1.00

# RADON AND THORON DAUGHTER MEASUREMENTS USING A PORTABLE RADON SNIFFER

EFENDI ZAINUDDIN and PHILIP JENNINGS
School of Mathematical and Physical Sciences
Murdoch University, Murdoch, WA, 6150

## ABSTRACT

A flexible version of the two-count method has been developed for use in determining radon and thoron daughter working levels in residential and industrial premises. The method is adapted for use with a simple radon sniffer based on a low speed pump and an alpha counter. The flexibility of the method derives from the sofware, where some freedom is permitted in the selection of sampling times and post sampling analysis of the filters. This method has been tested on a variety of radioactive materials and it gives consistent, reliable results over a wide range of working levels. It provides an inexpensive method of surveying radon and thoron levels in a variety of domestic and industrial premises. The results of a survey of Rn(222) and Rn(220) daughter levels in dwellings within the Perth Metropolitan Area are reported. This study shows that the mean concentration of indoor Rn(222) daughters was around 5.2 mWL (20.83 Bq $m^{-3}$ EEC) with a range from 0.8 mWL(3.0 Bq $m^{-3}$ EEC) up to 23.3 mWL (86.4 Bq $m^{-3}$ EEC). The mean thoron daughter concentration was 8.6 mWL (2.36 Bq $m^{-3}$ EEC) with a range from 1mWL( 0.27 Bq $m^{-3}$ EEC) to 64.5 mWL (17.74 Bq $m^{-3}$ EEC). Using conversion factors of 0.061 mSv/Bq $m^{-3}$ for Rn(222) daughters and 0.29 mSv/Bq $m^{-3}$ for Rn(220) daughters respectively (UNSCEAR,1982), it is estimated the average annual effective dose equivalent is 1.2 mSv for Rn (222) and 0.6 mSv for Rn(220) respectively.

## INTRODUCTION

Rn(222) and Rn(220) and their progeny are well known as health hazards for causing lung cancer in humans. This is based on the epidemiological evidence of a higher probability of mortality from lung cancer caused by Rn(222) gas among uranium miners (Archer et al, 1973; Kunt et al,1979; Howe,1988; Harley,1988; Fabrikant, 1990; Steinhausler, 1989) as well as non- uranium miners (Lundin, 1969; Radford et al, 1984). These gases are common contaminants of the air in mines. Significantly elevated levels of Rn(222) have also been found in dwellings in America, Canada and Europe (Edling et al, 1979; Eaton,1987; James, 1987). Rn(222) occurs naturally as a result of the decay of uranium in the rocks and the decay products of the gas are solid radioactive elements which occur in the atmosphere in the form of attached and non-attached fractions. Since the gas is continously inhaled by humans, both of these fractions will decay in the lung and will be deposited in the bronchial tree where they irradiate the nearby tissues with alpha, beta and gamma radiation. A serious risk occurs from irradiation by alpha particles emitted by these solid isotopes which may cause the induction of lung cancer. The risk of lung cancer from Rn(222) and its progeny is typically thousands of times larger than that from other enviromental pollutants. In fact, millions of Americans are exposed to more radiation in their homes than underground miners experience in the workplace (Nero, 1986).

Internationally accepted occupational levels are specified in radon daughter concentrations and the unit commonly used is the Working Level (WL). This unit can be converted to an equivalent equilibrium Rn(222) concentration (EEC) which corresponds to 3700 Bq $m^{-3}$ EEC for 1 WL of Rn(222) daughters and 275 Bq $m^{-3}$ EEC for 1 WL of Rn(220) daughters. In the U.S.A, the Environmental Protection Agency (EPA) has recommended a public exposure limit of an average background level of 20 mWL (74 Bq $m^{-3}$ EEC) and action levels of 10 mWL ( 37Bq $m^{-3}$ EEC). Based on 168 hour per week exposure Departement of Mines, WA (1982) has set limits for members of the public for Rn(222) and Rn(220) daughters of 10 mWL (37 Bq $m^{-3}$ EEC) and 100 mWL (27.5 Bq $m^{-3}$ EEC) respectively. In the latest recomendation was set by the Radiation Health Committee (1990), a remedial action level was set 54 mWL (200 Bq $m^{-3}$ EEC) for existing homes. The committee advised that remedial action is not necessary where the annual average radon concentration is below 200 Bq $m^{-3}$ EEC (Mason,1990).

