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Mathematical Modeling of Radon Concentration Measurements in Air by Charcoal Canisters Without Diffusion Barriers using Finite Difference Technique

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ABSTRACT

This research studies the behavior of the activated charcoal canister when exposed to different radon concentrations using finite difference technique to solve diffusion equation that simulates diffusion of radon in charcoal. Two cases of radon concentrations were studied, namely: Constant radon concentration and stepwise increase for predefined time duration in radon concentration. The present study suggests that the active adsorption time is 95% of the saturation time which equals 48 h in case of constant radon concentration using the specified charcoal canister in this work. It is also recognized that the amortization activity in case of a step increase in radon concentration is 5% more than the canister's activity in the case of constant radon concentration. It is found that for different values of step heights (in case of step wise increase in concentration) the timing of the charcoal activity peak does not change. This means that the peaking time depends only on the duration of the step in radon concentration not on the height of the step. The study indicated that the canister's activity is positively affected by increasing the step height. Finally, the calibration factor CF of the charcoal canister is found to be independent of step height. Nevertheless; it depends on the starting and ending times of the step increase in the radon concentration.

Key words: Radon measurements, activated charcoal canisters, mathematical modeling, simulation

INTRODUCTION

The background radiation dose due to radon gas forms more or less about 50% of the nominal background radiation dose all over the world (UNSCEAR, 2000). The measurements of Radon concentration in air is of vital importance especially in closed areas.

There are many measuring techniques of Radon concentration in air. These techniques vary in their accuracy and integration period. The techniques can be classified into two main categories: Short term grab sampling and long term integration methods (Wichmann *et al*., 2005). The most famous technique of the long term integration techniques is the solid state nuclear track detectors and the charcoal canisters (Chen, 2003). The former technique has better integration characteristics while the later technique is easier to manipulate and cheaper.

The performance of a charcoal canister in adsorbing radon was studied and modeled by many researchers (Cohen and Cohen, 1983; Cohen and Nason, 1986; Blue and Holcomb, 1989; Nagarajan *et al*., 1990; Blue and Jarzemba, 1992). They studied the diffusion of radon into

the charcoal canister and concluded that radon is desorbed out of the canister as well as adsorbed into it. Nikezic and Urosevic (1998) simulated the diffusion of radon in a bed of activated charcoal canister with diffusion equations. Two exact analytical solutions for the differential equation to get the charcoal calibration constant (CF) for the case of exposure to constant radon concentration in the atmosphere were introduced by Nikezic and Urosevic (1998). The first solution was for short time exposure while the second solution was for long time exposure. The detailed mathematical procedures and equations used to derive the two cases were explained in details in their study. The activity of activated charcoal adsorption determined by the finite elements method and experimental verification were given as well. They concluded that the principal advantage of such a formulation is easiness and simple receipt of canister responses for different types of activation. It provides an appropriate method for testing the effects of different parameters of monitoring and predicting responses to different variations of these parameters. According to their simulation results they suggested that the model adequately describes the phenomenon of radon diffusion and adsorption on activated charcoal, when taking into account the process of radioactive disintegration. This means that the model could be used as a new approach to the problem of determining radon concentration if the total radon activity in the canister is known. They analyzed both the situation when radon level is constant and the more realistic one when radon level is varying as a stepwise function with step duration time. They concluded that the calculation of the environmental radon level by the activated charcoal adsorption for the appropriate measurement conditions is possible by using their model. They also concluded that optimization could be applied when determining the optimal exposure time, the minimum thickness of the canister needed for the given exposure period and the number of canisters needed for attaining the measurements accuracy.

In order to examine the possibilities for improving the integration characteristics of active charcoal, simulation was done by using finite element method (Urosevic *et al*., 1999). Their study investigated the possibility of improving the integration characteristics of active charcoal by studying and employing simulation techniques. This simulation studied two cases of environmental radon concentrations. The first case is constant radon concentration, while the second case is stepwise increase in radon concentration. In constant radon concentration simulation gave a space time distribution of radon adsorption by the canister and an optimization of the active charcoal thickness. The space time distribution is determined during an active adsorption period and saturation period. The active adsorption period was defined as the time needed for achieving 85% of the equilibrium radon amount in the active charcoal canister. This equilibrium is achieved when the number of radon gas atoms that diffused into the active charcoal (adsorbed by the charcoal) is approximately equal to that disintegrated and escaped (desorbed) from the canister. The saturation period is the time taken to reach constant radon activity in the charcoal bed. They studied in their simulation the canister response to constant and variable radon concentrations. In case of exposure to constant radon concentration (C_0) they concluded that the time taken to achieve saturation period depends on the thickness of the active charcoal canister and the optimal time of the exposure should not be longer than the duration of the active adsorption period. While in the case of stepwise increase in radon concentration ($C = 5\ C_0$), they found that the time of the peak amortization is 24 h for active adsorption while the saturation period is 36 h according to their study (Urosevic *et al*., 1999).

