THE IMPLICATION OF THE TIME-DEPENDENT DIFFUSION THEORY
ON RADON-222 EXHALATION MEASUREMENTS

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By collecting the radon (Rn-222) emanating from a sample enclosed in an accumulation can, the mean exhalation rate (Bq s^{-1}) can easily be deduced by extracting gas from the can and analysing it for radon. This so-called closed-can method has been used in various forms since the beginning of this century. The radon exhalation rate from a sample free in air and in equilibrium with its surroundings (i.e. the radon concentration gradient inside the porous sample is constant in time) is referred to as the free exhalation rate of the sample. The natural aim of most exhalation measurement techniques is to determine this free exhalation rate. Unfortunately, the radon accumulating variant of the closed-can technique fails in this (except for samples that are very thick compared with the diffusion length), and what is really remarkable is that this failure can pass unnoticed, even under carefully controlled laboratory conditions. The objective of this paper is to illustrate, by means of theoretical calculations using diffusion theory and Fick's law, why it is so difficult to experimentally determine the free exhalation rate with closed-can methods. The temporal variation of the radon exhalation rate and the corresponding radon gas concentration in the sample enclosure (outer volume) for 'one-dimensional' samples will be presented. The conclusions drawn are, in principle, also valid for more realistic sample geometries, as long as the dimensions of the sample relative to the diffusion length are kept the same.
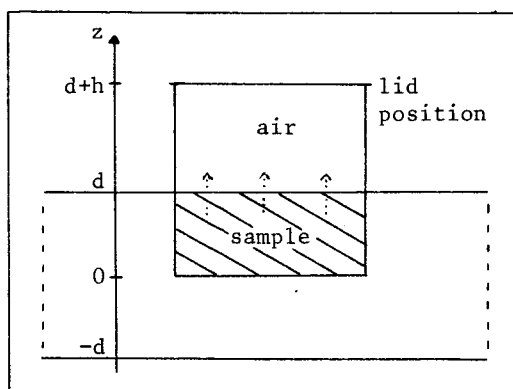

Fig. 1. A sample of thickness d placed in the bottom of an can is closed at time t=0, the enclosure exhibits the same areal exhalation as a semiinfinite slab of thickness 2d. The outer volume height with lid in place is denoted h.

In order to facilitate theoretical calculations, the simple sample geometry displayed in Fig. 1 is assumed. Furthermore, it is presupposed that before closing the sample is freely exhaling with an outer volume radon concentration equal to zero. In the absence of pressure and temperature gradients across the sample, the transport of radon from the sample pores into the outer volume is purely diffusive, i.e. is governed solely by a radon concentration gradient. If the can is closed at time t=0, the differential equation governing the pore concentration, C(z,t), of radon at time t and depth d-z in the sample is given by equation 1 (Sa84).

Eq. 1 $p \, \partial C(z,t)/\partial t = p\lambda\{C_m - C(z,t)\} + D_e \, \partial^2 C(z,t)/\partial z^2$

where D_e is the effective diffusion coefficient, p the sample porosity, C_m the maximum pore concentration in an infinite sample, and λ the decay constant of radon-222. C_m is related to the specific radium concentration, C_{Ra}, of the sample by the relation $pC_m = C_{Ra}\varepsilon\rho$, where ε is the emanation fraction (the fraction of the radon produced in the sample reaching the pore space), and ρ the bulk density of the sample. The initial condition is that $C(z,t)$ is zero for all $z>d$ (i.e. in the outer volume). The boundary conditions for the bottom and the open surface of the sample for $t>0$ are given by Eqs (2) and (3), respectively (instantaneous mixing of the air in the outer volume is assumed)

Eq. 2 $\partial C(z,t)/\partial z = 0$ for $z=0$ (bottom)

Eq. 3 $-pD_e \, \partial C(z,t)/\partial z - h(\lambda+\nu)C(z,t) = h \, \partial C(z,t)/\partial t$ for $z=d$