## METHODS

To measure indoor Rn(222) and Rn(220) daughter working levels, a flexible version of the two count method (Stranden, 1980) has been developed. In this method, radon daughter working levels may be computed as,

$$WL\,(Rn) = (1.044\ N_1 + 5.150\ N_2 + 3.805\ N_3) \times 10^{-3}$$

$$N_1 = \frac{\lambda_c}{\lambda_a} N_3, \qquad N_2 = \frac{\lambda_c}{\lambda_b} N_3$$

$$N_3 = \frac{I(1) - (D_d N_4 - D_e N_5 - D_f N_6)}{\left(D_a \frac{\lambda_c}{\lambda_a} + D_b \frac{\lambda_c}{\lambda_b} + D_c\right)} \qquad (1)$$

And thoron daughter working levels may be computed as;

$$WL\,(Tn) = (0.122\,N_5 + 0.0116\,N_6)$$

$$N_5 = \frac{\lambda_f}{\lambda_e}\,N_6,$$

$$N_6 = \frac{I(2)}{\left(D_D\,\frac{\lambda_f}{\lambda_e} + D_E\,\frac{\lambda_f}{\lambda_e} + D_F\right)} \qquad (2)$$

Where,

$\lambda_{a,\,..,\,f}$ = are the decay constants of RaA, RaB, RaC, ThA,ThB, and ThC respectively.

I(1) = Total observed alpha activity on the filter paper over counting period(1) from $T_0$ at the end of sampling to $T_1$.

I(3) = Total observed alpha activity on the filter paper over counting period (3) from $T_2$ to $T_3$.

$D_{a,b,c}$ = are integrated alpha counts for RaA, RaB, and RaC respectively over counting period (1) for a radon concentration of 100 pCi/litre at a sampling rate of 1 litre/min.

$D_{d,e,f}$ = are integrated alpha counts for ThA,ThB, and ThC respectively over counting period (1) for a thoron concentrations of 7.5 pCi/litre at a sampling rate of 1 litre/min.

$D_{D,E,F}$ = are integrated alpha counts for ThA,ThB, and ThC respectively over counting period (2) for a thoron concentrations of 7.5 pCi/litre at a sampling rate of 1 litre/min.

$N_1$ = pCi RaA/litre, $N_2$ = pCi RaB/litre, $N_3$ = pCi RaC/litre,
$N_4$ = pCi ThA/litre, $N_5$ = pCi ThB/litre, $N_6$ = pCi ThC/litre.

The optimal time for counting period (1) is 30 min ($T_0 = 0$, $T_1 = 30$) and for counting period (2) is 300 min to 600 min ($T_2 = 300$, $T_3 = 600$).

The method was used to determine relative concentrations of RaA, RaB, RaC,ThA,ThB,andThC in the atmosphere by measuring the alpha activity of a mixture of these isotopes on a filter paper through which air is sucked for a period of 1 or 2 hours. By observing the integrated alpha counts for a period of 0 - 30 minutes and 300 - 600 minutes after sampling, the Rn(222) and Rn(220) daughter working levels were calculated. The delayed observation of integrated alpha counts (300 minutes after sampling) is intended to eliminate Rn(222) daughters from the determination of Rn(220) daughter working levels, since the relatively short-lived Rn(222) daughters will have completely decayed during that period. Before applying this method to analyse field data, the Radon Sniffer was calibrated and tested using standard samples prepared in the laboratory. A computer program has been written to analyse integrated observed alpha counts after sampling, based on the mathematical analysis in this study. The method was found to give consistently reliable results over a wide range of working levels.

