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Volume IX: Radon Occurrence in the Natural Environment

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Radon Occurrence in the Natural Environment

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COMPARATIVE DOSIMETRY OF RADON IN MINES AND HOMES: AN OVERVIEW OF THE NAS REPORT

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ABSTRACT

The findings of the recent report by a National Academy of Sciences panel on radon dosimetry are reviewed. The committee was charged with comparing exposure-dose relations for the circumstances of exposures in mines and homes. The community first obtained data on the various parameters included in dosimetric lung models and then selected values that it judged to be best supported by the available evidence. Dosimetric modeling was used to calculate the ratio of exposure to radon progeny to dose of alpha energy delivered to target cells for various scenarios. The committee's modeling shows that exposure to radon progeny in homes delivers a somewhat lower dose to target cells than exposure in mines; this pattern was found for infants, children, men, and women.

INTRODUCTION

Radon, an inert gas, is a naturally occurring decay product of radium-226, the fifth daughter of uranium-238. Radon decays with a half-life of 3.82 days into a series of solid, short-lived progeny; two of these progeny, polonium-218 and polonium-214, emit alpha particles. When radon progeny are inhaled and these alpha emissions occur within the lungs, the cells lining the airways may be injured and damage to the genetic material of the cells may lead to the development of cancer.

Radon has been linked to excess cases of lung cancer in underground miners since the early decades of the twentieth century. Epidemiologic evidence on radon and lung cancer, as well as other diseases is now available from about 20 different groups of underground miners (1,2). Many of these studies include information on the miners' exposure to radon progeny and provide estimates of the quantitative relation between exposure to progeny and lung cancer risk (2,3); the range of excess relative risk coefficients, describing the increment in risk per unit of exposure is remarkably narrow in view of the differing methodologies of these studies (2).

As information on air quality in indoor environments was collected during the last 20 years, it quickly became evident that radon is ubiquitous indoors and that concentrations vary widely and may be as high as levels in underground mines in some homes. The well-documented and causal association of radon with lung cancer in underground miners appropriately raised concern that radon exposure might also cause lung cancer in the general population. The risk of indoor radon has been primarily assessed by using risk assessment approaches that extend the risks found in the studies of miners to the general population. Risk models that can be used for this purpose have been developed by committees of the National Council on Radiation Protection and Measurements (NCRP) (4), the International Commission on Radiological Protection (5) (1987), and the National Academy of Sciences (Biological Effects of Ionizing Radiation (BEIR) IV Alpha Committee) (1).

Extrapolation of the lung cancer risks in underground miners to the general population is subject to uncertainties related to the differences between the physical environments of homes and mines, the circumstances and temporal patterns of exposure in the two environments, and potentially significant biological differences between miners and the general population (Table 1). A number of these factors may affect the relation between exposure to radon progeny and the dose of alpha-particle energy delivered to target cells in the tracheobronchial epithelium; these factors include the activity-aerosol size distribution of the progeny, the ventilation pattern of the exposed person, the morphometry of the lung, the pattern of deposition and the rate of clearance of deposited progeny, and the thickness of the mucous layer lining the airways.

The activity-aerosol size distribution refers to the physical size distribution of the particles containing the alpha activity. The term "unattached fraction" has historically been applied to progeny existing models that it judged to be best supported by the available evidence. The committee then utilized a dosimetric model, developed in part by the Task Group of the International Commission for Radiological Protection, to compare exposure-dose relations for exposure to radon progeny in homes and in mines. While the report provides the exposure-dose figures, the committee expressed its principal findings as a ratio, termed K in the BEIR IV report (1). K, a unitless measure, represents the quotient of the dose of alpha energy delivered per unit of exposure in a home to the dose per unit exposure for a male miner exposed in a mine. If the K factor exceeds unity, the delivered dose per unit exposure is greater indoors whereas if it is less than unity, the delivered dose per unit exposure is less indoors.

Factors other than lung dosimetry of radon progeny also introduce uncertainty in extrapolating risks from the studies of underground miners to the general population. The committee briefly reviewed the evidence on cigarette smoking, tissue damage, age at exposure, sex, and exposure pattern. These sources of uncertainty were considered in a qualitative rather than a quantitative fashion.

THE COMMITTEE'S FINDINGS

The committee selected several different sets of exposure conditions in homes and in mines (Table 2,3). The mining environment includes the areas of active mining, the haulage drifts, and less active and dusty areas such as lunch rooms. In some analyses, the values for active mining and haulage ways were averaged to represent typical conditions. Separate microenvironments considered in the home included the living room and the bedroom. Parameters for the living room and the bedroom were averaged to represent a typical scenario for the home. The effects of cooking and cigarette smoking on radon progeny aerosol characteristics were also considered. While the contrast between the home and mining environments was somewhat variable across the scenarios, homes were characterized as having greater unattached fractions and smaller particles. Higher average minute volumes were assumed for the mining environment (Table 2,3).

The committee also examined uncertainties associated with other assumptions in the dosimetric model. Doses to basal and secretory cells in the tracheobronchial epithelium were calculated separately, because all types of cells with the potential to divide were considered to be potential progenitor cells for lung cancer. The committee also compared the consequences of considering: lobar and segmental bronchi rather than all bronchi as the target; radon progeny as insoluble or partially soluble in the epithelium; of breathing through the oral or nasal route exclusively; of varying the thickness of the mucus lining the epithelium and the rate of mucociliary clearance; and cellular hyperplasia leading to thickening or injury causing thinning of the epithelium.

Across the wide range of exposure conditions and exposed persons considered by the committee, most values of K were below unity (Table 4). For both secretory and basal cells, K values indicated lesser doses of alpha energy per unit exposure, comparing exposures of infants, as ions, molecules, or small clusters; the "attached fraction" designates progeny attached to ambient particles (6). Using newer methods for characterizing activity-aerosol size distributions, the unattached fraction has been identified as ultrafine particles in the size range of 0.5 to 3.0 nm (6). Typically, mines have higher aerosol concentrations than homes and the unattached fraction would be expected to be higher in homes than in mines. Because of differing sources of particles in the two environments, aerosol size distributions could also plausibly differ between homes and mines.

The physical work involved in underground mining would be expected to increase the amount of air inhaled in comparison with the generally sedentary activities of time spent at home. The greater minute ventilation of miners would result in a higher proportion of the inhaled air passing through the oral route, in comparison with ventilation during typical activities in residences. The physical characteristics of the lungs of underground miners, almost all adult males, differ significantly from those of infants, children and thickness of the epithelial layer could also plausibly differ, comparing miners with the general population, because of the chronic irritation by dust and fumes in the mines.

Methods are available for characterizing the effects of these factors on the relation between exposure to radon progeny and the dose of alpha energy delivered to target cells in the respiratory tract. Using models of the respiratory tract, the dose to target cells in the respiratory epithelium can be estimated for the circumstances of exposure in the mining and indoor environments. One of the recommendations of the 1988 BEIR IV Report (1) was that "Further studies of dosimetric modeling in the indoor environment and in mines are necessary to determine the comparability of risks per WLM [working level month] in domestic environments and underground mines". The BEIR IV Report had included a qualitative assessment of the dosimetry of progeny in homes and in mines, but formal modeling was not carried out.

Consequently, the U.S. Environmental Protection Agency asked the National Research Council to conduct a study addressing the comparative dosimetry of radon progeny in homes and in mines. This paper reviews the findings of the recently published report of the committee (Panel on Dosimetric Assumptions Affecting the Application of Radon Risk Estimates). The panel was constituted with the broad expertise, covering radon measurement and aerosol physics, dosimetry, lung biology, epidemiology, pathology, and risk assessment, needed for this task.

THE COMMITTEE'S APPROACH

To address the charge of undertaking further dosimetric modeling, the committee obtained data on the various parameters included in dosimetric lung models that contributed to uncertainty in assessing the risk of indoor radon. The committee not only reviewed the literature, but obtained recent and unpublished information from several investigators involved in relevant research. After completing this review, the committee selected values for parameters in dosimetric children, men and women in homes with exposures of male miners underground. While the highest values of K were calculated for children, the values for children did not exceed unity, suggesting that children exposed to radon progeny are not at greater risk for lung cancer on a dosimetric basis.

