Radon-222 Accumulation in Air Confined by Phosphogypsum-Bearing Boundaries: Simulation for Distinct Exhaling Scenarios

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Abstract: - Understanding radon-222 exhalation from phosphogypsum-bearing materials and its accumulation in confined air is fundamental for radiation exposure assessment and radiological protection design based on human health risks. Despite evoking time dependence, zero-order models with respect to space assume uniform distribution of radon-222 activity. If detailed description is desired, one must consider spatial dependence as well. Accordingly, this work proposes a time-varying two-dimensional diffusion-dominant model where radon-222 self-decay is accounted as a sink term in the governing differential equation while its exhalation from boundaries is modelled as Neumann-type boundary condition. By adapting an existing academic simulator for transport phenomena, distinct exhaling scenarios are numerically simulated and discussed bearing in mind the failure of the aforesaid homogeneous distribution hypothesis as well as future extensions of the simulator.

Key-Words: - mass transfer, modelling, simulation, phosphogypsum, radon-222, radiological protection

1 Introduction

Aiming at lowering costs, scientific attention has been dedicated to alternative construction materials with similar characteristics as those presented by conventional ones. Quest for surrogate and cheaper building materials can be two-fold interesting since it can be environmentally friendly (as low-value residues can be recycled and employed) while it may help reducing dwelling deficits (as construction costs diminishes).

Accordingly, research points to the replacement of gypsum. Basically composed by calcium sulphate dihydrate ($CaSO_4 \cdot 2H_2O$), gypsite is the raw material to obtain ordinary gypsum (i.e., calcium sulphate hemihydrate) via thermal dehydration. However, its industrial production can become restricted or even economically unfeasible thanks to transportation costs from production sites (usually near ores) to major urban consumption centres.

In contrast, due to large demands for phosphate fertilizers, enormous quantities of phosphogypsum have been by-produced worldwide for years [1], in a proportion of 4 up to 5 tons of phosphogypsum for each ton of processed P_2O_5. Despite it is essentially $CaSO_4 \cdot 2H_2O$, such industrial waste is likely to be simply piled up in the vicinity of phosphate fertilizer plants [2], thus requiring considerable open space.

Phosphogypsum has currently little economic value (if any) because its disposal and management may derive environmental issues, notably those with regard to radon (Rn-222). Even so, its commercial exploitation has been sought and one may point to soil amendment (although only in small amounts), mine recovery and alternative material for civil engineering (e.g., building component, embankment filling and road base) [2]. With respect to its use as a surrogate building material, there are radiological constraints about Rn-222 exhalation. If well above safe levels, lung cancer risks can be assigned to the radiation released by Rn-222 [3]-[5].

Together with its short-lived decay products, Rn-222 accounts for most human exposure to radiation from natural sources [6] and the assessment of its exhalation rate is central for radiological protection. Belonging to U-238 chain, Rn-222 is a radioactive inert gas resulting from the alpha-decay of Ra-226, the later being a radionuclide usually found among phosphogypsum impurities. Therefore, Ra-226 may decay to Rn-222, which can percolate the porous matrix towards the solid-air interface, mix up with air and be inhaled by occupants.

In actual fact, soil is a major source for indoor Rn-222 concentrations [7],[8]. The problem is that one may also claim for construction materials [9],

especially if phosphogypsum is somehow used as a substitute for ordinary gypsum. Hence, a model for Rn-222 exhalation from phosphogypsum-bearing materials and its accumulation in air can be useful for radiation exposure assessment and radiological protection design based on radon-induced human health risks.

Despite accounting for time-dependence, models for Rn-222 accumulation in indoor air or in building substructures (e.g., slab-on-grade, crawl space or basement) have dealt with bulk (i.e., average) values [10],[11]. Those are typified as zero-order models with respect to space-dependence so that Rn-222 activity concentration is presumably uniform, i.e., at any instant one deals with a representative value for the entire domain of interest.

One may obtain detailed knowledge of Rn-222 distribution via higher order models [12], which are convenient (or necessary) if the solution domain is partially or wholly filled with the phosphogypsum-bearing sample [13],[14]. In addition, simultaneous processes may influence Rn-222 exhalation and accumulation inside dwellings [15]. Comprehensive analysis may evoke numerical methods, particularly if transient effects and/or convective transfer must be considered. Since early models, Rn-222 transport in porous media has basically assumed pressure-driven interstitial fluid flow in line with Darcy's law [8],[16]-[19], while simulations have been extended to include pressure fluctuations [20].