The amortization time is defined as the time required for the canister activity to decrease back to its value corresponding to constant radon concentration and to recover from the effect of the stepwise rise in concentration. Radon adsorption by activated charcoal canisters were also studied

by Underhill (2003). He used the basic components of a diffusive sampler which are a windshield at the face of sampler; an air gap (or some other diffusion barrier) which serves as a valve to limit the radon input into the sampler; the adsorption for the radon and the radon impervious backing. Such barrier prevents the loss of radon through this section of the sampler. He used modified Fick's diffusion equation to include both the adsorption and the decay of radon. He applied two boundary conditions. The first is found by setting the flow of radon across the air gap equal to the flux of radon into the adsorbent. The second boundary condition states that there is no loss of radon through the impervious backing at the rear of the sampler. He used analytical solution using convolution integral for constant radon concentration. He used finite difference calculations otherwise. He concluded that radon is affected by desorption more than other air contaminants; and decayed after being sampled while others don't. Radon samplers have a much greater width than the diffusive samplers commonly used in industrial hygiene according to their conclusions (Underhill, 2003). Diffusive samplers for radon must give reliable results not only at normal air velocities but also under nearly stagnant conditions. Diffusive samples gave helpful data for epidemiologists to obtain a dose-effect relationship for human exposure to radon in order to set standards for exposure limits to low levels of ionizing radiation (including radon).

The aim of the present research is to deeply study the behavior of the activated charcoal canister when it is exposed to different values of stepwise rise in radon concentrations.

MATERIALS AND METHODS

Following the same model introduced by Urosevic *et al.* (1999), the diffusion equation is going to be used to represent adsorption of radon in charcoal. The model simulates all processes involved in the exposure of active charcoal to atmospheric radon. The effect of other gases in air is taken into account in the diffusion and adsorption constants. The radon diffuses into charcoal from the atmosphere or vise versa depending on the surrounding concentration of radon in the atmosphere and the concentration in the charcoal. The model takes into account the radioactive disintegration of radon adsorbed in the charcoal. The analysis given here is applied to open canisters without diffusion barriers. The governing differential equation is given by:

$$\frac{\partial}{\partial x_i}\left(Di\frac{\partial C}{\partial x_i}\right)-\lambda C=\frac{\partial C}{\partial t}$$

Where:

C = Radon concentration in the pores of active charcoal
D = Diffusion coefficient in coordinate (i)
λ = Decay constant of radon
t = Time
x_i = Coordinate system i = 1, 2, 3

Assuming isotropic diffusion coefficient and a single dimension for variation of radon concentration, the equation reduces to:

$$D\frac{\partial^2 C}{\partial Z^2}-\lambda C=\frac{\partial C}{\partial t}$$

The boundary conditions of the equation are:

$$\text{at, } z = 0, C(0, t) = k\,\rho\,C(t)$$

$$\text{at } z = L, \frac{\partial C}{\partial z} = 0 \text{ (non permeable boundary condition)}$$

Where:

ρ = Charcoal density (kg m^{-3})
k = Adsorption coefficient in charcoal (m^3 kg^{-1})

The initial conditions are taken to be zero every where for the concentration of radon in charcoal at the start up of exposure (C (z, 0) = 0). The control volume finite difference approach is employed to solve the equation in small control volumes. The control volumes at the boundaries are taken to be smaller than the control volumes inside the charcoal. The control volumes are hollow cylinders with finite height and with rectangular cross sections.