The committee explored the sensitivity of the K factors to underlying assumptions in the dosimetric model. The general pattern of the findings was comparable for secretory and basal cells. The K factors remained below unity regardless of whether the radon progeny were assumed to be insoluble or partially soluble in the epithelium. The K factor was also not changed substantially with the assumption that lobar and segmental bronchi, rather than all bronchi, are the target. Assumptions regarding breathing route also had little impact. After the committee had completed its principal analysis, new data became available suggesting that recent higher values for nasal deposition reported by Cheng et al. (7) might be preferable to lower values from the 1969 report of George and Breslin (8); other new evidence suggested that a value of 0.15 um should be used for aerosol size in the haulage drifts. Inclusion of these two modifications of the committee's preferred parameter values in the dosimetric model reduced the values of K by about 20 percent.

The committee did not attempt to reach quantitative conclusions concerning sources of uncertainty not directly addressed by the dosimetric modeling. It noted the paucity of data on such factors as cigarette smoking, age at exposure and particularly the effect of exposure during childhood, and exposure pattern. The evidence on these factors received detailed review in the BEIR IV report (1) and the present committee did not reach any new conclusions on these sources of uncertainty. The committee also commented on the potential effects of the miners' exposures to dust and fumes while underground. Increased cell turnover associated with these exposures may have increased the risk of radon exposure for the miners.

SUMMARY

The Panel on Dosimetric Assumptions Affecting the Application of Radon Risk Estimates comprehensively reviewed the comparative dosimetry of radon progeny in homes and in mines. The committee's modeling shows that exposure to radon progeny in homes delivers a somewhat lower dose to target cells than exposure in mines; this pattern was found for infants, children, men, and women. This finding was not sensitive to specific underlying assumptions in the committee's modeling. Assuming that cancer risk is proportional to dose of alpha energy delivered by radon progeny, the committee's analyses suggests that direct extrapolation of risks from the mining to the home environment may overestimate the numbers of radon-caused cancers.

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TABLE 1. POTENTIALLY IMPORTANT DIFFERENCES BETWEEN EXPOSURE TO RADON IN THE MINING AND HOME ENVIRONMENTS*

Physical Factors

Aerosol characteristics: Greater concentrations in mines; differing size distributions

Attached/unattached fractions: Greater unattached fraction in homes

Equilibrium of radon/decay products: Highly variable in homes and mines

Activity Factors

Amount of ventilation: Probably greater for working miners than for persons indoors

Pattern of ventilation: Patterns of oral/nasal breathing not characterized, but mining possibly associated with greater oral breathing

Biological Factors

Age: Miners have been exposed during adulthood; entire spectrum of ages exposed indoors

Gender: Miners studied have been exclusively male; both sexes exposed indoors

Exposure pattern: Miners exposed for variable intervals during adulthood; exposure is lifelong for the population

Cigarette smoking: The majority of the miners studied have been smokers; only a minority of U.S. adults are currently smokers

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*Taken from Table 1-2 in reference (6).

TABLE 2. ASSUMPTIONS FOR EXPOSURE SCENARIOS ASSUMED FOR MINES AND HOMES*

SUMMARY OF RADON PROGENY AEROSOL CHARACTERISTICS ASSUMED TO REPRESENT EXPOSURE CONDITIONS IN MINES AND HOMES

Exposure Scenario	f_p	AMD of Room Aerosol (μm)	AMD of Aerosol in respiratory tract (μm)
<u>Mine</u>			
Mining	0.005	0.25	0.5
Haulage drifts	0.03	0.25	0.5
Lunch room	0.08	0.25	0.5
<u>Living Room</u>			
Normal	0.08	0.15	0.3
Smoker - average	0.03	0.25	0.5
- during smoking	0.01	0.25	0.5
Cooking/vacuuming	0.05	0.02/0.15+ (15%/80%)	0.02/0.3 (15%/80%)
<u>Bedroom</u>			
Normal	0.08	0.15	0.3
High	0.16	0.15	0.3

*Based on Tables 3-1 and 3-2 in reference 6.

+The radon progeny aerosol produced by cooking/vacuuming has three size modes; 5% of potential alpha energy is unattached, 15% has an AMD of 0.02 m, and 80% has an AMD of 0.15 μm. The 0.02 μm AMD mode is hydrophobic and does not increase in size within the respiratory tract.

TABLE 3. ASSUMPTIONS FOR EXPOSURE SCENARIOS ASSUMED FOR MINES AND HOMES*

LEVELS OF PHYSICAL EXERTION AND AVERAGE MINUTE VOLUMES ASSUMED FOR UNDERGROUND MINERS AND FOR ADULTS IN THE HOME

Exposure Scenario	Level of Exertion	Average $\dot{V}_E$ (liters/min)	
		Man	Woman
Underground Mine			
Mining	25% heavy work/75% light work	31	--
Haulage way	100% light work	25	--
Lunch room	50% light work/50% rest	17	--
Home-Living Room			
Normal and smoker	50% light work/50% rest	17	14
Cooking/vacuuming	75% light work/25% rest	21	17
Home-Bedroom			
Normal and high	100% sleep	7.5	5.3

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*Based on Tables 3-1 and 3-2 in reference 6.

TABLE 4. SUMMARY OF K FACTORS FOR BRONCHIAL DOSE CALCULATED FOR NORMAL PEOPLE IN THE GENERAL ENVIRONMENT RELATIVE TO HEALTHY UNDERGROUND MINERS*

Subject Category	K Factor for Target Cells Secretory	 Basal
Infant, age 1 month	0.74	0.64
Child, age 1 year	1.00	0.87
Child, age 5-10 years	0.83	0.72
Female	0.72	0.62
Male	0.76	0.66

*Taken from Table 5-1 in reference 6.

Session IX:

Radon Occurrence in the Natural Environment

COMBINING MITIGATION & GEOLOGY: INDOOR RADON REDUCTION BY ACCESSING THE SOURCE

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ABSTRACT

Soil radon testing has shown that radon sources are concentrated in narrow linear areas congruent with local geology in the Eastern Piedmont, which should also hold true in any folded mountain belt region with heterogenous geology.

In existing buildings, if micromanometer tests indicate poor communication in the sub-slab environment, soil radon concentration gradients can be mapped with instantaneous sub-slab radon measurements. By then orienting these difficult-to-mitigate homes on a geologic map, we have been able to predict the location of the radon source adjacent to foundation walls. Tapping these source areas with a multi-duct sub-slab depressurization system has been shown to be effective in achieving optimum radon reductions.

By using this method of radon soil testing for the construction of new large buildings, such as schools, to locate areas of sub-slab depressurization, maximum indoor radon reductions can be achieved with minimal installations.

In large buildings, such as schools and office buildings, and in homes without good sub-slab air-flow communication, e.g. no aggregate, we have achieved significant indoor radon reductions by sub-slab ventilation at the source of maximum soil radon concentrations using quantitative diagnostic tests which incorporate the correlation between radon soil testing and local geology.

Recent measurements of soil radon availability numbers by the author (1) have yielded correlations between indoor radon levels in homes, office buildings, and schools and the various geologic units in the Coastal Plain, Piedmont, and Mesozoic Basin. The radon availability number was determined using the equations of Nazaroff, et al (2) and Tanner (3), whereby radon availability number is a function of soil radon content, permeability, and diffusion coefficient. The equipment used consists of a Pylon radon monitor with attached Lucas cell and soil probe developed by the author. The probe has an in-line flow meter and pressure gauge (which must have an appropriate range for the permeability values inherent in the particular soils being measured) and a drying tube, cut-off valve, and Swaglok connector which attaches to the in-ground section of the probe assembly. This in-ground section consists of a three foot long metal tubing surrounded near its base by an inflatable packer to prevent atmospheric dilution.

Because soil permeabilities in the Piedmont, Coastal Plain, and the Mesozoic Basin of Northern Virginia and Maryland are low enough that radon migration is predominately diffusion driven, it was decided to calculate the radon availability number based upon the soil radon concentration and diffusion coefficient of the soil. Soils were then tested around a number of basement homes and schools remediated by RCP. Therefore good data was available on the original radon values and construction characteristics.

Determined radon availability numbers, plotted against indoor radon levels, revealed two distinct populations (Figure 1). The lower population (i.e. those with a higher radon availability number to indoor radon ratio) consists entirely of buildings having one or more of the following four factors:

1. A vented crawlspace,
2. A tight or sealed slab-wall contact,
3. A controlled fill around basement walls that has a low radon availability number,
4. No basement.