Analogous to numerous heat and/or mass transfer problems, numerical simulation comes forward as a robust solution method, which the nuclear engineer or physicist may evoke to gather understanding on Rn-222 exhalation from phosphogypsum-bearing materials and its accumulation in air. In other words, one may rely on numerical simulations to obtain detailed Rn-222 distributions in space and time.

In view of that, the present work puts forward a time-dependent two-dimensional model for Rn-222 exhalation from phosphogypsum-bearing walls and its accumulation in confined air. Aiming at the study of distinct exhaling scenarios, the model was then numerically implemented by means of adaptating an existing academic (non-commercial) finite-volume simulator that has been successfully employed to investigate transport phenomena.

2 Model for Rn-222 exhalation and its accumulation in indoor air

Knowledge of Rn-222 activity concentration inside phosphogypsum-bearing materials is necessary to evaluate the corresponding exhalation rate. In order to derive a governing equation for Rn-222 activity, one may assume that its total activity in the solid matrix is composed by those in interstitial air (filling the pores), in interstitial water (for wet materials) and in phosphogypsum itself [19]. As far as the later is concerned, only a smaller amount is available for exhalation, together with the interstitial component. It comprises Rn-222 particles that succeed to reach the solid-fluid interface, after decaying from Ra-226 trapped among phosphogypsum impurities. For this reason, Rn-222 transport within the porous medium refers to the total mobile activity, which is here assumed to be homogeneously distributed inside the building material thus yielding a uniform exhalation into the neighbouring air.

2.1 Primitive variables formulation

Time-varying Rn-222 activity distributions within a two-dimensional domain have been investigated in a previous work [12]. By evoking the same model for Rn-222 exhalation and accumulation in air, Rn-222 activity concentration $C = C(x,y,t)$ is simulated for any instant t and position (x,y) in a square domain as sketched in Fig. 1. One may think of applying such model to a vertical x-y plane within a closed room or test chamber, suitably distant from the two vertical parallel end walls.

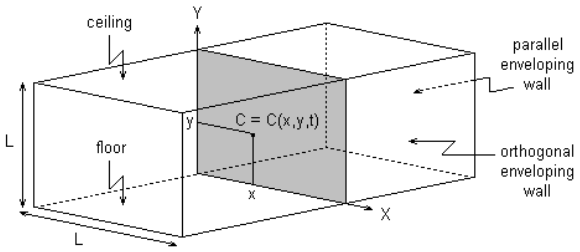


Fig. 1. Two-dimensional domain for simulations (xy-plane in a closed room or test chamber).

In simulations here performed, it is assumed that top and vertical boundaries (ceiling and enveloping walls) might contain phosphogypsum, except for the bottom horizontal boundary (floor), which is assumed to be impermeable. Consequently, the two vertical sides plus the top horizontal one may exhale Rn-222. By supposing constant Rn-222 diffusivity D in air while disregarding either thermal effects or convective transfer, Rn-222 activity concentration in air is governed by the following diffusive-dominant transport equation [12]:

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2}\right) - \lambda C \qquad (1)$$

where λ is Rn-222 self-decay constant so that $-\lambda C$ behaves like a sink term in Eq. (1).

Neumann boundary conditions are here assumed so that Rn-222 activity fluxes (exhalation) must be prescribed. Floor impermeability is expressed as a null-flux condition, $J_y = 0 = \partial C/\partial y$, whereas Rn-222 exhalation from a particular boundary obeys Fick's law of diffusion, $J_x = -D(\partial C/\partial x)$ or $J_y = -D(\partial C/\partial y)$. In opposition, if a particular boundary contains no phosphogypsum, negligible flux (or impermeability) is evoked, $\partial C/\partial x \approx 0$ or $\partial C/\partial y \approx 0$. Back-diffusion effects are neglected so that exhalation rates are not influenced by Rn-222 concentration close to the boundary. As discussed in [12], such assumption is reasonable on condition that the exhaling material thickness x_c is much smaller than the domain length L. Similar to the phosphogypsum building boards (panels) considered in [12], the present work equally assumes $x_c/L << 1$.

