess physical properties and decay half-lives that prevent them from being considered a natural radiation hazard. Radon, however, is a gas with a half-life just large enough for it to be able to diffuse out of the ground in which it is formed and migrate in open air from its origination point with changes in air flow. Whilst being solids, the decay products of radon are highly reactive and possess half-lives in the order of minutes. Consequently, the decay of these compounds, with the associated production of high energy α-particles, presents a possible health risk. Investigations into radon and its progeny in Australian caves are showing the presence within the caves of high levels of these substances. Described here are the factors that have been shown in studies worldwide to affect these levels over time within cave systems.

Introduction

Radon and its short-lived decay products are a major issue in human exposure to natural sources of radiation, especially with regard to enclosed spaces such as mines, buildings and caves. This has certainly become the case over the last few decades as research has linked elevated levels of radon to the increased numbers of fatal cancers experienced by miners in some mines (Lorenz 1944; Jackson *et al.* 1987; Prime and O'Hara 1991); the levels of radon in some abandoned mines have been measured as high as 240 million Bq m^{-3}. Unlike mines, the study of radon levels in caves has been limited. Nevertheless, it has been found that in many of the caves commonly used by the general public, such as the Carlsbad Caverns (Wilkening and Watkins 1976) in New Mexico, USA, and in wild caves, such as the Giants Hole (Bown 1992) in Derbyshire, UK, there are high levels of radon and radon decay products, the highest at present being 160,000 Bq m^{-3} in the latter wild cave.

Investigation into the levels of radon in Australian caves was initiated almost by accident in 1991 (James 1996 pers. comm.). Test studies were required on the use and suitability of track etch detectors in measuring radon levels in buildings, and these tests were performed in the caves below the Nullarbor Plains. The results of these tests, and of subsequent studies at other Australian cave systems, showed the existence of high levels of radon in these systems (Lyons 1992, Solomon *et al.* 1992).

The reasons for studying radon and its decay products in caves can be summarised as:

(i) determining whether there are health concerns with regard to the concentrations of radon and radon decay products for tourists and, more significantly, guides and maintenance workers;
(ii) determining possible sources for any noticeably raised levels of radon;
(iii) determining the distribution and flow of radon within the cave; and
(iv) suggesting methods of reducing exposure to any levels of radon or radon decay products deemed to be dangerous to health in ways that do not harm the sensitive cave environments.

Background

The only naturally occurring heavy radioactive isotopes are uranium-235 (^{235}U), uranium-238 (^{238}U) and thorium (^{232}Th). These primordial constituents of all rocks and soils possess extremely long half-lives, the time elapsed since their creation being insufficient to facilitate the decay of these isotopes into other more stable compounds. The decay paths of these three elements is long and complex, proceeding through various shorter lived radioactive atoms until the formation of the stable non-radioactive lead isotopes (Figure 1). Because of their chemical reactivity, these intermediates tend to stay within the material in which they were formed. However, in each decay process, an isotope of the noble gas, radon, is formed. Essentially inert in cave environments, radon can diffuse slowly through solid matter into the air (Hopke 1987), where it becomes a significant contributor to the background levels of natural radiation. Indeed, over 50 % of the background radiation is from radon decay.