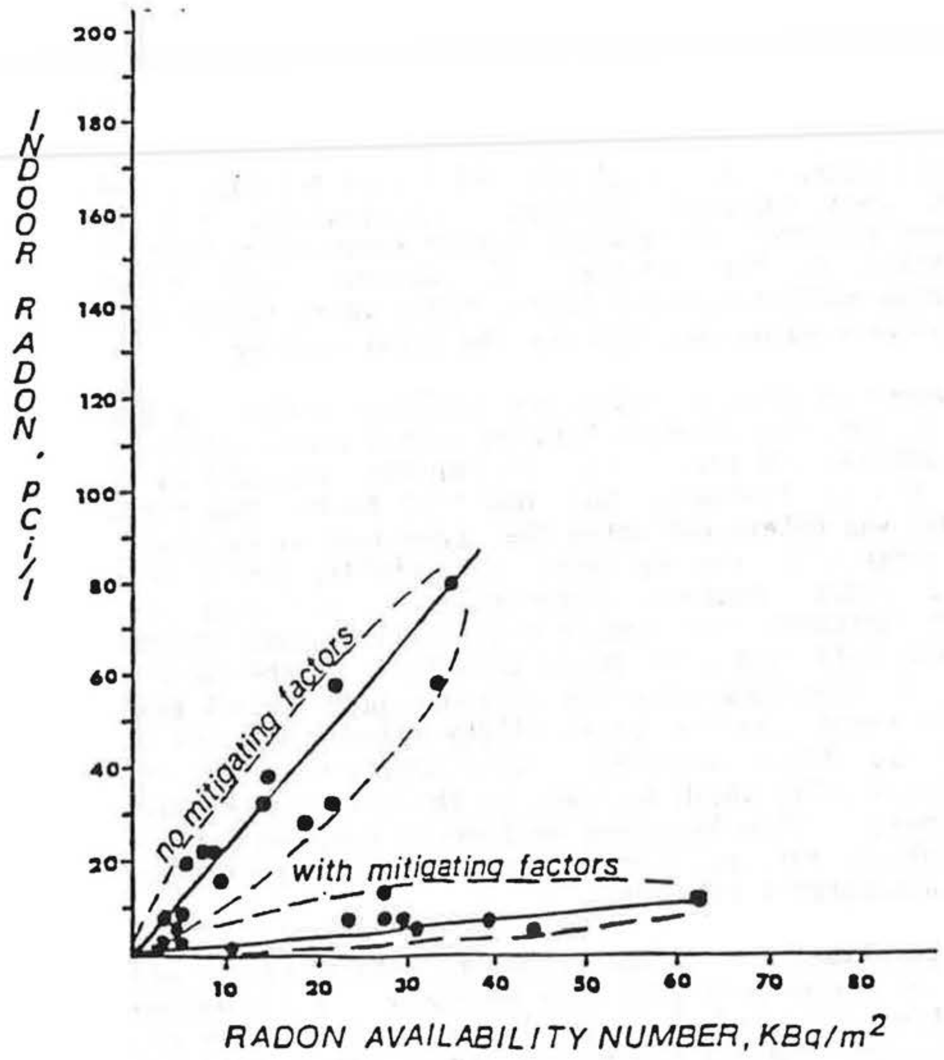

Figure 1. Correlation between soil test results (radon availability number) and indoor radon concentration.

Interestingly, the author had previously discovered in the George Mason University (GMU) Radon Study, that in the same geologic setting, basement homes with crawlspaces tended to have lower indoor radon than those without crawlspaces. Apparently this is a function of the fact that crawlspaces are normally attached at one end of the basement and have fresh air vents to the outside, at least in the local area studied. Crawlspaces are also usually separated from the rest of the basement by a wall, thereby acting as a decoupled unit without the decrease in indoor air pressure that the basement experiences.

The upper population (i.e. those with a lower radon availability number to indoor radon ratio) consists entirely of homes and schools with none of the factors inherent in the lower population. It is this trend that one would want to use to predict magnitudes of indoor radon problems, based upon soil tests for homes or buildings without any radon mitigating factors. Figure 2-7 (highest values darkened), illustrates that both slab-wall separation radon measurements (interior semi-circles) in partially completed schools have corroborated the location of maximum radon potentials determined from soil tests.

In most cases, elevated sub-slab radon levels and soil test results have been shown to be concentrated in linear areas for the various geologic units around the DC metro area, which should also hold true in any folded geologic region with heterogenous geology. These linear areas or "bands" can be one foot to a few tens of feet wide. Importantly, the orientation of the high radon potential lineations correlate well with the trend of local rock layers (generally N30°E), or with the trend of local shear fractures (generally N45°W to N60°W). For example, a boundary between high and low radon potentials is shown in Figure 2, along a N60°W fracture trend and in Figure 3, along a N45°W fracture trend. Figure 4 shows a diagonal band through the central area of the school along a N45°W trending fracture pattern. Figures 5 and 6 show correlations between high radon potentials and N30°E trending rock layers; both revealing a linear band through the interior area of the school.

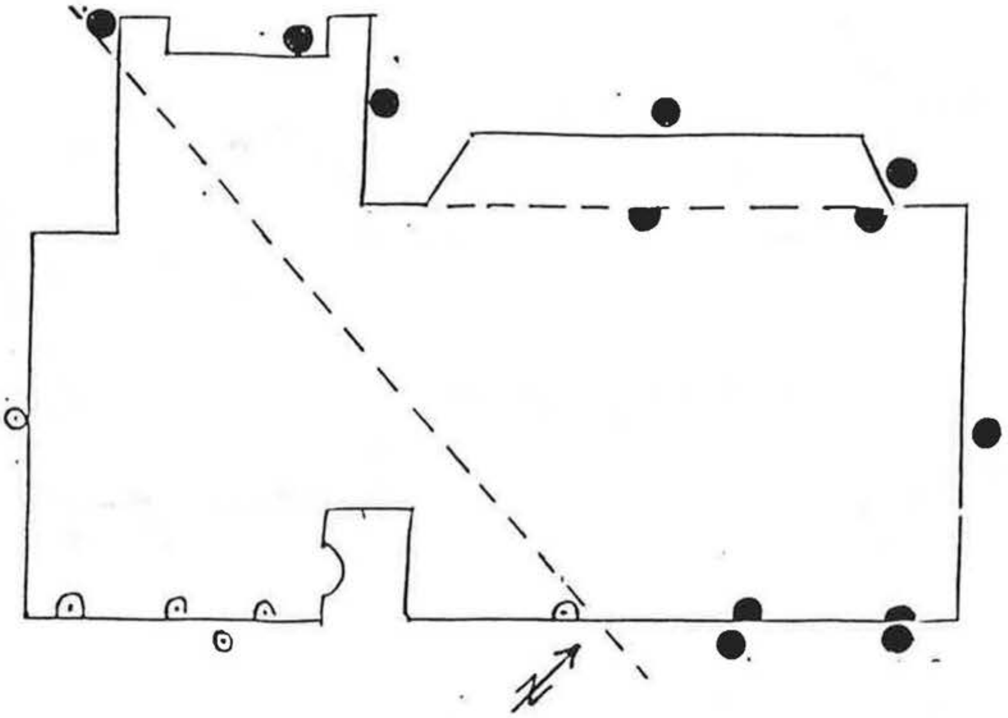

Figure 2. Footprint plan of a school showing highest soil radon potentials (RAN) and indoor slab/wall joint radon concentrations darkened. Highest soil radon potentials follow N60°W fracture trends.