## MEASUREMENT SITES

The Perth Metropolitan Area contains several different types of soils. Basically, it is divided into two main structures, the coastal plain and the steeply rising escarpment of the Darling Range. In order to simplify the analysis of the results of indoor radon monitoring, the geological structures will be grouped into four zones as shown in Figure 1. Zone 1 is entirely covered lime beach sand and limestone which are a major constituent of the coastal plain. Zone 2 is a part of the coastal plain with subsoils consisting of quartz and leached sand. Zone 3 is a part of the coastal plain with clay and loam overlying granitic and lateritic subsoils. Zone 4 is on the scarp and is mainly composed of precambrian rock, granite and laterite and other igneous rocks.

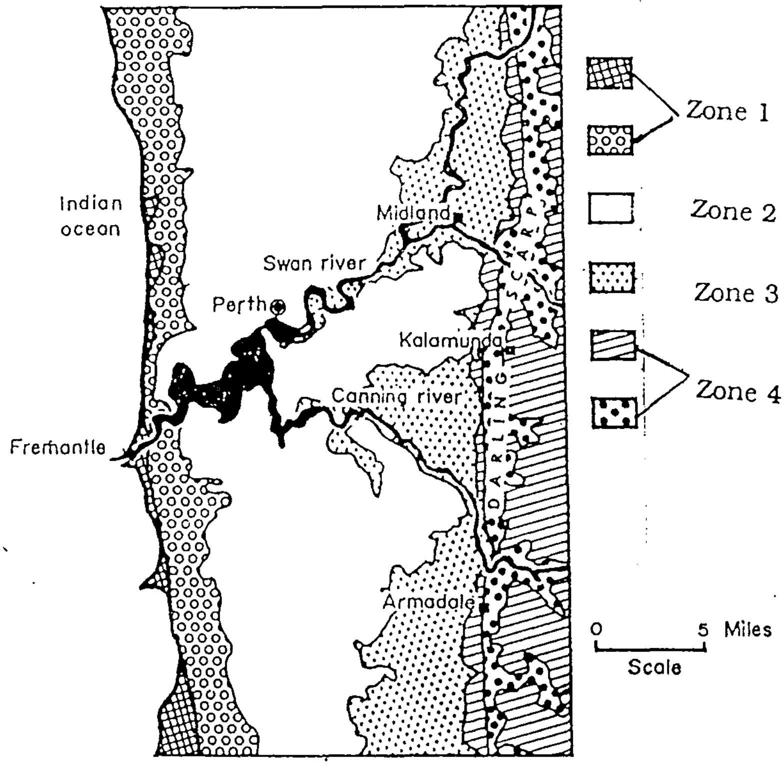

Figure 1. Geologigal structure of the Perth Metropolitan Area

## THE INSTRUMENT

The device used to conduct the measurements is a radon sniffer detector made by Thomson & Nielsen Electronics Ltd. It contains a battery and an air pump plus a filter holder, detector,and liquid crystal display. The heart of the instrument is the detector system which is based on silicon dosemeter chips. The instrument operates by sampling air from the environment at a constant rate (1 litre/min). When both power and pump are switched on, air is sampled through a standard 0.8 μm millipore filter. Radon and thoron daughter products are collected on the filter paper and alpha particles emitted are counted by the detector and display as "ALPHA COUNTS". This is the total integrated alpha count over the whole counting period (not the alpha activity).

The instrument is calibrated with a 10 nCi custom-mounted, Am-241 alpha emitter supplied by Thomson & Nielsen Electronics Ltd. This calibration source was checked by the laboratory of the Radiation Health Branch in Western Australia to confirm the activity of the source. The known activity reported was used to deremine the calibration factor and efficiency of the instrument, and then applied to compute radon and thoron working levels.

