The effect of domestic air treatment equipment on the concentration of radon-222 daughters in a sealed room

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ABSTRACT

REDUCTION OF the ventilation rate in houses in the interests of energy conservation can lead to a build-up of radon-222 in room air. This results in irradiation of the lungs by the radioactive daughters of radon. An electrostatic precipitator has been used to remove radon daughters from the atmosphere of a sealed room. It is shown that this can reduce the dose to the bronchial epithelium by an order of magnitude, and effectively counter the effects even of extremely low ventilation rates.

INTRODUCTION

URANIUM IS widely distributed in the earth's crust at typical concentrations of 2-4 ppm[1] and hence occurs in most commonly used building materials such as clay bricks. Uranium-238 is the parent of a chain of radioactive nuclides whose fifth member is radium-226 and hence this nuclide is also present in many building materials, usually in concentrations of a few parts in 10^{12} [2]. Radium-226 decays to produce the noble gas radon-222 which is itself radioactive with a half-life of 3·8 days. Radon-222 can diffuse from building materials and the subsoil beneath buildings into the air within the building. Radon-222 decays to form particulate daughter products which are isotopes of polonium, bismuth and lead. The first four daughter products of radon-222 are polonium-218 (α emitter of half life 3·05 min), lead-214 (β emitter of half life 26·8 min), bismuth-214 (β emitter of half life 19·7 min) and polonium-214 (α emitter of half life 164 μs). In view of its short half life polonium-214 is always in equilibrium with bismuth-214 and for practical purposes bismuth-214 may be regarded as a combined α, β emitter. Polonium-218, lead-214, bismuth-214 and polonium-214 are generally referred to as the short-lived daughters of radon-222 and they are inhaled, either as free atoms or attached to dust particles, and become deposited in the lungs. As their half-lives are short, essentially all of these deposited nuclides decay within the lung and give rise to a radiation dose to the cells of the bronchial epithelium. The largest contributors to this radiation dose are the alpha particles emitted by the two polonium isotopes.

Studies among uranium miners in Czechoslovakia[3, 4, 5] and in the United States[6, 7, 8] and in fluorspar miners in Newfoundland[9] demonstrated a correlation between an excess incidence of lung cancer and exposure to high concentrations of the short-lived daughters of radon-222. Although a statistically significant excess incidence of lung cancer can be demonstrated only for cumulative exposures to radon daughters an order of magnitude larger than the life-time exposure of a person occupying a normal building, the data are not inconsistent with a linear exposure-effect relationship with no threshold[10]. If there is a causal relationship between lung cancer and exposure to radon daughters, then a fraction of lung cancers in the general population are the result of exposure to the short-lived daughters of radon in the environment and particularly within buildings.

In general, radiation doses are given in units of the gray (the unit of absorbed dose 1 Gy = 1 joule per kilogram) or to account for the varying biological effectiveness of differing forms of radiation the unit of dose equivalent, the sievert (Sv) is used. However in the case of the short lived daughters of radon-222 special units of concentration and exposure are used. The unit of concentration is the Working Level (WL) defined as any combination of the nuclides polonium-218, lead-214, bismuth-214 and polonium-214 in one cubic metre of air which will result in the ultimate release of $1{\cdot}3 \times 10^8$ MeV of alpha particle energy. The unit of exposure is the Working Level Month (WLM) which corresponds to an exposure to air containing 1 WL for one working month of 170 hours[11]. In the studies of underground miners a statistically significant excess of lung cancers was exhibited by groups of miners whose total exposure exceeded 100 WLM. From a survey of radon daughter concentrations in dwellings in Great Britain[12] it was concluded that the average population exposure rate for the British population is 0·15 WLM/y. Thus over a 70-year life span the total average exposure is 10·5 WLM or about 10 per cent of that exposure above which a correlation with an excess of lung cancers can be demonstrated with reasonable certainty.

