EXTERNAL RADON DISTURBANCE OF ^{14}C MEASUREMENTS IN GAS-PROPORTIONAL COUNTERS

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ABSTRACT. Low-level detector systems for ^{14}C dating are frequently located in underground or basement laboratories to reduce the influence of cosmic radiation. However, if careful precautions are not taken, the presence of radioactive radon gas may severely disturb the analytical results. Measurements of the influence of radon on a proportional counter system, show that a radon level of 100 Bq m^{-3}, a level not uncommon in basement rooms, is sufficient to produce an unacceptable uncertainty in the ^{14}C results. Radon levels of up to 1500 Bq m^{-3} can be demonstrably reduced to about 30 Bq m^{-3}, using a separate ventilation system that generates a slight overpressure in the laboratory.

INTRODUCTION

International intercomparisons of results from radiocarbon dating have shown that a large number of laboratories state underestimated uncertainties. Close attention should thus be paid to every parameter in the measurement procedure that can contribute to the uncertainty. One such parameter is the contribution of external radon to the background rate recorded by gas-proportional counting systems.

In the early 1960s, fluctuations in background due to radon in laboratory air were reported for a detector system comprising a large NaI detector (Stenberg and Olsson 1968). Later, an increase in background due to radon was reported in a ^{14}C gas-proportional counting system (Freundlich 1972). Reports of similar disturbances have been made in recent years for ultra low-level germanium diode systems (Heusser and Wojcik 1992).

A few years ago, increased fluctuations in the background of the ^{14}C proportional counters were observed at the Laboratory for Isotope Geology in Stockholm. Increased inflow of radon was suspected as the cause of these fluctuations. The fluctuations have now been studied in order to determine quantitatively the relation between the radon concentration and the detector background levels.

Radon Concentration in Indoor Air

The element radon has three isotopes in the uranium and thorium series:

^{232}Th series (thoron):	^{220}Rn	half-life 54.5 sec
^{235}U series (actinon):	^{219}Rn	half-life 3.92 sec
^{238}U series (radon):	^{222}Rn	half-life 3.82 days

Radon is an inert gas that is soluble in water, invisible at room temperature and odorless. It is denser than N_2 and O_2, but is uniformly mixed in indoor air. It is environmentally widespread in bedrock, groundwater, soil and building materials. Radon concentration in indoor air is determined by three factors:

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1. *The concentration of radon in the ground, building material and tapwater.* Various types of concrete and plasters can outgas radon. Also, water may contain radon, which is brought into the air when the water is tapped. The most serious inflow of radon, however, is from the soil and the bedrock. Taking basement rooms as typical for radiocarbon laboratories, we can expect a mean radon concentration in the range of *ca.* 40–160 Bq m^{-3}, if no special ventilation precautions are taken (White *et al.* 1992). Large geographical differences exist, however, because of variations in the soil and bedrock radon levels.
2. *Ventilation.* Normally there is a small pressure difference between indoor and outdoor air. This difference can be maintained by a ventilation system, but is often the result of a temperature difference between the indoor and outdoor air. A lower indoor pressure may cause radon to diffuse into the house through the foundation. Natural ventilation due to the pressure difference will normally increase with increased difference between the temperature inside and outside the house. Thus, it is often possible to correlate the inflow of radon with the outside temperature. Depending on the ventilation system, the radon inflow can either increase or decrease with the outside temperature (Hubbard, Hagberg and Enflo 1992). This will cause the radon inflow to vary from day to night and from summer to winter. In addition, depending on how the wind is blowing, the pressure within a building varies, and, hence, so will the inflow of radon. It is often possible to reduce drastically the radon level in a house by increasing the inlet of outdoor air.
3. *Entryways leading radon into indoor air.* Being an inert gas, radon can diffuse far away from its origin. However, in indoor air, usually only the concentration of ^{222}Rn is critical, given the short half-lives of the other isotopes. ^{220}Rn is sometimes present in wall material, but its contribution can often be removed by an extra layer of paint.