## RESULTS AND DISCUSSION

### 1. Indoor Rn(222) Daughter Concentrations

Table 1. shows the mean concentration of indoor Rn(222) daughters for each zone and all zones in the Perth Metropolitan Area. It appears that all values are much lower than the limit set by the Radiation Health Committee (Mason, 1990). The mean concentration was around 5.2 mWL (20.83 $Bq^{-3}$ EEC) and a geometric mean of 4 mWL (14.9 Bq $m^{-3}$ EEC) with a range from 0.6 mWL (2.22 Bq $m^{-3}$ EEC) up to 23.3 mWL (86.4 Bq $m^{-3}$ EEC). The geometric mean was in good agreement with the national survey conducted by Solomon et. al, (1987). However, the mean concentration was found to be higher than the total mean value for all States Australia (12 Bq $m^{-3}$) obtained by the Australian Radiation Laboratory (Mason, 1990). The average values found in Perth are much lower than the action level set by the USEPA which is 10 mWL (37 Bq $m^{-3}$ of EEC). Only 10 % of total measurements obtained exceeded this value. Most of these concentrations were found in zone 4 where the subsoils are composed of granite and laterite. Roleystone was the main area measured which is situated in this zone.

Table (1)
Rn (222) DAUGHTER WORKING LEVELS, MEASURED IN THE PERTH METROPOLITAN AREA

| Zone | N | Arithmetic mean(mWL) | Geometric mean(mWL) | Minimum (mWL) | Maximum (mWL) |
|---|---|---|---|---|---|
| Zone 1 | 18 | 2.9 | 2.5 | 0.6 | 6.3 |
| Zone 2 | 25 | 4.3 | 3.4 | 1.1 | 17.9 |
| Zone 3 | 23 | 6.4 | 4.3 | 0.6 | 19.0 |
| Zone 4 | 38 | 7.0 | 5.0 | 1.2 | 23.3 |
| Whole | 104 | 5.2 | 4.0 | 0.6 | 23.3 |

## 2. Indoor Rn(220) Daughter Concentrations

Based on 168 hours per week exposure, the limits for the public for Rn(220) daughters is 0.1 WL (27.5 Bq $m^{-3}$ of EEC) (Departement of Mines,WA, 1982). From 104 total houses measured, the thoron daughter concentrations were found to be below this public limit. Table 2. shows the mean concentration of Rn(220) daughters for each zone and all zones. It covered a range from 1 mWL to 64.5 mWL ( 0.27 to 17.73 Bq $m^{-3}$ EEC) and the mean concentration was 8.6 mWL (2.36 Bq $m^{-3}$ EEC) with a geometric mean of 5.5 mWL (1.48 Bq $m^{-3}$ EEC). The highest arithmetic mean and geometric mean over the four zones was in zone 3. Zone 3 has soils composed of quartz sands mixed with clays and loams washed down from the Hills. It is conceivable that thoron gas can percolate more readily through such soils than it can through the heavier soils in the Hills. There is a significant correlation between indoor thoron daughter concentrations and the nature of the geological structure, particularly in zones 3 and 4.

So far, there are no published results of the national survey of thoron daughter concentrations conducted in Australia. Therefore no comparison of the results can be made. However, it is worthwhile to compare the results with those of thoron daughter concentration measurements from dwellings in other countries. Measurements of thoron at Madras City, India carried out by Lakshmi et. al, (1989) show that in 68.38 % of 136 total houses measured, thoron concentration had the values between 0 to 1 WL (0 to 275 Bq $m^{-3}$ EEC) and the values were found to be in the range of 0 to 7.5 WL ( 0 to 2062.5 Bq $m^{-3}$ ). In the United Kingdom, thoron daughter concentrations were found to range from 0.43 to 4.32 mWL (0.12 to 1.2 Bq $m^{-3}$ EEC), (UNSCEAR, 1982). A survey carried out in the Federal Republic of Germany indicated that thoron daughter concentrations were found in the range from 0.4 to 7.9 mWL (0.11 to 2.2 Bq $m^{-3}$ EEC) and the mean value was 1.3 mWL (0.37 Bq $m^{-3}$ EEC) (UNSCEAR, 1982). It seems that thoron daughter concentrations in the Perth Metropolitan Area are much lower than these values obtained in Madras City , India. While 83 % of the measurements gave thoron daughter concentrations below 10 mWL (2.7 Bq $m^{-3}$ EEC), these values are higher than the values found both in United Kingdom and the Federal Republic of Germany.