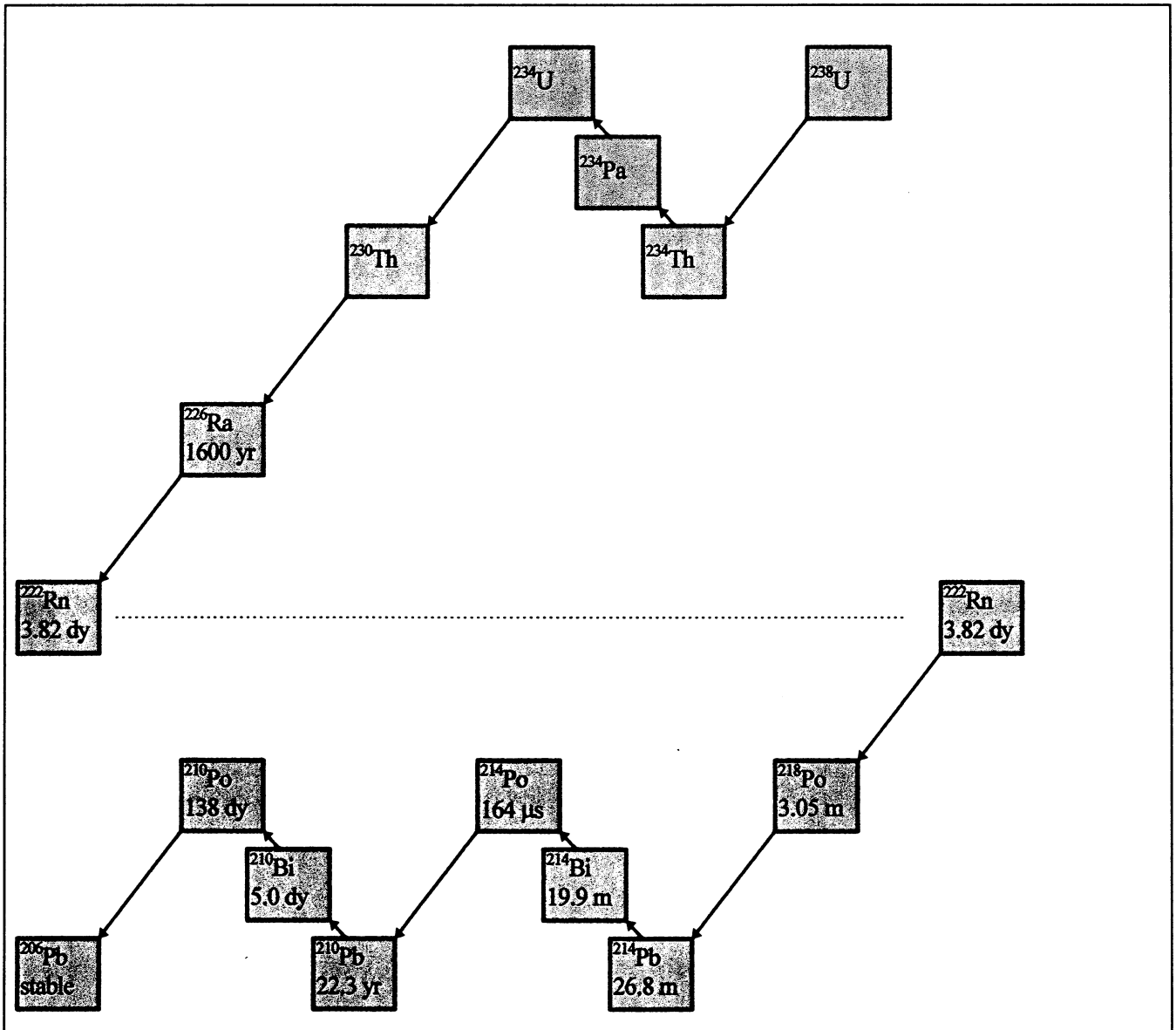

Figure 1. Decay chain and half-lives for the radon isotope, ^{222}Rn, and its decay products.
long downward arrow = a-decay short upward arrow = b-decay

Radon

The normal background level of radon in outdoor air is about 7 Bq m^{-3} ‡. Radon levels for enclosed spaces such as caves will generally be significantly higher than outdoor levels, because the dispersion of the gas by airflow changes is more restricted and thus less effective. Furthermore, the average air pressure differential experienced by caves often results in a pooling of gases within these spaces. Common radon levels found in caves range from 200 Bq m^{-3} to several thousand Bq m^{-3} averaged over a year For instance, levels at Wombeyan caves average 240 Bq m^{-3} annually whilst those at Jenolan (northside) caves are around 1500 Bq m^{-3} (Figure 2).