where ν is the leakage rate constant of the can. Equation 3 is the mathematical expression for the balance between the exhalation according to Fick's law (first term) and the loss of activity (second term) due to decay and leakage. The relative leakage factor, μ, is equal to $(\lambda+\nu)/\lambda$ and the diffusion length, L, is defined as $L=(D_e/p\lambda)^{1/2}$. Analytical solutions to Equation 1 for $\mu=1$ (i.e. no gas leakage out of the can) have been published (Kr71, Sa84).
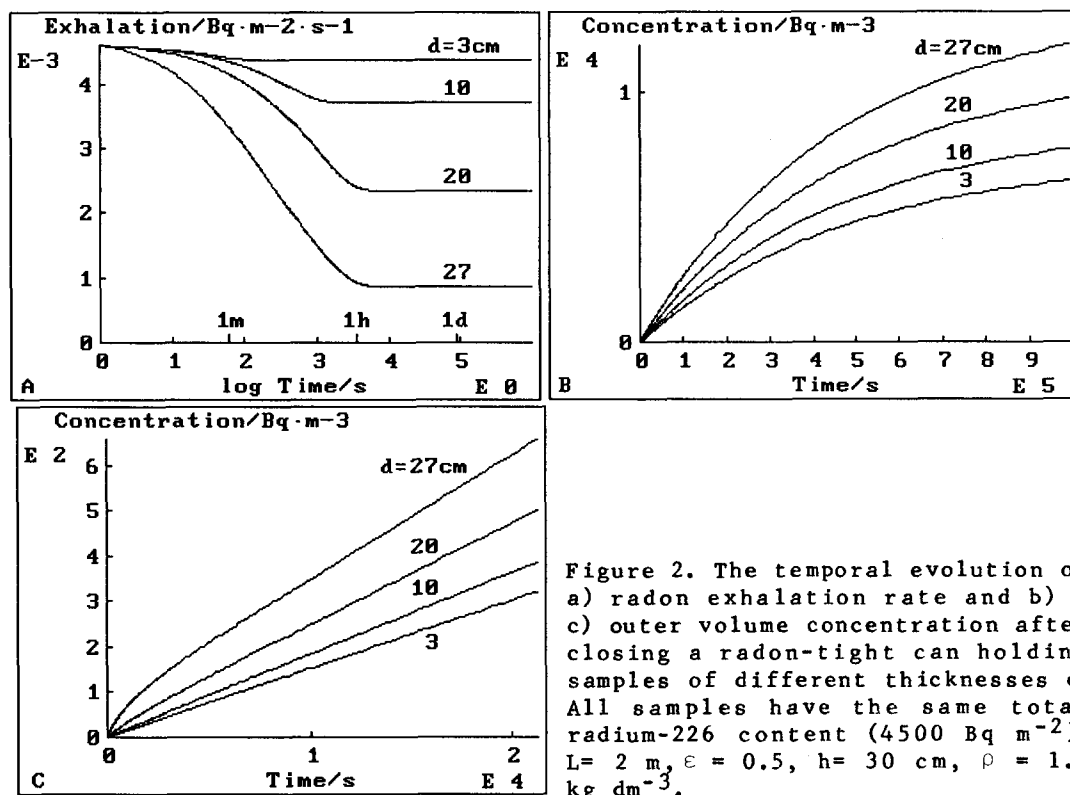
Figure 2. The temporal evolution of a) radon exhalation rate and b) & c) outer volume concentration after closing a radon-tight can holding samples of different thicknesses d. All samples have the same total radium-226 content (4500 Bq m^{-2}). L= 2 m, ε = 0.5, h= 30 cm, ρ = 1.5 kg dm^{-3}.

In this paper, numerical solutions for $C(d,t)$ (i.e. the outer volume concentration as a function of time) and the corresponding exhalation rate (Bq m^{-2} s^{-1}) are presented in Figures 2-4. The radium-226 activity of all samples, irrespective of sample thickness, is normalized to 4500 Bq m^{-2}. The time scale of the exhalation part of the figures is

logarithmic in order to emphasize the initially rapid change in exhalation rate after closure. The concentration versus time curves are drawn on linear scales. All samples in figures 2-3 are thin compared with the diffusion length, L= 2m. Nearly all radon atoms available to the pore volume of the samples will therefore escape to the outer volume initially. In other words, all samples will exhibit about the same free exhalation rate. Let us, with this in mind, examine the radon-tight ($\mu=1$) enclosure illustrated in figure 2. As soon as the lid is put on the increase in radon activity in the outer volume will lead to an increase in the radon concentration in the outermost pores, thereby decreasing the exhalation rate. Within a few hours a final exhalation rate is reached. Thereafter, the increase in radon concentration is uniform throughout the whole pore volume and outer volume, a state corresponding to an exhalation rate which is constant in time. The described behaviour is typical for all samples that are thin in comparison with the effective diffusion length L.

For $d \leqslant 20$ cm the rapid initial change in exhalation rate is well hidden in the concentration curves even if the time scale is expanded (Fig. 2c). In the past, experimentalists have judged the initial part of activity growth as truely linear and corresponding to the free exhalation rate. This is in conflict with the results of the time-dependent diffusion theory predicting that the mean exhalation rate during the first few hours is much closer to the final steady-state exhalation rate than the free exhalation rate.