Table (2)
Rn (220) DAUGHTER WORKING LEVELS, MEASURED IN THE PERTH METROPOLITAN AREA

| Zone | N | Arithmetic mean(mWL) | Geometric mean(mWL) | Minimum (mWL) | Maximum (mWL) |
|---|---|---|---|---|---|
| Zone 1 | 18 | 2.6 | 2.2 | 1.0 | 7.0 |
| Zone 2 | 25 | 7.8 | 5.1 | 1.4 | 44.9 |
| Zone 3 | 23 | 13.4 | 7.7 | 1.4 | 54.0 |
| Zone 4 | 38 | 10.5 | 6.9 | 1.2 | 64.5 |
| Whole | 104 | 8.6 | 5.5 | 1.0 | 64.5 |

### 3. Rn(222) Daughter Doses

The annual effective dose equivalent was estimated using a conversion factor of 0.061 mSv $Bq^{-1}$ $m^3$ (UNSCEAR,1982) for Rn(222) daughters. Table (3) shows a summary of the mean calculated effective dose equivalent for radon daughters for each zone and all zones using the appropriate indoor radon daughter level from Table (1). The mean value of the annual effective dose equivalent for all zones is 1.10 mSv which is associated with exposures from Rn(222) daughter concentrations averaging 20.83 Bq $m^{-3}$ (EEC). This dose rate is considerably lower than the total effective dose equivalent contributed by cosmic rays and terrestrial radiation received by the United States population which is estimated to be 3 mSv $y^{-1}$ and approximately 2 mSv $y^{-1}$ is estimated to come from inhalation of Rn(222) daughters (NCRP, 1987). The annual effective dose equivalent from inhalation of U(238) decay series, which are mainly due to Rn(222) decay products, was estimated to be 0.77 mSv (ICRP,1990) and 1.24 mSv (UNSCEAR,1988). It appears that the annual effective dose equivalent from inhalation of Rn(222) daughters in this study is within the range of values estimated by ICRP (1983) and UNSCEAR (1988).

Figure (2) shows the frequency distribution of the annual effective dose equivalent for Rn(222) daughters for all zones. It can be seen that that about 56 % of the total calculated annual effective dose equivalents are below 1 mSv and 27 % are between 1 and 2 mSv, while 17 % are estimated to be between 2 and 6 mSv. Most of the relatively high annual effective dose equivalents were found in zones 3 and 4 which are associated with granite and laterite formations.

Table (3)
Mean and Extreme Values of Effective Dose Equivalents for Indoor Rn(222) Daughters Calculated for all zones, in the Perth Metropolitan Area

| | Annual effective dose equivalent (mSv $y^{-1}$) | | |
|---|---|---|---|
| Zone | Minimum | Mean | Maximum |
| Zone 1 | 0.13 | 0.65 | 1.42 |
| Zone 2 | 0.25 | 0.97 | 4.05 |
| Zone 3 | 0.13 | 1.44 | 4.29 |
| Zone 4 | 0.25 | 1.57 | 5.27 |
| Whole | 0.13 | 1.10 | 5.27 |

## 4. Rn(220) Daughter Doses

The conversion factor used to estimate the annual effective dose equivalent per unit inhaled potential alpha energy concentration of Rn(220) daughters is 0.29 mSv Bq $m^{-3}$ (UNSCEAR, 1982) for indoor Rn(220) daughter concentration. The contribution of Rn(220) gas itself to the effective dose equivalent is relatively small and may be neglected. The conversion factor is based on a similar occupancy factor and breathing rate used for evaluating the annual effective dose equivalent for Rn(222) daughters.