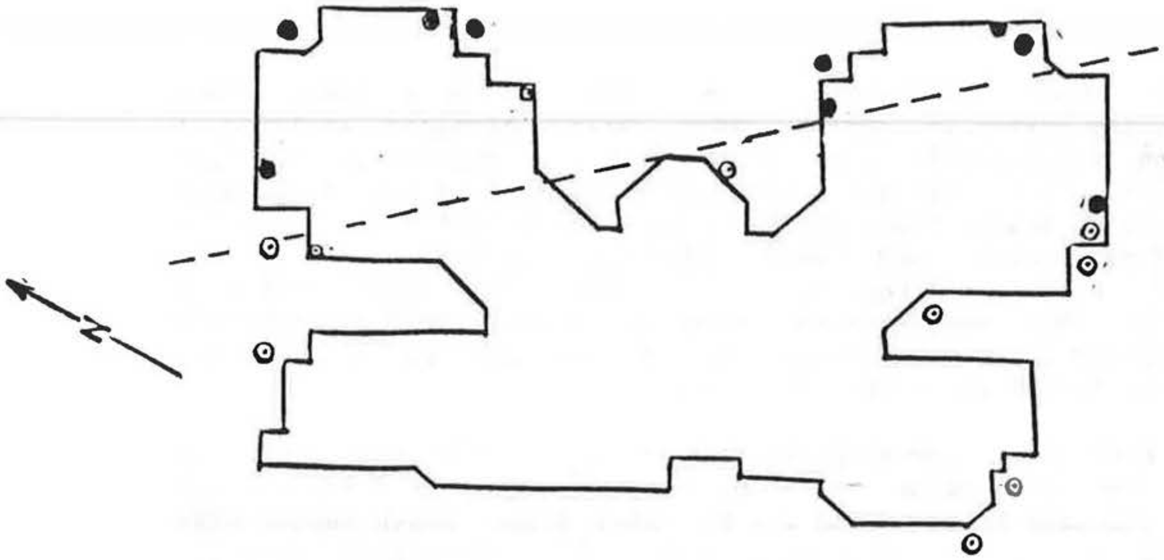

Figure 3. Footprint plan of a school showing highest soil radon potentials (RAN) and indoor slab/wall joint radon concentrations darkened. Highest soil radon potentials follow N45°W fracture trends.

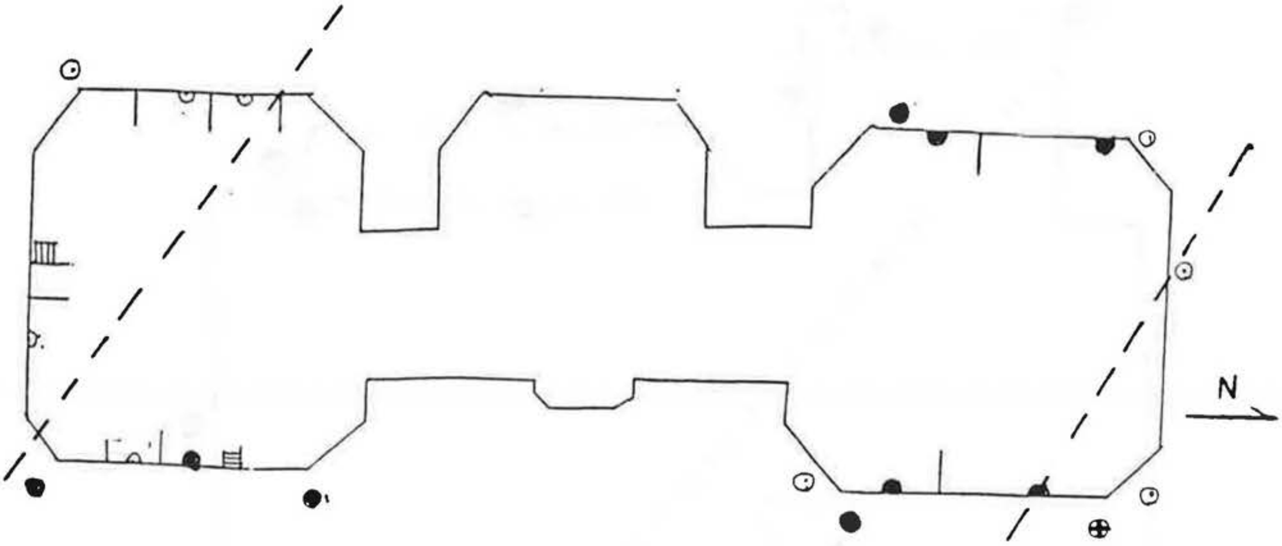

Figure 4. Footprint plan of a school showing highest soil radon potentials (RAN) and indoor slab/wall joint radon concentrations darkened. Highest soil radon potentials follow N45°W fracture trends.

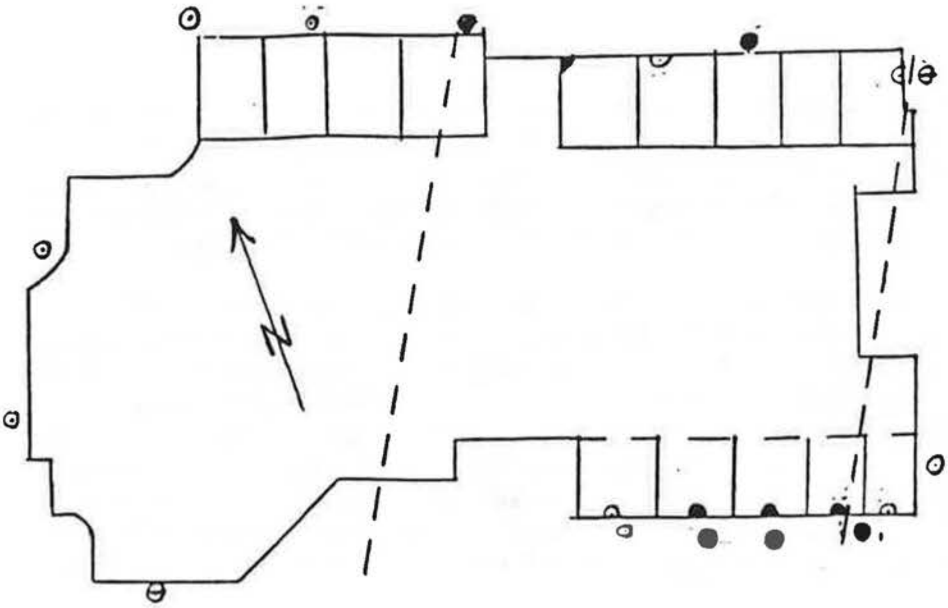

Figure 5. Footprint plan of a school showing highest soil radon potentials (RAN) and indoor slab/wall joint radon concentrations darkened. Highest soil radon potentials follow N30°E trending rock layers.

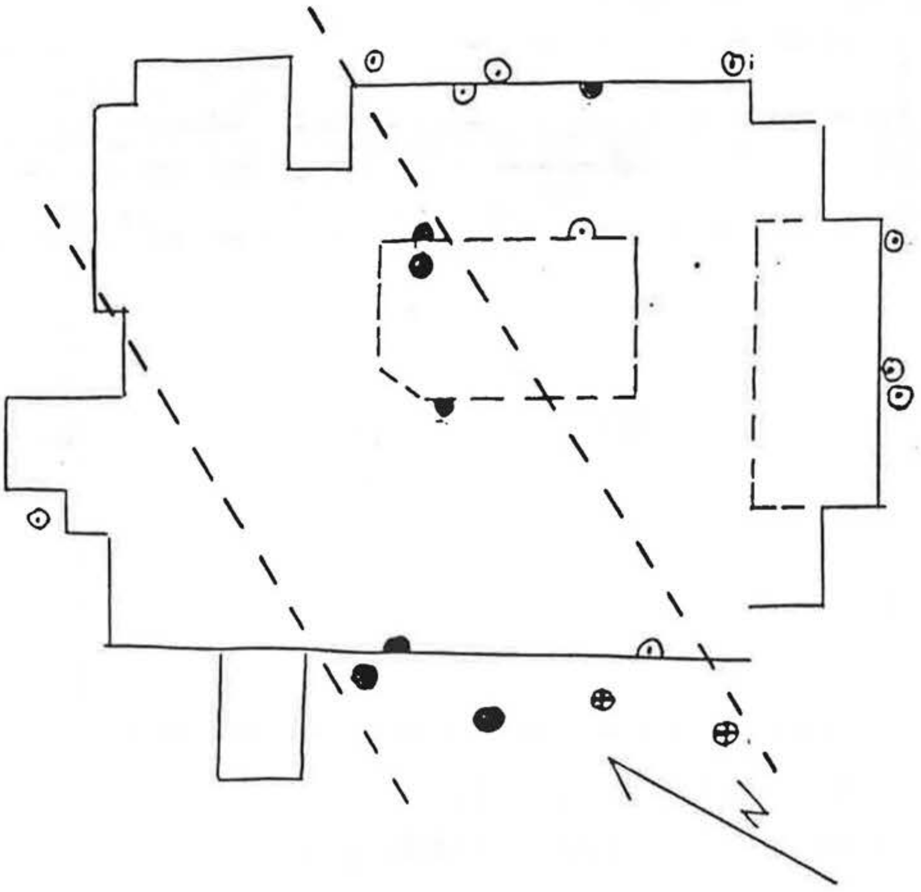

Figure 6. Footprint plan of a school showing highest soil radon potentials (RAN) and indoor slab/wall joint radon concentrations darkened. Highest soil radon potentials follow N30°E trending rock layers.