The influence of leakage is exemplified in Figure 3. The initial slope of all concentration curves is approximately the same, corresponding roughly to the final steady-state exhalation of the radon-tight can ($\mu=1$). A few hours after closure the exhalation rate starts to increase again for all leaking cans since the increase in radon concentration in the outer volume is not fast enough due to radon atoms leaking out. This unexpected behaviour has been explained in detail elsewhere (Sa87).
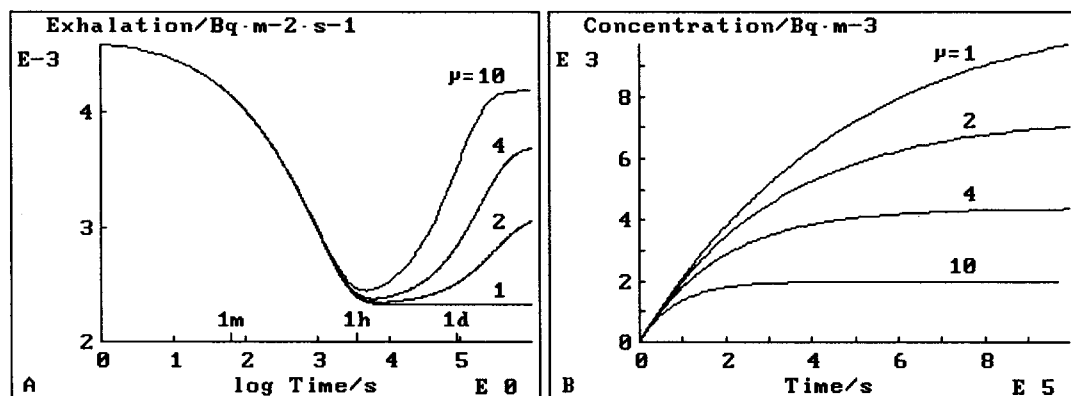

Figure 3. Exhalation and outer volume concentration for the 20 cm thick sample in Fig.2 but in leaking cans. The relative leakage $\mu = (\lambda+\nu)/\lambda$.

The diffusion theory predictions for samples with different diffusion lenghts are displayed in Figure 4. If the thickness of the sample is much larger than the diffusion length, the free exhalation rate prevails for a substantial time and thus can be easily measured with the closed-can technique. The prolonged period of free exhalation is due to the slow increase in radon concentration in the outer volume. The radon
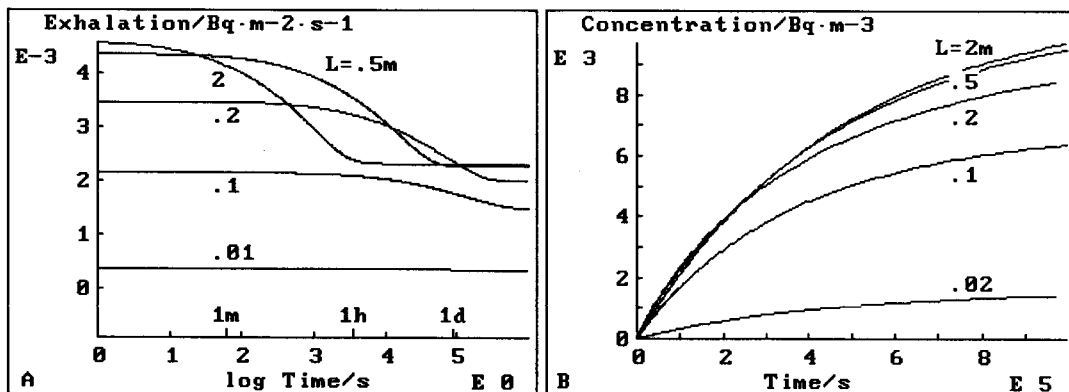
Fig. 4. The exhalation and concentration caused by the d= 20 cm sample in Fig. 2 but with different diffusion lengths L.

concentration in the pore volume has, in this case, enough time initial-ly to 'follow' the concentration increase in the outer volume. The conclusion from Figure 4 is that the thicker the sample (relative to the diffusion length) the more the disturbance of free exhalation is de-layed, and for very thick samples free exhalation prevails for such a long time that an experimental determination is feasible.

The results in this paper are based on time-dependent diffusion theory and the following main assumptions:
 1) Fick's law is valid for porous samples.
 2) Prior to time t=0 the sample is in equilibrium with a surrounding radon concentration equal to zero.
 3) There is an instantaneous mixing of the gases in the outer volume.
None of these assumptions is neccessarily true to 100% in the practical use of the closed-can method. Despite these reservations it should be stressed that the exhalation school, which identifies the free exhala-tion rate with the mean exhalation rate during the first few hours after closure, has no support from the time-dependent diffusion theory, except for samples which are very thick compared with the diffusion length.

References:
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