Radon Effects on Proportional Counters

The daughter products of ^{222}Rn (Table 1) easily attach to dust particles and aerosols, as well as air molecules. Variation in the amount of aerosols will affect the amount of radon daughters in the air. Radon and its daughter products will produce α, β and γ radiation, of which the α particles have no importance to the detector background. On the other hand, β particles from the decay of ^{214}Bi may penetrate detector walls several millimeters thick and cause background pulses. Following an α or β decay, the nucleus often emits γ rays. The dominant γ radiation is produced by ^{214}Pb and ^{214}Bi. These γ rays may penetrate the proportional counters, and thus increase the background.

Radon Background Disturbances in the Laboratory for Isotope Geology in Stockholm

The Stockholm Radiocarbon Dating Laboratory has two multidetector proportional counter systems, the older system 1 and the more modern system 2, situated in separate rooms on the ground floor of the Swedish Museum of Natural History. Both systems include six sample detectors. System 1 consists of two sections, each with a separate proportional guard counter ring, inside a common iron chamber. System 2, built by Wallac OY, has pulse-height and rise-time filters and a liquid scintillation guard.

Long-term monitoring of the background levels showed an increase in 1990 (Fig. 1). A serious water leak in 1990 made it necessary to repair the floor of the laboratory. During repairs, the ground underneath was filled with a porous material for draining water. After this operation the background increased and the fluctuations were greater than before. Close study of measurement results after the repair indicated daily fluctuations in the detector background as well as guard counters, with higher values recorded at night (Fig. 2). Fluctuations decreased during the weekends, but the count rate stayed high. The fluctuations were found to match the active periods of the ventilation system, which was turned off during weekends and nights, indicating the presence of radon. To confirm this, a quick test was made during a weekend by simply opening one of the laboratory windows. The background of all sample detectors and the count rate of the guard counters decreased then stabilized at a low level for as long as the window remained open. It seemed plausible that the porous material under the floor of the detector room was a passageway for radon inflow from the ground below. Work was initiated to reduce the radon level and to measure quantitatively the relation between radon concentration and background count rate. These measurements were made on both systems; the results from system 1 are presented in detail here.

TABLE 1. The main decay branch of ^{222}Rn with the daughters' half-lives and their most common γ radiation. Potential disturbance of proportional counters derives mainly from daughters ^{214}Pb and ^{214}Bi.

Isotope	Half-life	Decay	Decay energy (MeV)	Most common γ radiation (MeV)
^{222}Rn	3.82 day	α	5.59	
^{218}Po	3.1 min	α	6.11	
^{214}Pb	26.8 min	β^-	1.023	0.24
				0.30
				0.35
^{214}Bi	19.9 min	β^-	3.27	0.61
				0.77
				1.12
				1.24
				1.76
				2.20
^{214}Po	0.00016 sec	α	7.83	
^{210}Pb	22.3 yr	β^-	0.064	
^{210}Bi	5.01 day	β^-	1.163	
^{210}Po	138.4 day	α	5.407	
^{206}Pb	Stable			

After installation of separate ventilation, the background level dropped to a level even lower than before the repair of the floor. Hence it is likely that a certain level of radon disturbance was also present before the repair of the floor. The new ventilation system blows outdoor air through filters into the laboratory, creating an overpressure of about 20 Pa. This pressure is enough to keep the radon level below 30 Bq m^{-3}.

In the second laboratory, the floor was sealed, the walls painted with an extra layer and a separate ventilation system installed. Here the sample detectors also fit tightly into the tubes in the scintillation guard so that only a small free space remains between the detector and the guard. The radon fluctuation in the second detector room is smaller than in the first.

METHODS AND RESULTS

Radon and radon daughter levels were measured with two different types of detectors. The radon daughter level was measured with a semiconductor detector (Scintrex/Nucletron RTA30), and the radon level with a Lucas cell (Eberliner RGM3).