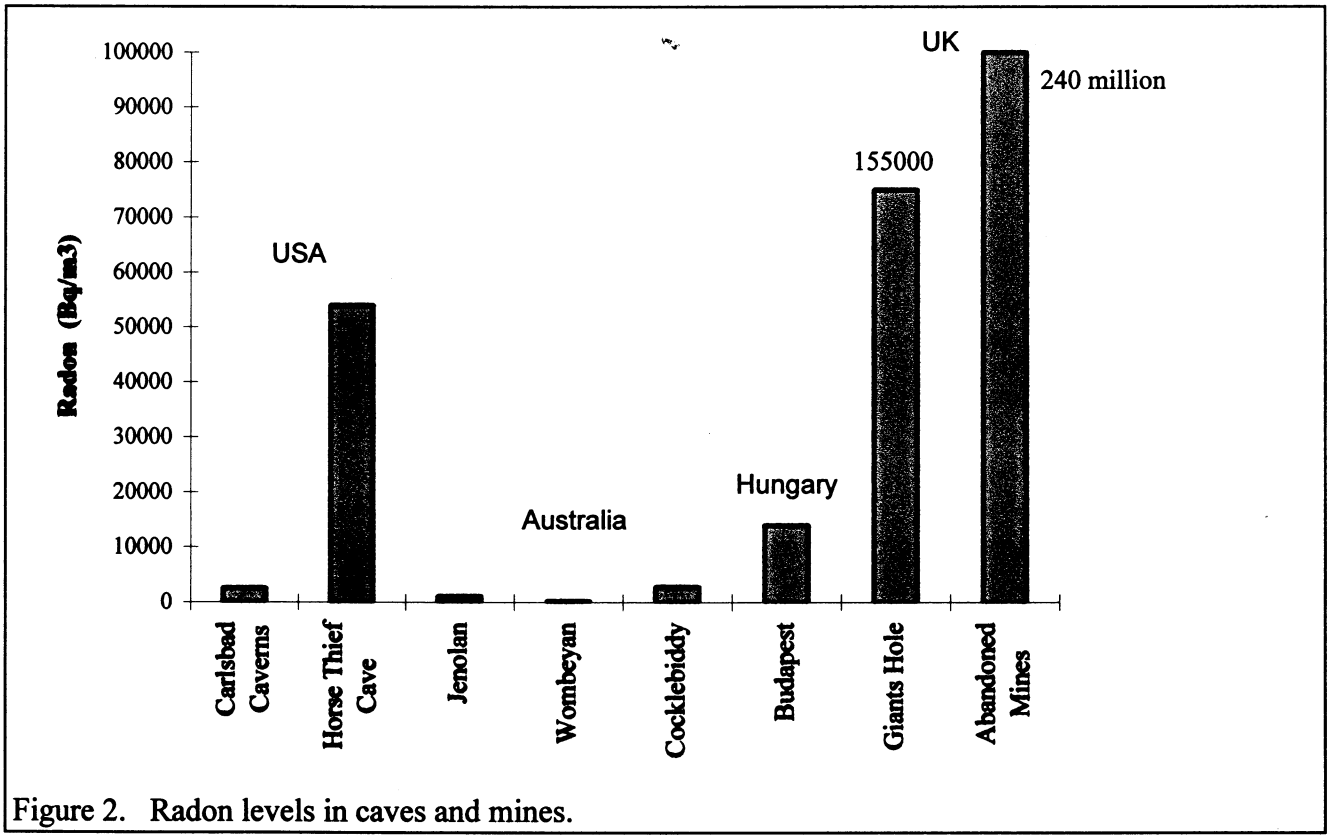

Figure 2. Radon levels in caves and mines.

The study of radon levels in caves began in the 1970's and, since 1974, radon levels have been monitored and analysed in both wild caves and show caves around the world. For example, radon levels have been measured in caves in Derbyshire, UK (Gunn *et al.* 1991a,b), New Mexico, USA (Wilkening and Watkins 1976), Australia (Solomon *et al.* 1996) and Budapest, Hungary (Géczy 1993). These measurements have shown radon to vary both seasonally and diurnally, with maximum levels observed in summer and during daylight hours, and minimum levels in winter and at night for most cave systems.