Figure 1 shows the finite difference cross section of the grid which is used to represent the canister. A software program was developed to perform such calculations. In order to establish confidence in the results of the software, the case of constant concentration of radon in the atmosphere had been solved and compared to the analytical solution given in the reference (Nikezic and Urosevic, 1998). The solution uses the data from reference (Nikezic and Urosevic, 1998). The data list is:

D = 1.27×10^{-9} (m^2 sec^{-1})
k = 3.9 (m^3 kg^{-1})

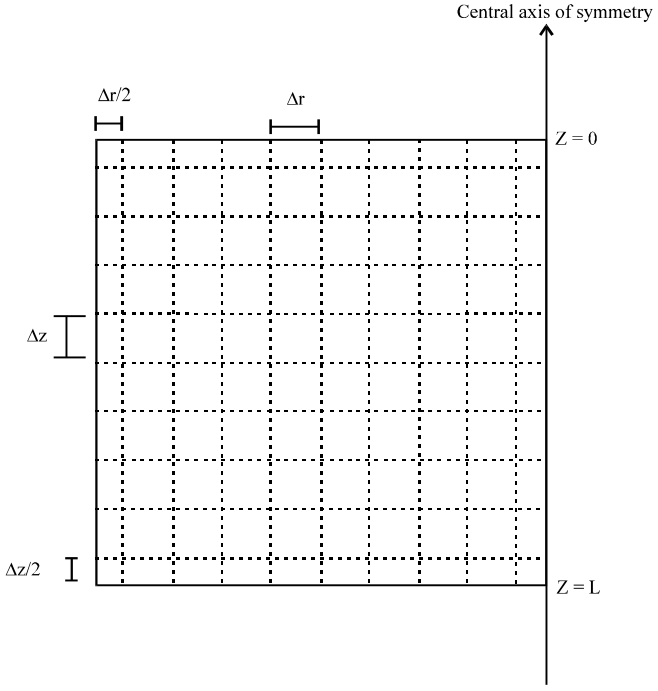

Fig. 1: Finite difference cross section of the grid in calculations

$\rho = 500\ (kg\ m^{-3})$
$L = 0.017\ m$

where, L is the canister thickness.

$$S = 0.0081\ m^2$$

where, S is the canister surface area.

$$C_0 = 1\ (Bq\ m^{-3})$$

where, C_0 is the normalized value of radon concentration in air.

$$\lambda = 2.1\times10^{-6}\ sec^{-1}$$

The results of the simulation are given in Fig. 3. The results show a very good agreement with the analytical solution. The developed software allows the use of the explicit time scheme, implicit time scheme and Crank Nicholson time scheme. Crank Nicholson time scheme (Crank and Nicolson, 1996) is used for solution to avoid the limitations on the stability of explicit scheme. The resulting system of finite difference linear algebraic equations is solved using Gauss-Seidel iteration technique (Barrett *et al*., 1994) to a relative accuracy of 10^{-7}. A grid of 20×20 is used to represent the canister. Using this method the response of the canister with activated charcoal to different outside constant and time step rise mentioned before. The output of the model includes the variation of concentrations in the canister with time and space. The total activity of adsorbed radon in the charcoal is calculated using numerical integration and different parameters had been studied.

RESULTS AND DISCUSSION

The first study case is the canister described by Urosevic *et al*. (1999). This canister is open at one end and there were no radon atoms inside the charcoal before the exposure. The other sides of the canister are closed so that radon leakage in lateral directions is neglected. Following these assumptions, the diffusion problem becomes a two dimensional one (z and r) with equal diffusion coefficients in different directions. Physical characteristics of the canister and activated charcoal used for radon measurements according to the EPA standard and EPA protocol which were used by Nikezic and Urosevic (1998), are mentioned in the above data of simulation. Radon diffusion and adsorption are simulated during 144 h with a step of 1 h. The output of the model is a space time distribution of radon in active charcoal. Using numerical integration over the volume of charcoal, the activity of charcoal is calculated at each time step.

Figure 2 shows a Schematic diagram of radon concentration in air with time predefined by Urosevic *et al*. (1999) and used for this study.