The concentration of radon daughters within a building is dependent upon the rate of ingress of radon into the room air from the sub-soil and the building materials, and on ventilation rate, typically one room change of air per hour. With ever-increasing energy costs efforts are being made to design houses with very low ventilation rates (less than 0·2 room changes per hour). Reducing ventilation rates leads to increasing concentrations of radon daughters and hence higher exposure rates. Thus it is desirable to consider methods of removing radon daughters from room air without increasing ventilation rates. To this end investigations were carried out to determine the effect of various air treatment devices on the radon daughter concentrations within a room. The work was carried out under a contract from the Electricity Research Council and the test room used was an environmental chamber at the Electricity Council Research Centre at Capenhurst, Cheshire. Tests were carried out using an electrostatic precipitator, a humidifier and a dehumidifier.

MEASUREMENT TECHNIQUE

MEASUREMENTS WERE carried out in an environmental chamber measuring 3·60 × 3·60 × 2·55 m (33 m^3 volume). Into this volume radon-222 gas was introduced to produce a concentration of 400-800 Bq m^{-3} (11-22 pCi l^{-1}) of radon-222. In a hermetically sealed enclosure the relative concentrations of radon-222, polonium-218, lead-214 and bismuth-214 2½ hours after the release of pure radon-222 gas should be 1, 0·98, 0·96 and 0·92 respectively[13].

Although the environmental chamber did not have zero ventilation rate the measured rate of 0·05 air changes h^{-1} was very low and hence the atmosphere was presumed to be essentially stable 2½ hours after the introduction of radon-222. Ventilation rate was measured using the tracer gas technique[14]. In this case krypton-85 was used as the tracer gas and its decline in concentration measured using the scaled counts from a Geiger-Muller tube. For a constant ventilation rate the concentration of tracer gas decreases exponentially with time.

The above quoted relative concentrations of radon-222 and its short lived daughters at 2½ hours after the introduction of pure radon-222 gas assumes that there is no removal of daughter products. However, when the daughter products are first formed they exist as free charged ions with a high diffusion coefficient. In the absence of condensation nuclei a large proportion of these free ions will attach (plate-out) to room surfaces and hence be removed from the room air producing lower relative concentrations than those predicted from considerations of simple radioactive decay. Thus an important parameter to consider when assessing the expected equilibrium conditions is the condensation nucleus concentration in the air. The condensation nucleus concentration was monitored throughout the measurement period with an Environment One Corporation Rich 100 condensation nucleus counter. In this instrument, air samples are drawn into a small automatic cloud chamber. On expansion of the air in the chamber dust particles act as condensation nuclei and their concentration is found by measuring the attenuation of a beam of light passed through the chamber. A sample is taken and the chamber air expanded about once every second.

Radon daughter concentrations were measured using an instrument based on the radon daughter monitor (RDM) developed by James and Strong[15]. In this instrument air is drawn through a filter paper which is viewed by a silicon surface barrier alpha particle detector. Three integrated gross alpha counts are recorded, one as sampling takes place and two over periods following the end of sampling. From the three recorded gross alpha counts equations can be solved to yield the concentration of polonium-218, lead-214, and bismuth-214 and also the number of Working Levels. The method is described by Cliff[16, 17].

In order to ensure uniform conditions within the test chamber fans were operated throughout the measurement period to keep the air well mixed. Rapid mixing of air containing radon daughters leads to increased plate-out to surfaces as the air-surface contact time is increased. This was shown to be an effective method of reducing radon daughter concentrations in mines[18]. Also recent work has shown that radon daughter products will plate-out on the blades of operating air fans[19]. Thus although operating the fans will have reduced the radon daughter concentration the effects of the air treatment equipment can still be assessed by measuring the radon daughter concentrations before and during the operation of these additional items of equipment.