For the construction of new large buildings, such as schools, radon soil testing has proven valuable in locating the sources of maximum radon availability. By locating sub-slab ventilation points in the vicinity of these areas maximum indoor radon reductions can be achieved with minimal sub-slab ventilation installations.

In existing homes with a footprint area of less than 2000 ft², if sub-slab micromanometer tests indicate good air-flow permeability (good sub-slab communication), the location of ventilation points is not critical because one fan with one penetration will draw radon from everywhere under the slab. However, when sub-slab communication is poor, sub-slab and blockwall radon concentration gradients can be mapped with instantaneous radon measurements to determine the orientation and location of high radon potential lineations under the building, based on a knowledge of the local geology.

For example, Figure 7 shows a home with all the high sub-slab and blockwall radon levels along the NW side, indicating a linear source at the NW end oriented N30°E, parallel to the local rock layers. Micromanometer tests indicated negligible sub-slab communication. A sub-slab ventilation system, installed with one wall and three slab penetrations along the NW end, brought indoor radon levels down to 0.5 pCi/l.

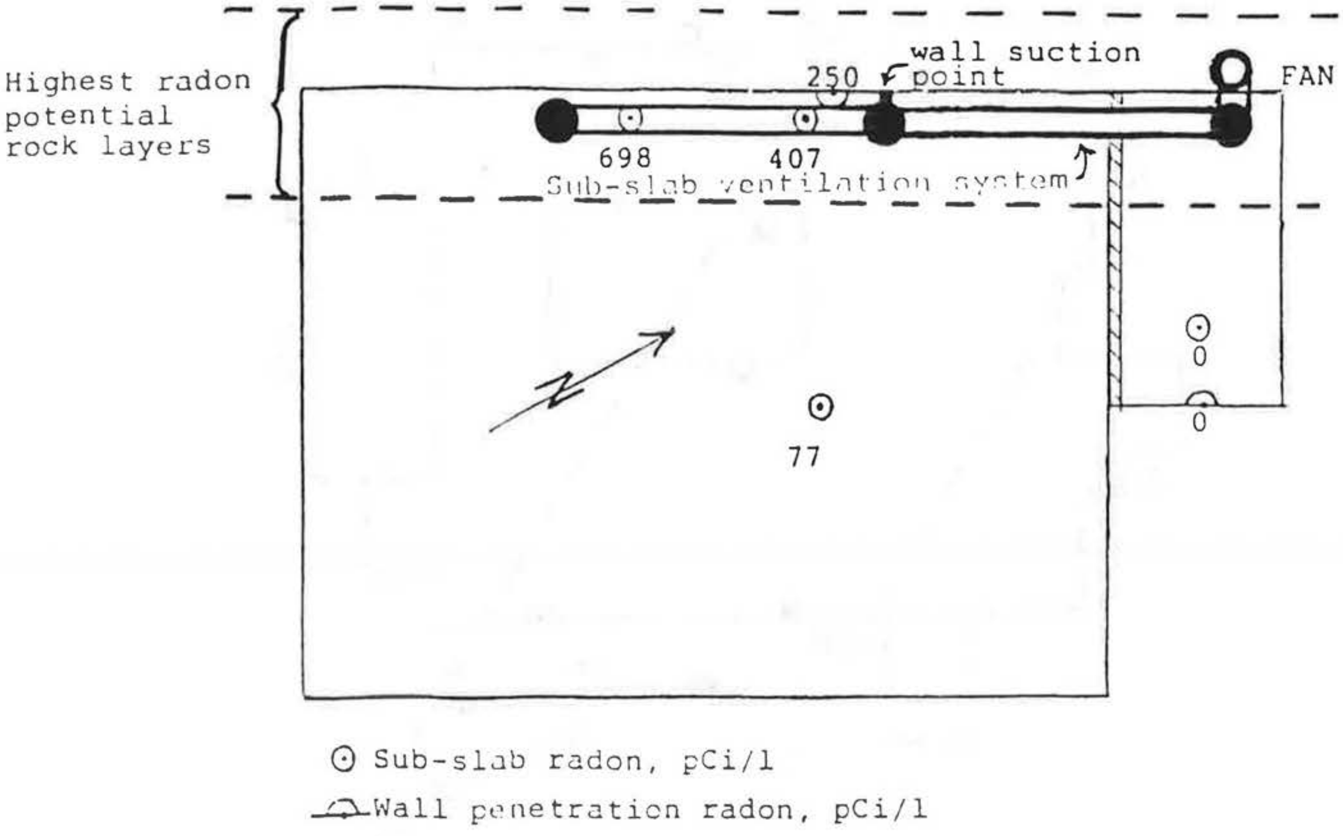

Figure 7. Footprint plan of a home showing numerical values of sub-slab and blockwall radon concentrations that indicate that the radon source is following N30°E rock layers, delineated by dash lines. Sub-slab ventilation systems penetrations are shown as darkened circles.

Figure 8 illustrates a similar house situation where high radon potentials are parallel to N30°E rock layers and generally increase toward the SE. Sub-slab ventilation as shown brought indoor radon levels from 30 pCi/l to 1.5 pCi/l.