Figure 3 gives the activity change of charcoal with time for constant concentration of radon in air. For the sake of comparison the concentration of radon in air is normalized to unity. Since the effect of radon concentration is linear in the model equation and the boundary conditions, the results are going to be multiplied by a constant value of radon concentration. The end of active

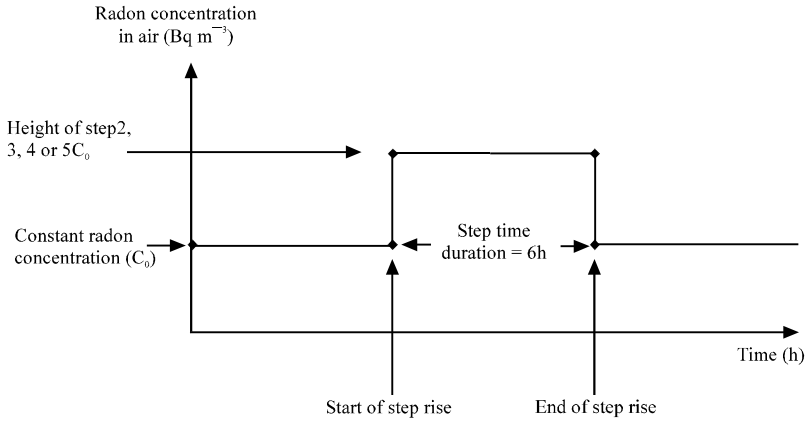

Fig. 2: Schematic diagram of radon concentration in air with time

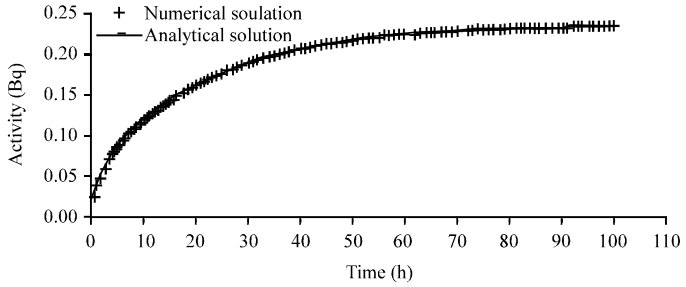

Fig. 3: Comparison of numerical and analytical solutions for charcoal canister's activity at constant radon concentration

adsorption which is defined as 85% of the equilibrium of radon amount in the active charcoal is found to be at 33.3 h. This value has been determined using linear interpolation to get the exact 85% value of the solution of charcoal.

Figure 3 shows a very good agreement between the present numerical solution and the analytical solution given by Nikezic and Urosevic (1998). This agreement confirms the accuracy of the numerical solution. Figure 3 also shows that the total saturation for radon adsorption into the canister occurred at exposure time of about 68 h. At this time the canister's activity exceeded by 5.4% its value of activity at exposure time of 48 h. This concludes that exposure for 48 h could be quite enough. Finally, Fig. 3 suggests a value of 95% of the saturation time as a good representative for the active adsorption time. This value is higher by 10% than the value suggested by Urosevic *et al.* (1999). For the sake of comparison the three numerical schemes explicit, implicit and Crank Nicholson are used for the calculations.

Figure 4 shows that the three schemes are almost identical provided that the explicit scheme satisfies the condition of stability. Thus, Fig. 4 suggests the use of Crank Nicholson scheme to avoid numeric instability of the solutions and massive calculations and output results since Crank Nicholson scheme is unconditionally stable.

In order to deeply study the behavior of the charcoal canister, we tried to simulate a real radon concentration environment. Firstly, it is assumed that radon concentration is constant (i.e., $C = C_0$). Secondly, we assumed that radon concentration is varying as a step function that appears in an

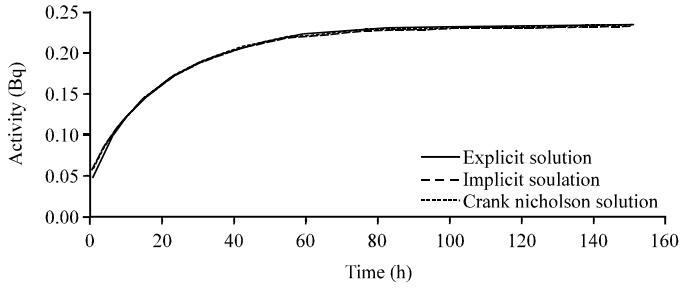

Fig. 4: Comparison between different numerical time schemes in the solution

interval of 6 h duration. The concentration of radon during these step functions are C = 2, 3, 4 and 5 C_0 and then go back to the original concentration before the 6 h increase as shown in Fig. 2. These step functions covered respectively 72 h of exposure time.