To measure radon gas concentrations the method of Roberts and Davies[20] was used. In this method a previously evacuated 3-litre chamber is opened in the atmosphere of interest with a filter over the inlet port to prevent the entry of particulate materials. The polonium-218 ions formed as the radon-222 decays are positively charged and are collected on an electrode maintained at a high negative potential (−2 kV). This electrode is coated with a scintillator and the alpha decays of deposited polonium-218 and polonium-214 are recorded using a photomultiplier tube which views the collecting electrode. Counting of alpha particle induced scintillations in the collecting electrode is started at least 3½ hours after the application of the negative potential to the collecting electrode. This time period ensures that when counting starts the daughter products on the collecting electrode are in equilibrium with the parent radon in the chamber.

AIR TREATMENT DEVICES

ALL DEVICES used were commercially produced equipment. The electrostatic precipitator consists of two stages, an ionizing stage followed by a collector and there is also an activated carbon filter to remove odours. The maximum throughput is nominally 5 cubic metres per minute and the precipitator was always used on this setting. The effects of the precipitator and the carbon filter were tested separately and together.

The humidifier that was used is a device which emits a jet of steam and the dehumidifier consists of a heat pump and fan. In the dehumidifier air is blown over cold pipes on which the water vapour condenses and then over warm pipes.

It was predicted that the electrostatic precipitator would reduce the radon daughter concentrations by direct removal of the daughters (see Appendix), both attached and unattached, and also by reducing the condensation nucleus concentration which increases the plate-out of daughters to room surfaces. The dehumidifier might also remove radon daughters directly if significant amounts of dust were to stick to the wet cooling coil. Apart from that, the humidifier and dehumidifier were expected to affect the radon daughter concentrations only indirectly by changing the condensation nucleus concentration in the air.

MEASUREMENTS

FOLLOWING THE introduction of daughter-free. radon-222 into the air a period of at least 2½ hours was allowed to elapse before the air treatment devices were switched on. Measurements of radon daughter concentrations were carried out during this 2½ hour period and for 1½-2 hours during the operation of an air conditioning device.

Initially several runs were carried out without the air conditioning equipment being operated in order to establish conditions in an unmodified atmosphere, see figure 1. It was found that the condensation nucleus concentration was reduced by a factor of 4 over a period of 3½ hours, probably due to agglomeration and sedimentation. This was thought to be untypical of conditions in dwellings where greater ventilation with outside air, various sources of household dust, and, in many cases, smoking will increase the nucleus concentration. Therefore during tests with the air conditioning equipment increased condensation nucleus concentrations were produced periodically by allowing a cigarette to smoulder or by burning a night-light candle.

RESULTS

WITHOUT AIR treatment equipment in use the radon daughter concentrations stabilised at a level where the energy concentration measured in Working Levels was 45-65 per cent of that expected if radon and its daughters were in equilibrium. These lower than expected WL values were probably the result of some loss due to the measurable ventilation rate and by plate-out to room surfaces and the blades of the air mixing fans as mentioned earlier. The use of the humidifier, dehumidifier or carbon filter alone resulted in no significant alteration to the radon daughter concentrations in the room. The electrostatic precipitator, however, reduced the concentrations considerably and was equally effective when operated with or without the carbon filter.

As shown in the appendix, the precipitator would be expected to reduce the WL by a factor of 8 if the daughters were initially in equilibrium or by a factor of 4 if the WL was initially at 50 per cent of the equilibrium value as was the case during these experiments. In fact the measurements showed that the precipitator reduced the WL by factors of between 6 and 19 on different runs. The reduction was greatest if there was no generation of additional condensation nuclei by smouldering a cigarette or candle. If the WL had been reduced by the precipitator, it was found to double if a cigarette was lit even if the precipitator was still operating, as is shown in figure 2.

DISCUSSION

THE LARGE reductions in radon daughter concentrations resulting when the precipitator was in use were caused by two effects. The electrostatic precipitator was very effective in reducing the aerosol concentration and hence directly reduced the radon daughter concentration by removing the daughters attached to these aerosols as well as being effective in removing unattached radon daughters. The second effect resulted from the reduction in condensation nucleus concentration which more readily plated out to room surfaces and the blades of the air-mixing fans[19].