The mean value of annual effective dose equivalent for Rn(220) daughter concentrations for each zone and all zones is presented in Table (4). The average value for all zones is 0.66 mSv. This dose rate is associated with an average indoor Rn(220) daughter concentration of 2.36 Bq $m^{-3}$ (EEC). If we compare this annual effective dose equivalent value with the estimated annual effective dose equivalent from the inhalation of Th(232) decay series which are mainly due to Rn(220) daughters given by UNSCEAR, (1988), and ICRP, (1983), it appears that an average value given in this study is more than 3 times higher than that given by UNSCEAR, (1988) and ICRP, (1983). This may be explained because most high values of dose rate come from zones 3 and 4 where the geological formations are associated with granite and laterite. In addition, more samples were taken from zone 4 than any other zone. Therefore the mean annual effective dose equivalent for all zones could be biased.

The frequency distribution of annual effective dose equivalent for all zones is presented in Figure (3). It appears that 81 % of total annual effective dose equivalents fall below 1 mSv and 15 % have the values in the range 1 to 2.5 mSv, whereas 4 % are estimated to be between 3.5 and 5 mSv.

## CONCLUSION

There is a significant correlation between Rn(222) and Rn(220) daughter concentrations in zones 3 and 4 and the type of soils and geological structure in these zones. This was indicated by elevated indoor Rn(222) and Rn(220) daughter levels and significant annual effective dose equivalents found in these zones. The occurrence of higher Rn(222) and Rn(220) daughter concentrations in zones 3, and 4 may be explained on the basis that these zones have soils composed of granite and laterite which have a relatively high uranium and thorium content. Since the Rn(222) and Rn(220) are the decay products of these decay series, the presence of elevated indoor Rn(222) and Rn(220) concentrations could be related to the significant amount of uranium and thorium in the soils and rocks in these areas.

Table (4)

Mean and Extreme Values of Effective Dose Equivalents for Indoor Rn(220) Daughters Calculated for all zones, in the Perth Metropolitan Area

| | Annual effective dose equivalent (mSv $y^{-1}$) | | |
|---|---|---|---|
| Zone | Minimum | Mean | Maximum |
| Zone 1 | 0.01 | 0.20 | 0.56 |
| Zone 2 | 0.11 | 0.62 | 3.59 |
| Zone 3 | 0.11 | 1.10 | 4.30 |
| Zone 4 | 0.10 | 0.84 | 5.14 |
| Whole | 0.01 | 0.63 | 5.14 |