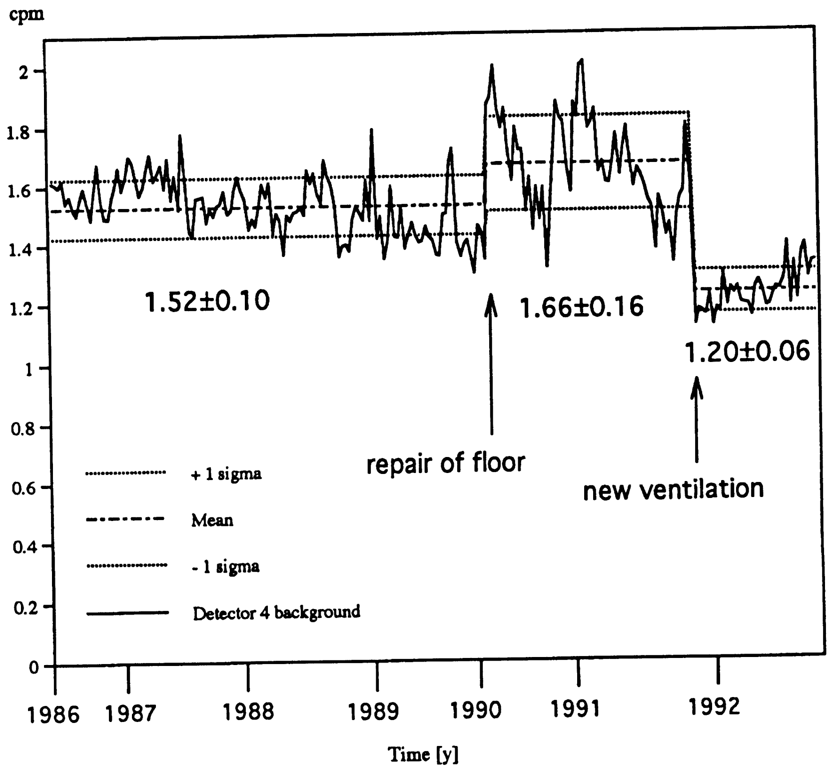

Fig. 1. Background in detector 4, in system 1, showing increase in background fluctuations after floor repair. The mean background and the mean square standard deviation are shown for three time periods. (Time scale not linear).

During the measurements the doors and windows in the laboratory were closed and the new ventilation was turned off, in order to allow the radon concentration to build up. As the humidity level was extremely low during the period, we brought it up to a normal level with an air humidifier. (A higher air humidity will often increase the quantity of radon daughters in the air).

The effect that increased radon concentration had on one guard and on detector 0 during a 90-h period is shown in Figures 3 and 4. When the ventilation was turned off after 20 h, the radon level quickly increased in the laboratory. The decrease in radon level after 36 h is a typical day and night effect caused by the temperature-dependent pressure change described earlier. After 64 h the ventilation was turned on again and the radon level immediately dropped. This quick response of the guard counters to the rise of radon concentration in the laboratory indicates a rapid air exchange between the interior and the exterior of the passive shield.

From the measurements that are plotted in Figures 3 and 4 we have calculated the average disturbance in the counters and in the guards for a 28-h period starting at 36 h and ending at 64 h, the period during which the radon level was high. From these average values we have calculated the increase in background, Δn_x, in detector x for every 100 Bq m^{-3} of radon. n_x is the normal background without radon. m_x is the net modern (95% of NBS HOxI) (Table 2). In the proportional counter guards, the count rate went up by 15 cpm per 100 Bq m^{-3} of radon. The normal count rate is about 2000 cpm.

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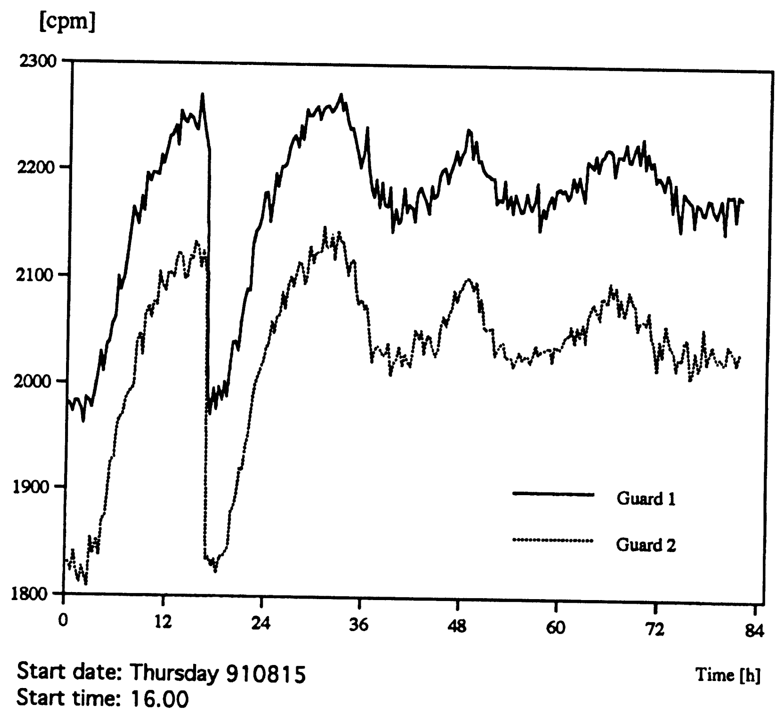