The influence of ventilation on this situation is complex. If outside air mixes with room air it will have a lower radon concentration. Taking a ventilation rate of 0·1 air change per hour and the precipitator working in the largest recommended room (57 m^3), the air will be processed by the precipitator about 50 times more often than it is changed by ventilation. Assuming complete mixing, the condensation nucleus (cn) concentration would be expected to be reduced by a factor of 50 from the concentration outside. George[21] measured condensation nucleus concentrations in outside air and found that they varied from about 7,500 cn/ml in a rural area to 190,000 cn/ml in a city street. If the outside air that entered the room came from either of these extremes and the dust levels were reduced by a factor of 50, the concentration would be 150 or 3,800 cn/ml. Duggan and Howell[22] found that the fraction of unattached polonium-218 increases dramatically as the condensation nucleus concentration is reduced below 10,000 cn/ml and for concentrations less than 4,000 cn/ml the unattached fraction exceeds 50 per cent. Thus in all cases the operation of the precipitator would lead to increased plate-out of radon daughters as well as removing the daughters directly, thereby reducing the airborne radioactivity remaining to be breathed into the lungs.

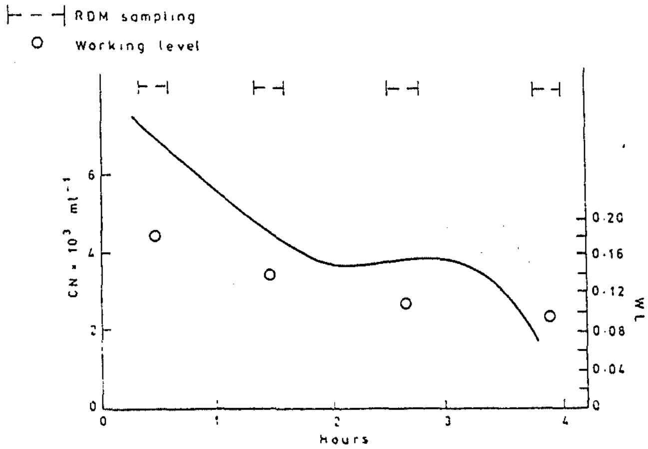

Figure 1

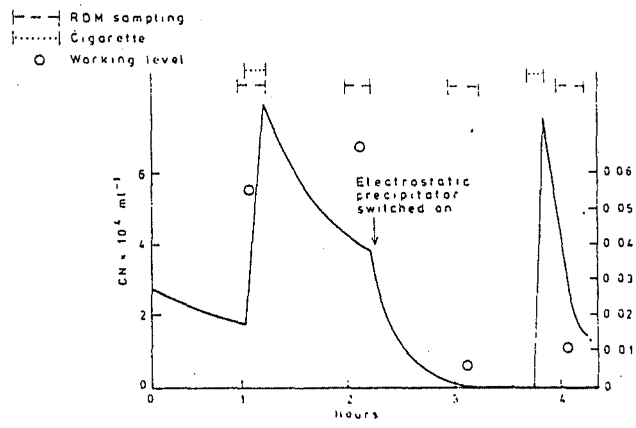

Figure 2

Cliff (1978a) calculated the effects of different ventilation rates on WL, on the assumption that the only removal mechanisms for radon daughters were radioactive decay and ventilation. This showed that reducing ventilation rates from 1 room change per hour (a typical current figure) to 0·2 or 0·1 room changes per hour would increase the WL by factors of 6·5 and 13·5 respectively. The results of the measurements carried out in the environmental chamber show that operation of an electrostatic precipitator will reduce radon daughter working level by a factor of at least 4 and possibly as high as 20, in a room with a low ventilation rate. The higher factor of 20 is probably unrealistic in normal domestic premises as this reduction was due in large part to increased plate-out resulting from the operation of air-mixing fans. The reduction in WL actually achieved will depend on whether the room occupants smoke and the condensation nucleus concentration in the air entering the room.