‡ The becquerel (Bq) is the SI unit for the measurement of radioactive decay, and is equal to one decay per second.

Radon Decay Products

The decay products (progeny) of radon are polonium-218 and -214 (^{218}Po, ^{214}Po), lead-214 (^{214}Pb) and bismuth-214 (^{214}Bi). Lead-210 (^{210}Pb), with a half-life of 22 years, is considered to be stable and, therefore, the end point for the progeny decay sub-chain (Figure 1). The chemistry of the radon progeny is of some importance. While both lead and bismuth are relatively unreactive, polonium isotopes are both reactive and extremely toxic. Consequently, high levels of these could be harmful in and of themselves, without the added danger of their being radioactive. In practice, the concentration of polonium present at any one time within a cave is extremely small, and so the radiation hazard predominates.

All of the radon decay products are solids, and so can "plate out" of the air onto other surfaces, such as those provided by the walls of caves, dust particles, moisture droplets and condensation nuclei. Condensation nuclei are small particles formed by photochemical or combustion processes in the outside atmosphere that have been transported into the cave atmosphere through air exchange. Because of the small size and high relative abundance of, and thus the large surface area provided by, condensation nuclei, radon decay products are more likely to attach to these particles rather than dust or water droplets. Therefore, the radon decay products are a mixture of "attached" and "unattached" particles.

As condensation nuclei are airborne, and dust and moisture can be, both the attached and unattached radon progeny have the capability of being inhaled into the lungs where, depending on the size of the particles, they become trapped anywhere along the respiratory tract from the nose to the lung tissue itself. Due to their more extreme reactivity, the unattached fraction pose a greater danger to health than the attached decay products. Moreover, the unattached fraction deposit in the respiratory tract with 100 % efficiency (Robkin 1987). Just like the decay of radon, the decay of the two Po isotopes produces a-particles which, when the nuclides are trapped in the lungs, can damage the sensitive lung tissues.

For a chain of radioactive isotopes where the parent is relatively long-lived, the amount of each chain member will adjust until the sources and losses of each are in balance. When the only source is an original amount of the parent and the only losses are decay processes, the equilibrium reached is one where the activities of the parent and its decay products are equal. That is, they are said to be in secular equilibrium. For the radon decay chain, in the absence of external influences, it takes approximately 4 hours for this state to be achieved (Robkin 1987). Processes, such as plate-out onto walls or those that cause changes in airflow, which operate within this time frame and which alter the abundance of the particles by introducing or removing them, can lead to departure from this "ideal" state.

The earliest research of radon decay products in caves was undertaken during the late 1970's in the Carlsbad Caverns, New Mexico, USA (Ahlstrand 1980). These studies showed that there existed temporal variations in the levels of radon decay products and that these levels were probably related to seasonal, diurnal and pressure related phenomena. A subsequent study of the decay product levels within Giants Hole, Derbyshire, UK (Middleton *et al.* 1991), found that changes in the radon decay product levels were primarily the result of changes in the direction of airflow within the cave and that this in turn was related on a seasonal and diurnal scale to temporal variations in the temperature gradient between the cave and external air.

Factors Affecting the Levels of Radon and Radon Decay Products Within Caves

Radon and radon decay product levels in cave atmospheres are dependent upon the whereabouts of the parent (radium) source, the diffusion rate of radon through and emanation from the ground, and the rate of introduction and removal of radon in air and water (Prime and O'Hara 1991; Lyons 1992; Nazaroff 1992; Blaauboer and Smetsers 1996) (Figure 3). The rate of diffusion varies with depth and geology. As the surface is approached, the diffusion rate will increase, as release of radon into larger air cavities like caves becomes important. The geology and physical processes such as weathering help determine the 'porosity' of the earth, and thus to some extent the moisture content and permeability of the soils. Consequently, levels of radon in caves is controlled by the radium concentration in the cave structure, the physical structure of the cave itself which affects the diffusion rate, and by atmospheric pressure changes.