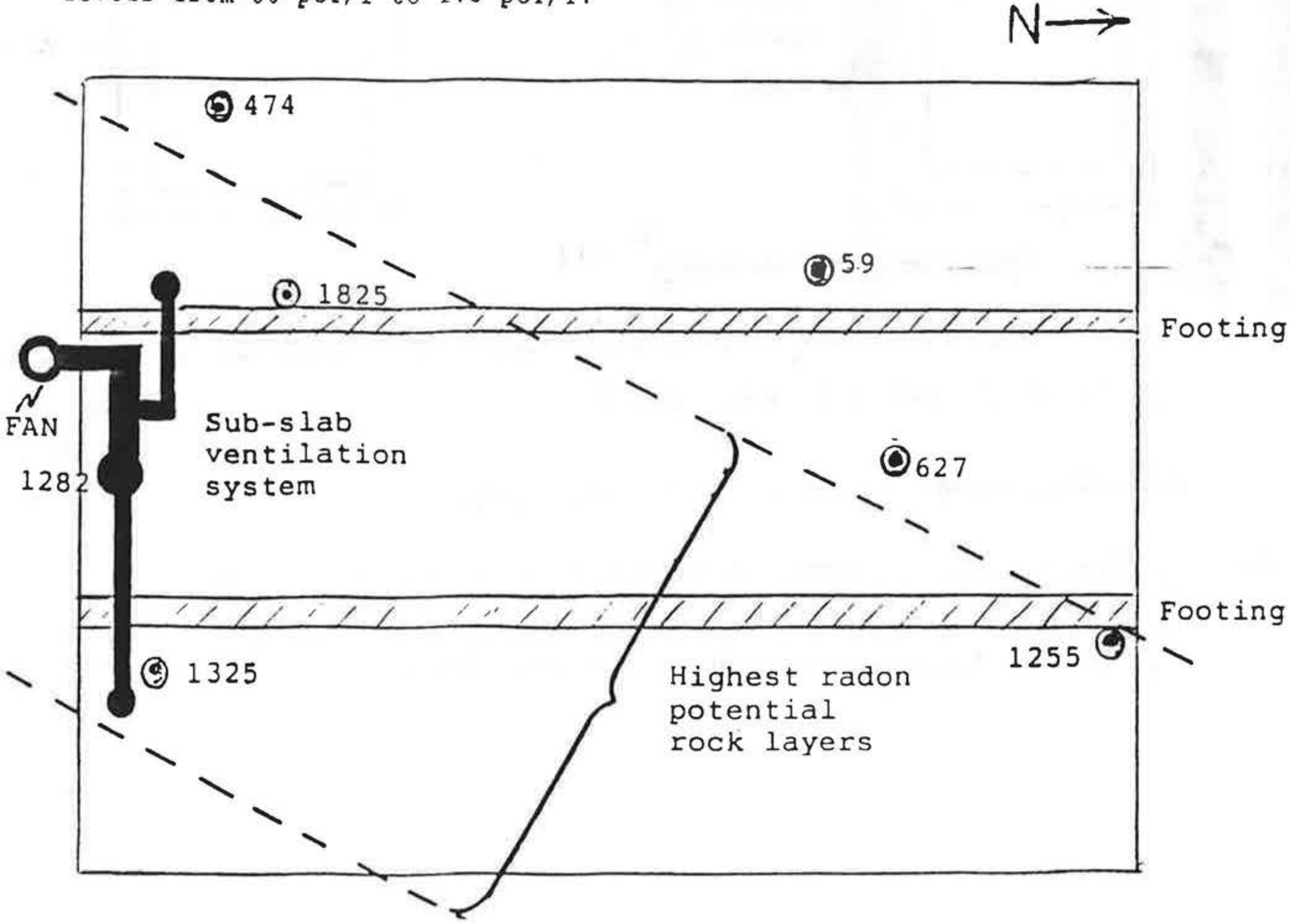

Figure 8. Footprint plan of a home showing numerical values of sub-slab and blockwall radon concentrations that indicate that the radon source is following N30°E rock layers, delineated by dash lines. Sub-slab ventilation systems penetrations are shown as darkened circles.

Figure 9 shows a workplace building with a footprint area less than 2000 ft² where micromanometer readings indicated no sub-slab communication because the slab was poured directly on compacted clay. Construction material radon levels tested negative. However, sub-slab radon levels increase towards the SE, congruent with local rock layers oriented N30°E. Thirteen slab penetrations with 2" pipe were necessary to deplete most of the source from the SE end of the building because the negative pressure field around each penetration was so small due to the very poor communication. Indoor radon, which initially was measured as high as 120 pCi/l, was reduced to less than 4 pCi/l.

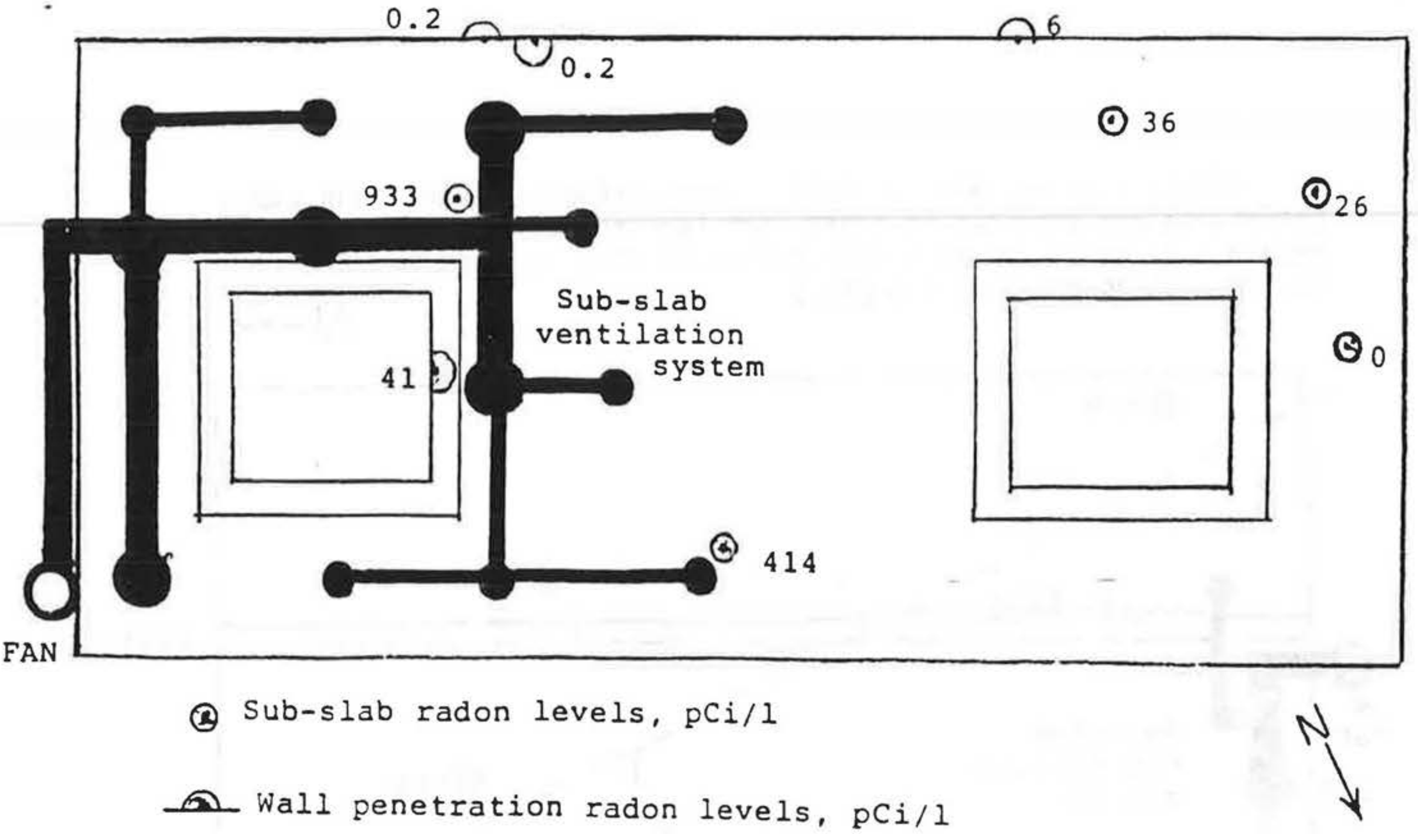

Figure 9. Footprint plan of a home showing numerical values of sub-slab and blockwall radon concentrations that indicate that the radon source is following N30°E rock layers. Sub-slab ventilation systems penetrations are shown as darkened circles.

Therefore, by knowing local geology and by tapping these high radon potential source areas with a multi-duct, single fan, sub-slab ventilation system optimum radon reductions can be achieved in buildings with poor sub-slab communication. Likewise by combining geologic knowledge with sub-slab and blockwall radon measurements in large buildings such as schools, the radon source can be located to determine where to place sub-slab ventilation systems that will achieve maximum radon reductions.

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TECHNOLOGICAL ENHANCEMENT OF RADON DAUGHTER EXPOSURES DUE TO NON-NUCLEAR ENERGY ACTIVITIES

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ABSTRACT

Natural radioactivity is a part of our natural surrounding and concentrations of natural radionuclides in the environment increase with the development of technologies. This is the case with phosphate ore processing in fertilizer industry and during coal combustion in coal-fired power plants. A major source of exposure to the population in the vicinity of non-nuclear industries results from inhalation of Rn-222 daughters. Exposure to radon daughters has been also associated with lung disorders that include cancer among workers. For that reason the radon daughter concentrations in different atmospheres are discused in this paper.

Working levels were measured as "grab samples" for several years at several stations on-site and off-site of the coal-fired power plant as well as the phosphate fertilizer plant, both located in Croatia. The average mean values of working levels are presented, and measurement techniques are reviewed.

INTRODUCTION

The exposure from man-made natural sources is called "technologicaly enhanced natural radiation" (TENR)(1). One of the first sourcesof uranium and thorium which was detected not being connected with the nuclear industry, was found during energy production using fossil fuels.

Uranium is widely distributed in nature and is a minor contaminant in all rocks, sand, and soil. Typical values for uranium are in the domain of 12 - 50 Bq/kg. Hence in ordinary back-yard soil there is of the order of 30 tons of uranium and 10 g of radium per square mile to a depth of 5 ft. Each cubic yard of ordinary soil or rock contains the order of 74 kBq of radium. This radium transforms at a constant rate into its daughter product, radon (^{222}Rn), and maintains a constant activity of about 74 kBq of radon per cumic yard of rock. Because all rock and soil is slightly porous some radon diffuses out of any exposed rock or soil surface. A typical value for the flow of radon from ordinary surface soils into the atmosphere is 3.7 µBq/sec.cm^2, or about 3.7 kBq/day per square yard (2). This radon is diluted in the atmosphere so that typical values for the radon concentration in outdoor air are in the domain of 3.7 - 37 Bq per cubic meter of air. Radon levels will build up near the surface under still, inversion conditions when mixing is minimal. The actual volume of radon in an uranium orebody is extremely small. 37 GBq of radon occupies only 0.66 mm^3 at normal conditions of pressure and temperature. Thus in the 1000 tonnes of ore, with 11 GBq of Ra-226, and therefore also 11 GBq of Rn-222, there is only about 0.2 mm^3 of radon.