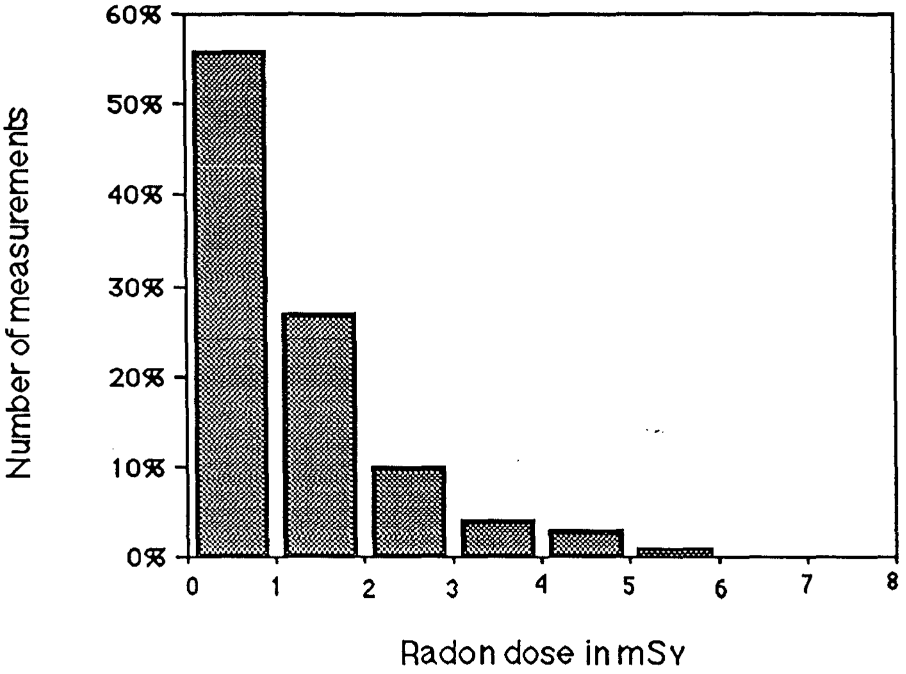

Figure 2. Frequency distribution of annual effective dose equivalent for indoor radon daughters for all zones, expressed in units of mSv.

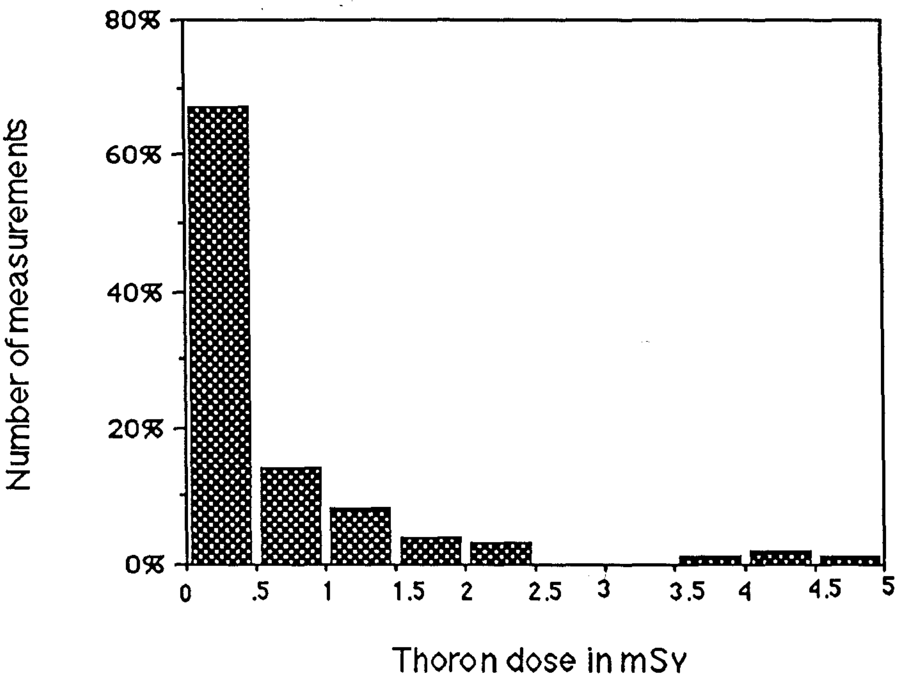

Figure 3. Frequency distribution of annual effective dose equivalent for indoor thoron daughters in all zones, expressed in units of mSv.

## REFERENCES

Archer, V.E., Wagoner, J.F. and Lundin, F.E. " *Lung Cancer Among Uranium Miners in the United States* ", Health Phys. **25** , pp.351 - 71(1973).

Departement of Mines, W A, " *Code Practice in the Mining of Mineral Sands Depaertement of Mines* ", (1982).

Edling, C., Atelson, O., and Kling, H. " *Lung Cancer and Residency - A case referent It's Daughters in Dwellings* ", Scand . J. Work Environ, Health **5**, pp. 10 - 15 (1979).

Eaton, R., " *Radon Issues in the U.S.A. and Canada* ", Canadian Radiation Protection Association (CRPA) Conference, Saskaton, May (1987).