Fig. 2. Daily variations in guard count rate in system 1 due to radon inflow. Variations decreased but stayed on a higher level during weekends owing to less ventilation.

TABLE 2. Results of Measurements in System 1

Detector x	Δn_x (cpm per 100 Bq m^{-3})	n_x (cpm)	m_x (cpm)
0	0.023	0.764 ± 0.02	12.8 ± 0.03
2	0.046	1.374 ± 0.03	18.65 ± 0.03

Analysis of these measurements requires consideration of the physical dimensions of the detector system. Large volumes inside the passive shielding should increase the effects of radon. Details of the physical dimensions of system 1 are as follows:

The volume between the guard systems and the passive shielding in system 1 ~ 100 liters.

The volume between detectors and inner lead shield:	Detector 0 ~ 0.7 liter
	Detector 2 ~ 0.2 liter
Thickness of detector walls:	Detector 0 = 3–13 mm
	Detector 2 = 1 mm

Detector 2 has shown a higher sensitivity to radon then detector 0 even though it has a smaller surrounding free space. This is probably due to the thinner detector wall, which increases the contribution from β radiation.

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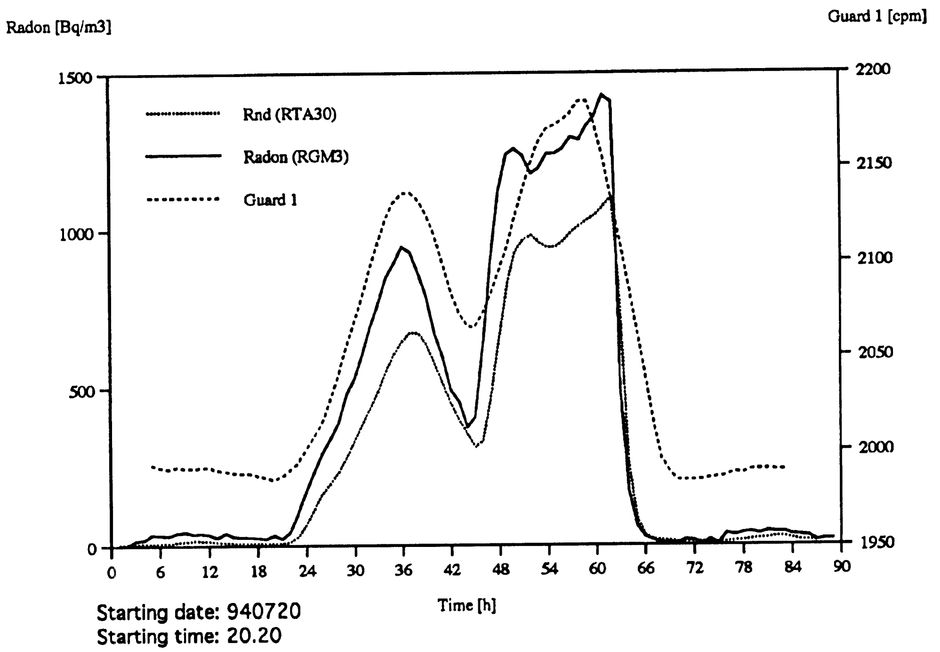

Fig. 3. The radon level increases after 20 h when the ventilation is turned off. The decrease after 36 h is a typical day-night effect that often is seen in houses with radon problems. The radon level drops after 64 h when the ventilation is turned on. The plots are smoothed with a 5-h window.