Water can also be an important source of radon in caves, depending on the rock over (or through) which it has flowed (Prime *et al.* 1991) before entering the cave. Water flowing over or through rock of high uranium or radium concentration often has high radon levels. Despite the low solubility of radon in water, the high concentrations of radon found in most soils readily forces some radon into groundwater and circulating subsurface waters. Water flow is thus a significant transport path for radon dispersion, allowing radon to migrate along caverns and fractures as far as the transporting waters allow (Nazaroff 1992; Hakl *et al.* 1993a), leading to possible increases in radon levels at sites removed from the point of origin of the radon.

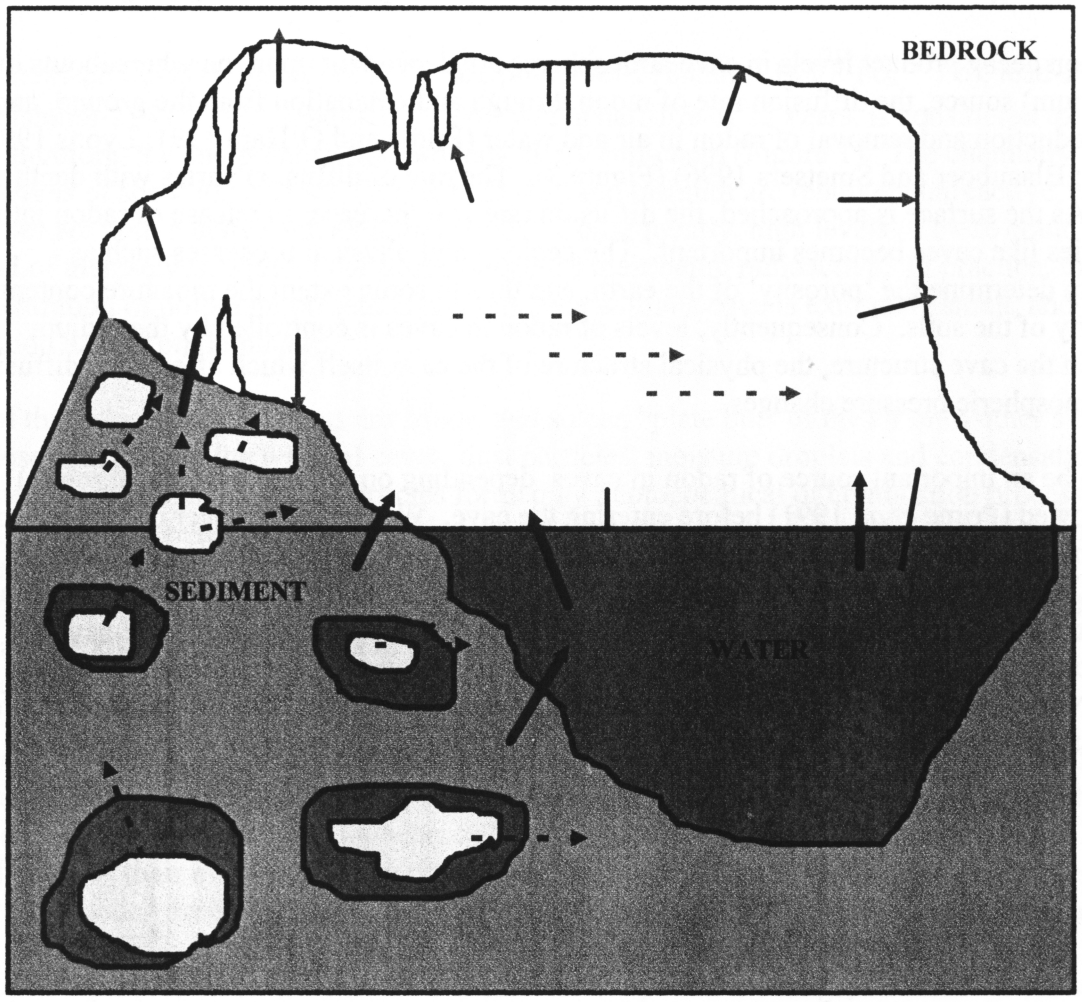

Figure 3. Factors affecting radon concentrations in air.