EXPERIMENTAL PROCEDURES

MEASUREMENTS TECHNIQUES

The radon or radon daughter measurement techniques vary considerably from modified film badge type detectors (3) to highly elaborate alpha or beta counting equipment and even solid state alpha spectrometry (4). It is desirable, for the long-term monitoring of an atmosphere, that the measurement techniques be simple, accurate and require a minimum of equipment. The techniques in this paper allow direct evaluation of the working level value which is ultimately the quantity correlated with biological hazard.

The working level (WL) is defined as any combination of short-lived radon daughters in one liter of air that will result in the emission of 1.3 x 10^5 MeV of potential alpha energy. Under conditions of secular equilibrium 3.7 kBq/m^3 (100 pCi/l) of Rn-222 produces 1 WL (5). The definition is given in Table 1.

TABLE 1. DEFINITION OF THE "WORKING LEVEL" UNIT (WL)

Radionuclide	Alpha energy (MeV)	Half-life	Number of atoms per 100 pCi	Ultimate alpha energy per atom (MeV)	Total ultimate alpha energy (MeV/100 pCi)
Ra-222	5.49	3.8 d	1,770,000	excluded	-
Po-218	6.00	3.05 m	977	6.00 + 7.68	0.134×10^5
Pb-214	-	26.8 m	8,580	7.68	0.659×10^5
Bi-214	-	19.7 m	6,310	7.68	0.485×10^5
Po-214	7.69	0.0027 m	0.0008	7.68	0.000×10^5
					1.278×10^5
					or
					1.3×10^5

Measurements of radon daughters can be converted to working levels by an exact calculation if the state of daughter equilibrium is known. Several authors (6) have developed methods to determine the state of radon daughter equlibrium relative to Po-218, by alpha counting a filtered air sample. The most widely applied measurement technique in the uranium mines is that of Tsivoglou, than Kusnetz.

The Thomas-Tsivoglou method for calculation of radon daughter concentrations is inconvenient for field use. The irregular counting times require manual control of the scaler with consequent probabilities of error, and an error renders the complete data set useless. The 30-min counting period limits the processing rate to two samples an hour, so at least two scalers are required if rapid changes in daughter concentrations are to be measured. With the method developed by Scott (7) and our equipment it is possible to transfer a filter from air pump to portable scaler within 40 sec, and next 15 sec is ample time to note down the scaler reading and restart. Our procedure is therefore to take an air sample from 0 to 5 min, and then count the filter from 6 to 11 minutes (the M count), and from 11.25 to 16.25 min (the R count). These are the only fixed counting times. The third 5-min count (K count) is made on the filter at a time between 45 and 90 min. The rapid estimation of WL is:

$$WL = \frac{R}{5550 \times V \times E} \qquad (1)$$

where "R" is the total number of alpha counts, "V" is the sample flow rate (liters/min), and "E" is the counting efficiency. The value for the average daughter ratio is 5539 counts, which is rounded to 5550 for convenience.

The radon monitor consists of an alpha scintillator (ZnS/Ag), photomultiplier tube, a light-tight outer housing for the detector with passive air entry and an electronic package to convert the measured pulses to a digitally recorded signal, all battery operated for field use.

For estimating WLs, paralel with alpha counting we used for a long time a single beta-counting of air sampler filters, using the method developed by Holmgreen (8), based on the Eberline Air Particulate Monitor and total low-level beta counting sistem, battery operated for field use. Since the method is unjustly forgotten, here is a reminder of the basis for WL calculation.

The method for calculation of WLs from total beta activity concentrations is based upon Table 1, using two simplifying assumptions:

1. Since at equilibrium Pb-214 and Bi-214 account for 90% of the total ultimate alpha energy, a WL estimation based on Pb-214 and Bi-214 concentrations would approximate 90% of the actual value, so a factor "F" may be introduced to compensate for the exclusion of the Po-218 contribution as a result of counting only beta activity.

2. The radon daughter concentration ratios 1:0.65:0.35 (Po-218:Pb-214:Bi-214) are employed in the model.

The ultimate energy assigned to an atom of Po-218 is 13.68 MeV, the energy of its own alpha plus the alpha energy of Po-214, its great-granddaughter. Also, Pb-214 and Bi-214, although only beta emitters, are assigned the alpha energy of Po-214, as Po-214 will ultimately be produced from either of these atoms. The energy contribution of Po-214 present in the 1 litre volume is nearly zero, because of the small population of the extremely short-lived Po-214 atoms. Equation |2| defines the WL unit:

$$WL = \frac{(13.68\mathrm{MeV/atom}_A)(N_A) + (7.68\mathrm{MeV/atom}_{B+C})(N_B+N_C)}{1.3 \times 10^5\ \mathrm{MeV/WL}} \qquad (2)$$

where "N_A" is number of atoms of Po-218, "N_B" number of atoms of Pb-214, and "N_C" is number of atoms of Bi-214. The numbers of atoms of each daughter can be determined from Table 1. Substitution of numbers of atoms of each daughter into equation |2| yields:

$$WL = 0.001028(\mathrm{pCi}_A/\mathrm{l.}) + 0.005069(\mathrm{pCi}_B/\mathrm{l.}) + 0.003728(\mathrm{pCi}_C/\mathrm{l.}) \qquad (3)$$

Based upon two assumptions given above, equation |3| may be modified to become:

$$WL = F|0.005069(\mathrm{pCi}_B/\mathrm{l.}) + 0.003728(\mathrm{pCi}_C/\mathrm{l.})| \qquad (4)$$

Also: $\mathrm{pCi}_B/\mathrm{l.} = 0.65\ C_a$ (5)

and $\mathrm{pCi}_C/\mathrm{l.} = 0.35\ C_a$ (6)

where C_a is the total measured beta activity concentration (pCi/l.). Substitution into equation |4| of equations |4| and |5| and factoring, and taking into account that parameter "F" has an empirically determined value of 1.25, substitution into equation |4| gives:

$$WL = C_a(0.00575) \qquad (7)$$

In all our measurements we used glass fiber filters (General Electric), even we tried with molecular filters, but they were not convinient in very dusty atmosphere.

WL IN COAL-FIRED POWER PLANT

As the combustion of coal increases, so will the magnitude of environmental and human health hazards associated with trace elements and radionuclides mobilized by the coal fuel cycle. The large fraction of coal ash that does not find a commercial application is usually dumped in the vicinity of the coal-fired power plant (CFPP). When the dumping is finished, most dry ash dumps are covered by topsoil and converted into areas for agricultural or recreational use, but not yet in Yugoslavia (9).

The coal ash may contain enhanced levels of the natural radionuclides in the uranium and series, especially fly ash. Among the decay products are the radon isotopes, Rn-222, Rn-220 and Rn-219, which are noble gases and thereby pose special problems in assessing the radiological hazard of fly ash. The fractional amount of radon lost from the parent-containing material is called the emanation coefficient or emanating power. It is important to stress the difference between radon which escapes the physical confines of the parent-containing material (emanation) and that which occurs in a gas atmosphere which may be sampled (emanation + diffusion). Beck measured the emenation coefficients of coal ash obtained from three different power plants (10). For all samples he studied, the emenation coefficients were less than 0.1. Gamma radiation from a tailings dump is in general not a serious problem. Radiation levels 1 m from the pile surface tend to be less than 0.01 mGy/h and average around 0.005 mGy/h though "hot spots" with much higher dose rates have been reported (9). As with radon emenation, higher surface dose rates are to be expected over the tailings from higher grade coal, such as in the investigated case.