As an example of the results, Fig. 5a and b show the charcoal canister activity for four different values of step heights and two different values of starting and ending times of the step function keeping a duration of 6 h constant. It was found that the peaking time depends only on the duration of the step in radon concentration. It does not depend on the step height. The amortization time depends on the value of the radon concentration during the step height. However, it does not depend on the start and end times of such step. Varying the order of the interval of 6 h along the 72 h period of simulation does not affect the amortization time. The calibration factor in m^3 (CF) of the canister is defined as the ratio of radon activity adsorbed by active charcoal and average radon concentration in the air during the period of exposure.

The amortization times for different concentrations were found to be as follows:

- For a step height of 2 C_0 the amortization time was 36 h
- For a step height of 3 C_0 the amortization time was 48 h
- For a step height of 4 C_0 the amortization time was 54 h
- For a step height of 5 C_0 the amortization time was 60 h

The values suggest that canister's amortization is 5% more than the activity at constant radon concentration.

Figure 6 gives the calibration factor CF for different radon concentration step wise heights; namely; 2, 3, 4 and 5 C_0. The x-axis represents the ending time of the step rise. Since, the step rise duration is 6 h, then the ending time is going to be multiplies of 6 h. The model values are relatively lower than those given by Urosevic *et al.* (1999).

The difference may be attributed to the finer mesh in finite difference grid and shorter time steps used in finite difference solution. Hence we used 20×20 grid of the canister instead of 10×20 used by Urosevic *et al.* (1999). In addition we applied one hour time step while their graph indicate that they applied six h. The figure shows that the values of CF do not depend on the height of the step function. Nevertheless, CF depends on the starting and ending time of step rise in radon concentration. The values are slightly different from those determined by Urosevic *et al.* (1999).

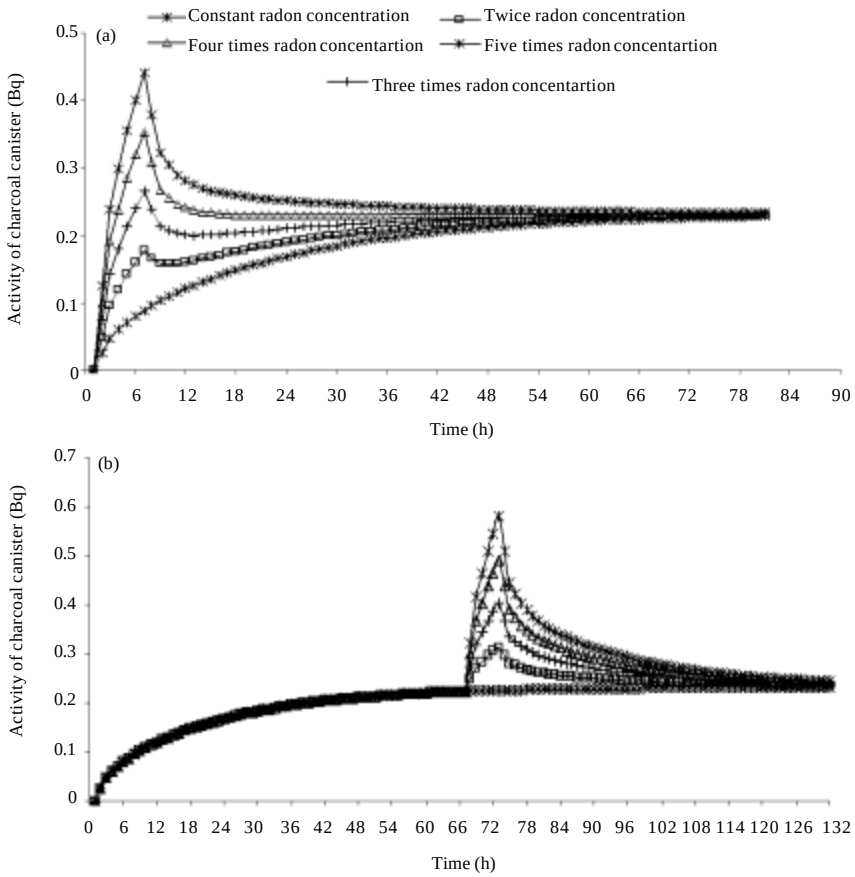

Fig. 5(a-b): Constant versus variable radon concentrations in (a) The first 6 h and (b) Twelfth 6 h time step