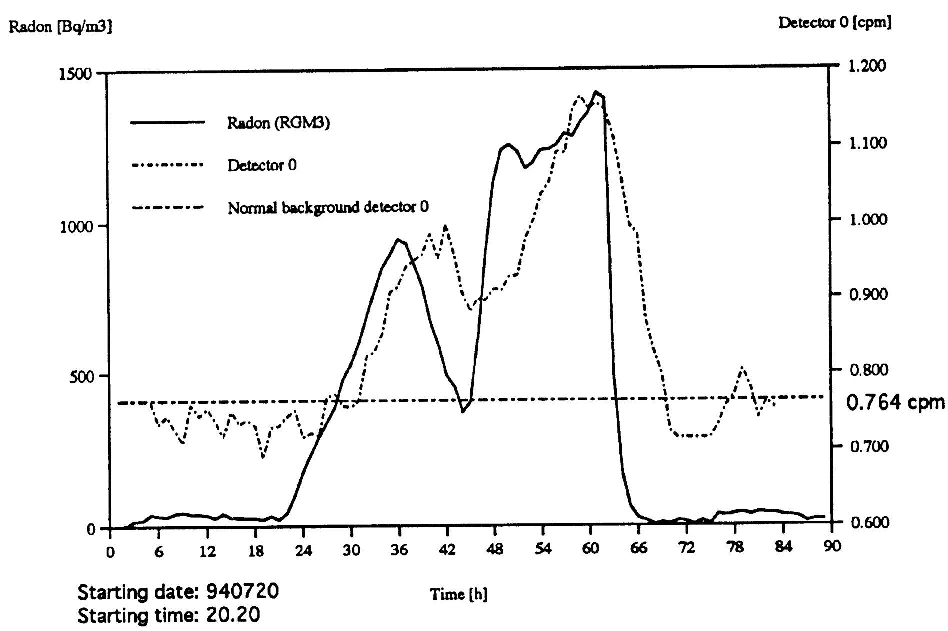

Fig. 4. Increase in radon and detector 0 count rate when ventilation is turned off after 20 h. The radon level drops after 64 h when the ventilation is turned on. The plots are smoothed with a 5-h window.

Conclusion

The effect of radon on a proportional counter system depends on the physical dimensions of the counters and the amount of free space between the guard systems and the passive shielding. The aerosol level can also change the effect of radon. These factors must be taken into consideration when the background contribution of radon is evaluated. Although our investigation was made on a proportional counter system, it is also likely that low-level liquid scintillation systems can be disturbed by radon.

It is difficult to calculate a strict upper radon limit that can be tolerated, owing to the irregularities of the distribution of the radon fluctuations. The radon disturbance can show extreme temporal variations, giving very high radon levels one day and almost nothing the next. The large fluctuations increase the necessity to minimize the amount of radon present in the laboratory. As an example, we consider the effect of adding 100 Bq m^{-3} radon disturbance to a sample measurement in detector 0, which in our case had the smallest sensitivity for radon. It would increase the age for a modern sample by *ca.* 15 yr, and by 1100 yr for a 35,000-yr-old sample.This indicates that even small radon amounts of *ca.* 100 Bq m^{-3} must be considered, and suggest that the radon level should be reduced as much as possible. Fortunately, as shown in the Stockholm laboratory, levels of up to 2000 Bq m^{-3} can quite easily be minimized. The procedure used in the Stockholm laboratory to reduce radon disturbance with a separate ventilation system is neither expensive nor complicated.

An easy way to check the radon level is to use the guard system as a detector of radon, by monitoring the count rate and fluctuations when it is flushed with radon-free air compared to when it is not. (This was how the radon problem first was studied in the Stockholm laboratory.)

Our measurements indicate that it is necessary to look carefully into the possibility of radon disturbances in high-precision ^{14}C dating with gas-proportional counters, especially as these detector systems are frequently placed in basement rooms, where the risk of a high radon level is greater.

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