CONCLUSIONS

THREE AIR treatment devices, an electrostatic precipitator, a humidifier and a dehumidifier, were tested for their effect on the radon daughter concentration in a closed room. The precipitator was the only device of those tested that produced a significant reduction in radon daughter concentrations. Operation of the electrostatic precipitator resulted in reduction of the radon daughter Working Levels by factors of from 6 to 19 depending upon conditions in the room. The use of an electrostatic precipitator would therefore counteract the increase in radon daughter concentrations resulting from reduced ventilation rates.

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APPENDIX

IT CAN be shown that the number of atoms, Q_A, of polonium-218 in a closed system containing Q_{Rn} atoms of radon-222 is given by

$$Q_A = \frac{\lambda_{Rn} Q_{Rn}}{\lambda}(1 - e^{-\lambda t})$$

where λ_{Rn} = decay constant of radon-222
$\lambda = \lambda_A + \lambda_E$
λ_A = decay constant of polonium-218
λ_E = removal constant for methods of removal other than radioactive decay, such as plate-out.

(Here the decline in the activity of radon-222 has been ignored because of its relatively long half-life of 3·8 days).

For $t \gg 1$, $Q_A = \frac{\lambda_{Rn} Q_{Rn}}{\lambda}$

If radioactive decay is the only method of removal, $\lambda = \lambda_A$, but the activity of polonium-218 in Becquerels (Bq), disintegrations per second, is $\lambda_A Q_A$, so the activity of polonium-218 is

$$\lambda_A Q_A = \frac{\lambda_{Rn} Q_{Rn}}{\lambda_A}\lambda_A = \lambda_{Rn} Q_{Rn}$$

Similarly for the other daughters of radon-222, if the only method of removal is radioactive decay, their activities will also be $\lambda_{Rn} Q_{Rn}$, and radioactive equilibrium between radon and its daughters exists.

If however, there are other methods of removal of the daughters with a total removal constant λ_E, then the activity of polonium-218 will be

$$\lambda_A Q_A = \lambda_{Rn} Q_{Rn} \cdot \frac{\lambda_A}{\lambda_A + \lambda_E}\ \text{Bq}$$

The activity of lead-214 with radioactive decay constant λ_B will be

$$\lambda_B Q_B = \lambda_A Q_A \frac{\lambda_B}{\lambda_B + \lambda_E} = \lambda_{Rn} Q_{Rn} \frac{\lambda_A}{\lambda_A + \lambda_E} \frac{\lambda_B}{\lambda_B + \lambda_E}\ \text{Bq}$$

and the activity of bismuth-214 with radioactive decay constant λ_C

$$\lambda_C Q_C = \lambda_B Q_B \frac{\lambda_C}{\lambda_C + \lambda_E} = \lambda_{Rn} Q_{Rn} \frac{\lambda_A}{\lambda_A + \lambda_E} \cdot \frac{\lambda_B}{\lambda_B + \lambda_E} \cdot \frac{\lambda_C}{\lambda_C + \lambda_E}\ \text{Bq}$$

The electrostatic precipitator used had a specification which would allow 8 per cent of the room air to be processed each minute when operated in a room of maximum recommended size (for the precipitator). Under those conditions, the activity of polonium-218 will be 73 per cent of its equilibrium value, lead-214 will be 17 per cent and bismuth-214 5 per cent. This gives a reduction in WL by a factor of 5·5 from [illegible] equilibrium value. Since the room used in this experiment was smaller than the maximum for the precipitator about 14 per cent of the air was processed each minute, giving an expected reduction in WL by a factor of 8·2.

Note: A closed system was assumed, but a constant level of radon may be obtained in a room with a constant production rate of radon and a constant ventilation rate. In this case the same equations hold if λ_T is substituted for λ_E with
$\lambda_T = \lambda_E + \lambda_V$
λ_V = removal constant for ventilation.