For all that reasons, investigations of the hazards were undertaken in the CFPP in Croatia, because the anthracite coal used for combustion has an average 10% sulphur and a variation of uranium. In the seventies the uranium content in coal was between 500 - 1200 Bq/kg. After 1980 it declined to an average 250 Bq/kg due to opening of an different vein in the coal mine. This requested a thorough monitoring programme which included measurements of activity concentration of radionuclides in coal and ash samples, and measurements of WL. First measurements of WL were carried out at 1977. In the CFPP seven locations have been chosen, because of long-time occupational exposure, and five on-site in places with natural air flow. Measurements have been repeated in 1983, when CFPP used coal with lower uranium content. In 1977 we used only Holmgreen`s method, and in 1983 we used both, Holmgreen`s and Scott`s method. Tables 2 and 3 summarize the estimated WL values, together with occupancy time limit.

TABLE 2. WL OF OCCUPATIONALLY EXPOSED PERSONS INSIDE THE CFPP

Work place	mWL * (1977)	Occupancy time limit	mWL (1983)	Occupancy time limit
1. Conveyour belt (coal)	8.0	42 h/week** unlimited	7.0	42 h/week unlimited
2. Conveyour belt (coal)	15.0	24-42 h/week	6.0	42 h/week unlimited
3. Below the automatic control (ash hooper)	80.0	21 h/week	12.0	24-42 h/week
4. Below the automatic control (ash hooper)	60.0	42 h/week	12.0	24-42 h/week
5. Waste pile fresh	80.0	21 h/week	-	-
6. Waste pile old	-	-	60.0	42 h/week
7. Bottom ash	80.0	21 h/week	20.0	24-42 h/week

TABLE 3. WL ON-SITE IN PLACES WITH NATURAL AIR FLOW

Work place	mWL* (1977)	Occupancy time limit	mWL (1983)	Occupancy time limit
1. Area around the steam generator building	6.0	unlimited	6.0	unlimited
2. Under the stack	5.0	unlimited	6.0	unlimited
3. Near the furnice	5.0	unlimited	6.0	unlimited
4. Office building (500 m from the CFPP)	3.0	unlimited	-	-
5. 10 km from the CFPP	3.0	unlimited	6.0	unlimited

*mWL = 1 x 10^{-3} WL. All WL values are an arithmetic mean of 3 measurements.

**42 h/week was taken as the occupancy time limit to comply with the US general population standards, since the workers in the CFPP were never considered as people occupationally exposed to radiation.

The WLs have shown great variations between two measurements depending on the radioactivity of the coal and combustion products present at the time of the measurements in the CFPP. Places on-site with good ventilation had 3 - 6 mWL. The highest WL was besides the bottom ash and fresh waste pile where even an occupancy time limit should be considered. The values for the WL are changing, so that the new data are lower than these presented in Table 2 and 3. Table 4 summarizes the estimated WL values measured in 1990, when we used only Scott`s method.

TABLE 4. WL MEASURED ON-SITE AND OFF-SITE CFPP IN 1990.

Location	mWL
1. Coal storehouse	6.0
2. Below the automatic control (ash hooper)	11.0
3. Area around the steam generator building	6.0
4. Slag and ash pile	6.0
5. Štrmac	6.0
6. Vozilići	5.0
7. Stepčići	5.0
8. Luka Plomin	4.0
9. Rabac	3.0

There were no differences in WLs between measurements done by one or the other method. As we expected, the highest values were obtained on-site of the CFPP. Locations 5 - 9 were at different directions and distances from the CFPP, chosen in dependency on the wind-rose (Table 5).

TABLE 5. ALTITUDES, DISTANCES AND DIRECTIONS FROM THE CFPP

Location	Altitude (m)	Distance (km)	Direction
Štrmac	120	3	SW
Vozilići	100	5	NW
Stepčići	80	2	W
Luka Plomin	10	1	SE
Rabac	0	20	S

The most interesting case is the location Štrmac, where a hamlet was built on a ninety years old tailing site, where already the second and even the third generation of same families are dweling in the same houses.

At the location Rabac, which is at the sea shore the WL is slightly lower, since the radon levels over the sea and the ocean are much lower than over the land, due to the lower Ra-226 content of the sea. For this reason, radon levels in the atmosphere at coastal sites are very dependent on whether the wind is blowing from the land or the sea.

WL IN FERTILIZER INDUSTRY

Three years after the beguining of the WL measurements at the CFPP, the same type of investigations has started in a fertilizer plant.

The activity mass concentrations of natural radionuclides in phosphate ore for a given radionuclide and type of fertilizer vary markedly from one country to another, depending on the origin of the components. General features are that the activity mass concentrations of K-40 and Th-232 and its decay products are always low and that the activity mass concentrations of the radionuclides of the U-238 decay series are 5 - 50 times higher than in normal soil. The degree of radioactive equilibrium between U-238 and its decay products in a given type of fertilizer depends essentially on the relative contribution of phosphoric acid, since phosphoric acid usually has a very low Ra-226 concentration. For the purpose of this, it is assumed that Th-230 and U-234 are in radioactive equilibrium with U-238 and that Pb-210 and Po-210 are in radioactive equilibrium with Ra-226.

A typical concentrations of U-238 and Ra-226 in sedimentary phosphate deposits are 1500 Bq/kg, which are generally found to be in radioactive equilibrium. When these rocks are processed into fertilizer most of the uranium and some of the radium accompanies the fertilizer, and than in the fields through crops enters the food chain.

In the production of fertilizers, phosphate rocks are used in two different ways. The first method, the acidulation of phosphate rocks was ensured by sulphuric acid, where phosphoric acid and gypsum result as normal superphosphate. The second method, where the phosphate rock is treated by nitric acid, the final product is phosphoric acid and gypsum as residue, which contains most of the radium (11).

Almost all of Ra-226 originally in the phosphate ore is discharged in the piles. The concentration of Ra-226 in piles is about 700 Bq/kg. Since the rate of radon production equals the rate of radium decay, the rate of radon production can be readily calculated. The answer is, 1 g of Ra-226 (this is also 1 Ci or 37 GBq of Ra-226) produces 74 kBq/sec of Rn-222. Thus the radon production rate in piles containing 700 Bq/kg od Ra-226 is 1.4 mBq/kg/sec. The density of dry piles is about 0.7 g/cm^3, which means that the production rate of radon per unit volume is about 1 $mBq/m^3/sec$.

The highest occupational radiation exposure during the process are to be expected in the fertilizer production or in the fertilizer storehouse. To check the level of radiation dose, a monitoring programme was introduced, including the determination of specific activities of natural radionuclides in ambient air, phosphate ore, phosphate fertilizers, waste products, trickling and well waters. Measurements of WLs were carried out at ten locations, twice a year for the last ten years. Five of them were inside the phosphate fertilizer plant, one on the gypsum`s pile. The off-site locations were at four different directions and distances, chosen on the basis of the wind-rose. Results are presented in Table 6.

TABLE 6. WL MEASURED ON-SITE AND OFF-SITE THE FERTILIZER PLANT

Location	mWL
1. Phosphate ore storehouse	12.0
2. KCl storehouse	4.6
3. Fertilizer package store (NPK)	21.0
4. Inside the fertilizer production	9.4
5. Phosphoric acid production	4.4
6. Gypsum`s pile	3.0
7. Off-site locations	1.2

All values are an arithmetic mean of ten years measurements performed in summer and winter, always three times on each location. During the first year only beta measurements (Holmgreen) were done, and later only alpha measurements (Scott)(7,8). There were no significant differences observed during the years. For the comparison in Table 7 one year data are presented measured once by alpha and once by beta measurement.

TABLE 7. WL MESURED BY DIFFERENT METHODS

Location	Holmgreen	Scott
	mWL	
1. Phosphate ore storehouse	3.1	3.2
2. KCl storehouse	2.5	1.1
3. Fertilizer package store (NPK)	3.5	4.2
4. Off-site locations	1.4	1.2

The WL rate differs slightly not because of different measuring methods, but also due to different phosphate ore origin.

CONCLUSION

This paper introduces WL measurements in industries where TENR is present. The CFPP is a specific case with the appearance of natural radioactivity which was very similar to open pit uranium mining, where WL measurements are routinelly done. For that reaso WL measurements were applied also in this case. When some places of occupational exposure in the CFPP were detected, the authors have tried to find out if the same problem also exists in the fertilizer industry. The appearence of places with an increase of natural radioactivity in nonnuclear industries have left the legislator, at present without a ready solution in Yugoslavia, how to systematize occupationally exposed workers, especially after the Chernobyl accident, when the public become sensitive to radiation of any origin.

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