Fabrikant, J. I., " *Radon and Lung Cancer : The BEIR IV Report* ", Health Phys, **59** (1), pp. 89 - 97 (1990).

Harley, N.H., " *Updating Radon Daughter Risk Projections* ", in : Radon and the Environment, (ed : Makofske, W.J, and Edelstein, M.R), Noyes Publication, New Jersew. USA., pp. 170 - 78 (1988).

Howe, G. R., L'Abbe, K.A. " *The Association Between Radon Daughter Exposure and Lung Cancer: Evidence and Implications from Studies of Miners* ", in: Radon and the Environment, (ed : Makofske, W.J, and Edelstein, M.R), Noyes Publication, New Jersey, USA. pp 161 - 169, (1988)

ICRP, " *1990 Recommendations of the International Commision on Radiological Protection* ", ICRP Publication **60**, Pergamon Press (1990).

ICRP, " *Principles For Limiting Exposure of the Public to Natural Sources of Radiation* "", ICRP Publication **39**, Annals of the ICRP, 14(1),Pergamon Press (1983).

James, A.C, " *Radon in the Building* ", (CPRA) conference Saskaton, May (1987).

Kunz, E., Placek, V. and Horacek, J., " *Lung Cancer in Man in Relation to Different Time Distribution of Radiation Exposure* ", Health Phys, **36**, pp.699 - 706 (1979).

Laksmi, K.S and Meenakshi V. S. " *Radon, Thoron Measurements in Dwellings at Madras City, India* ", in : Radioprotection, Environmental Radioactivity and Earth Sciences, ( ed: Tommasino, L., et al, World Scientific Pub, Singapore, (1990)).

Lundin, F.E, Loyd, J.W, Smith, E.M., Archer, V.E. and Holaday, D.A. " *Mortality of Uranium Miners in Relation to Radiation Exposure, Hard Rock Mining and Cigarette Smoking - 1950 through 1967* ", Health Phys. **16**, pp. 571 - 78 (1969).

Mason, C., " Radon in Homes ", ARL( Australian Radiation Laboratory), (1990).

NCRP, " *Exposure to the Population in the United States and Canada from Natural Background Radiation* ", NCRP Report 94, National Council on Radiation Protection and Measurement, Betheseda, Maryland (1987).

Nero, A.V. " *The Indoor radon Story* ", The Technology Review, January (1986).

Radford, E.P. and Renard, K.G., " *Lung Cancer in Swedish Iron Miners Exposed to Low Doses of Radon Daughters* ", New England Journal of Medicine, **310** ,pp. 1495 - 99 (1984).

Solomon, S.B., Peggie, J.R., Pojer, P.M., Wise, K.N., " *Preliminary Measurements of Radon in Australian Homes Using a Charchoal- Based Monitor* ", presented at 4th Int. Symp. on Natural Radiation Environment, Lisbon, December(1987).

Steinhausler, F., " *Indoor Radon : Health Effects and Future Research Trends* ", Proceedings of the International Workshop on Radon Monitoring, April 3 - 14 (1989) Trieste, Italy, in : Radioprotection, Environmental Radioactivity and Earth Sciences, ( ed: Tommasino, L. et al, World Scientific Pub, Singapore, pp. 32 - 50(1990)).

Stranden, E., " *A Two Count Filter Method For Measurement of Rn(222) and Rn(220) Daughters in Air* ", Health Phys. **38**, pp. 73 - 76 (1980).

UNSCEAR, " *Ionizing Radiation : Sources and Biologigal Effects* ", United Nations Scientific Commitee on the Effects of Atomic Radiation 1982 Report to General Assembly ", United Nations, New York (1982)

UNSCEAR, " *Sources, Effects and Risks of Ionizing Radiation* ", United Nations Scientific Commitee on the Effects of Atomic Radiation 1988 Report to General Assembly ", United Nations, New York (1988).