Exchange rates between cave and external air can be complicated, and are dependent upon temperature differences between the cave and outside and the cave configuration (entrance above or below the limestone voids, mazes, complex passages, chimneys etc.). For example, outside caves, a temperature inversion can occur some metres above the ground at night, resulting in an increased radon concentration at ground level. During the day, the air heats more uniformly and so mixes more thoroughly. This effect is enhanced seasonally, as the summer heat is more intense leading to warmer soils, and the winter nights are less stable leading to greater mixing and less likelihood of an inversion. Wind direction can also be important to cave airflow as can be the presence of active streamlets. Indeed, any factor likely to change airflow patterns within a cave is also likely to alter the levels of radon and its decay products.

Middleton *et al.* (1991) and Prime and O'Hara (1991) confirmed that the direction of airflow is one of the dominant factors affecting both the spatial and temporal distribution of radon decay products within caves. On both a seasonal and diurnal scale, airflow direction is determined by the temperature gradient that exists between the cave and external air; some of the temporal variations have been related to changes in atmospheric pressure. On the other hand, good correlations have been found between the variation in radon levels in the caves and outside temperatures (Géczy 1993) and changes in air pressure within the caves themselves.

Exposure to Radon-222 and Its Decay Products

Knowledge of the health effects of radon and its decay products is derived mainly from studies of cancers in underground miners, all of which have unambiguously shown that prolonged exposure to high concentrations of these substances leads to a high number of fatal lung cancers (Behounek 1970; Eisenbud 1987; Jackson *et al.* 1987). However, extrapolation of these results to situations where humans are exposed to much lower radon levels, for example, in caves, has severe limitations despite being of prime interest with respect to general public health.

Radon decays to ^{218}Po, emitting an a-particle in the process. Polonium-218 is a chemically active solid material that can bind to lung tissues, which has a half-life of about 3 minutes. It too decays by also emitting an a-particle, to ^{214}Pb. The properties of a-particles are such that they are only a health hazard if the emitter is in contact with living tissue. The relatively large, positively charged a-particles rapidly lose their energy to surrounding atoms. This means that they do not penetrate the skin and are stopped by a few centimetres of air. However, if the a-emitter is in the body (*e.g.* on the lung tissue), all of the energy is released to a much more sensitive tissue.

Being a gas, ^{222}Rn can be inhaled, although it is generally exhaled again. Radon is also somewhat soluble in blood and body fat (Eisenbud 1987). However, should radon decay within the body, it will not only emit an a-particle but deposit radon progeny as well, whereupon a further two a-particles will also be emitted within the body. Radon decay products produced outside the lungs can also be inhaled. Indeed, the amount of radon progeny deposited as a result of radon decay in the lungs is negligible compared to the amounts of radon decay products that are inhaled. These solids can be attached to condensation nuclei and other airborne particles or unattached, thus altering the overall size of the particle. Only a small fraction of those radon decay products breathed in will be exhaled again, with the particles possibly becoming trapped some distance along the respiratory tract in correlation to the size of the particle. The unattached radon decay products, which are smaller than the attached decay products, are more dangerous because they can travel further into the lungs before either being exhaled again or becoming trapped.

The chances of the energy released by a-particles breaking chemical bonds within organic molecules or causing ionisation or forming reactive free radicals are relatively high. All of these changes have the potential to disrupt the normal metabolism and produce changes which may either be easily reversed or affect cell replication, cause cell death, or in a limited number of cases, induce cancerous or abnormal growth. As those a-particles released by the radon decay products are more energetic than those emitted during radon decay, the progeny are once more revealed as being of greater concern to public health.

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