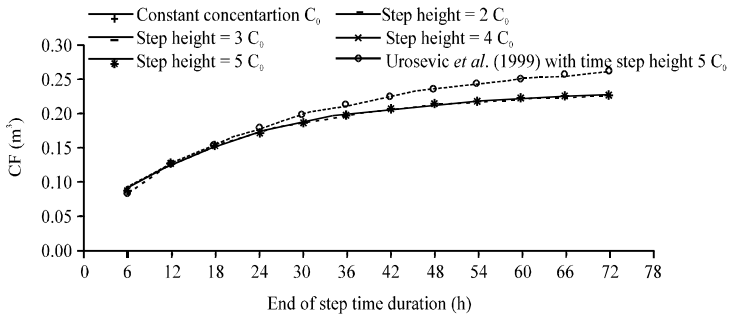

Fig. 6: Calibration factor (CF) versus time for different step heights in radon concentration with 6 h time duration

CONCLUSION

There is a reasonable agreement between the analytical solution given by Nikezic and Urosevic, (1998) and present numerical solution. This confirms the accuracy of numerical solution. Crank Nicholson Scheme is preferred to be used in numerical simulations to avoid numerical instability

in the solutions. The present study suggests that the active adsorption time is 95% of the saturation time which equals 48 h for the given dimensions of the charcoal canister. Using mesh of (20×20 grid) and applying only one hour time step gives more accurate results than coarser mesh size. The present study suggested that amortization is 5% more than the canister's activity at constant radon concentration. It is found that for different values of step heights the time of the charcoal activity peak dose not change. The peaking time depends only on the period of the step in radon concentration. The amortization time depends only on the value of radon concentration. The canister's activity is positively affected with increase in the step height. Finally, it is found that CF depends on the start and end time of step rise in radon concentration.

REFERENCES

Barrett, R., M. Berry, T.F. Chan, J. Demmel and J. Dongarra *et al.*, 1994. Templates for the Solution of Linear Systems: Building Blocks for Iterative Methods. 2nd Edn., SIAM Press, Philadelphia, ISBN: 9780898713282, Pages: 112.

Blue, T.E. and X.X. Holcomb, 1989. A global kinetic model for the adsorption of radon gas in humid air on charcoal for US EPA canister. Radiat. Prot. Dosimetry, 27: 173-178.

Blue, T.E. and M.S. Jarzemba, 1992. A model for radon gas adsorption on charcoal for open-faced canisters in an active environment. Health Phys., 63: 226-232.

Chen, J., 2003. Estimate of annual average radon concentration in the normal living area from short-term tests. Health Phys., 85: 740-744.

Cohen, B.L. and E.S. Cohen, 1983. Theory and practice of radon monitoring with charcoal adsorption. Health Phys., 45: 501-508.

Cohen, B.L. and R. Nason, 1986. A diffusion barrier charcoal adsorption collector for measuring Rn concentrations in indoor air. Health Phys., 50: 457-463.

Crank, J. and P. Nicolson, 1996. A practical method for numerical evaluations of the heat conduction type-conduction type. Adv. Comput. Math., 6: 207-226.

Nagarajan, R., L.D. Michaels, V.B. Menon, D.T. Wasan and D.S. Ensor, 1990. A mathematical model for Rn sampling by activated C adsorption. Health Phys., 58: 575-581.

Nikezic, D. and V. Urosevic, 1998. A theoretical study of radon measurement with activated charcoal. Nucl. Instrum. Methods Phys. Res. Sect. A, 406: 486-498.

UNSCEAR, 2000. Exposures from natural radiation sources (Annex B). Report to the General Assembly, United Nations, New York.

Underhill, D.W., 2003. Diffusive sampling of radon. Health Phys., 84: 287-295.

Urosevic, V., D. Nikezic, S. Vulovic and M. Kojic, 1999. Optimization of radon measurements with active charcoal. Health Phys., 76: 687-691.

Wichmann, H.E., A.S. Rosario, I.M. Heid, M. Kreuzer, J. Heinrich and L. Kreienbrock, 2005. Increased lung cancer risk due to residential radon exposure in a pooled and extended analysis of studies in Germany. Health Phys., 88: 71-79.