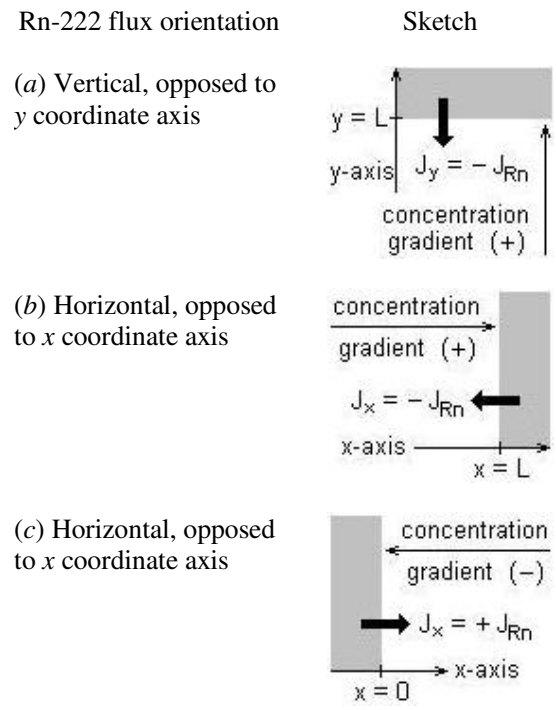


Fig. 2. Sign considerations when prescribing Rn-222 exhalation (species fluxes) at domain boundaries.

As far as the flux sign is concerned, care must be exercised when prescribing Rn-222 exhalation rates. Let first be considered species fluxes opposed to the coordinate axis orientation, as sketched in Fig. 2(*a*) for $y = L$ (ceiling) or in Fig. 2(*b*) for $x = L$ (right vertical wall). If J_{Rn} is the absolute value of Rn-222 exhalation flux, at those boundaries fluxes must be respectively set as (*a*) $J_y = -J_{Rn}$ and (*b*) $J_x = -J_{Rn}$. As shown in Fig. 2(*c*) for $x = 0$ (left vertical wall), Rn-222 flux is then expressed as $J_x = +J_{Rn}$ since it is in the same orientation of the coordinate axis.

2.2 Dimensionless variables formulation

The model framework is cast in dimensionless form as an attempt to lump concurrent effects into fewer dimensionless parameters. Given the characteristic length L of the solution domain, the following dimensionless variables are introduced:

$$X = \frac{x}{L} \quad , \quad Y = \frac{y}{L} \quad , \quad \tau = \lambda t \quad , \quad \phi = \frac{C - C_0}{\Delta C} \qquad (2)$$

where ΔC is an activity concentration scale suitably defined according to the scenario investigated while C_0 is Rn-222 activity concentration at $t = 0$. For the sake of simplicity, one may assume that the domain is initially free from Rn-222 thus leading to $C_0 = 0$. In terms of the dimensionless activity concentration ϕ, one casts Eq. (1) into the following form:

$$\frac{\partial \phi}{\partial \tau} = \frac{1}{R}\left(\frac{\partial^2 \phi}{\partial X^2} + \frac{\partial^2 \phi}{\partial Y^2}\right) - \phi \quad , \quad R = \frac{\lambda L^2}{D} \qquad (3)$$

where R is a dimensionless parameter measuring the magnitude of Rn-222 self-decay in relation to its diffusion, which has been referred to as decay-to-diffusion ratio [12]-[14]. In those references, one may also find additional dimensionless parameters related to Rn-222 transport (namely, emanation-to-diffusion and emanation-to-decay ratios).

Due to the Neumann nature of evoked boundary conditions, one may suitably define the scale ΔC for Rn-222 activity concentration as:

$$\Delta C = \frac{J_{Rn} L}{D} \qquad (4)$$

Accordingly, dimensionless boundary condition for a vertical exhaling boundary results as:

$$\frac{\partial \phi}{\partial X} = -1 \quad (\text{for } 0 \le Y \le 1) \qquad (5)$$

at the left boundary ($X = 0$) or as:

$$\frac{\partial \phi}{\partial X} = +1 \quad (\text{for } 0 \le Y \le 1) \tag{6}$$

at the right one ($X = 1$). The dimensionless boundary condition for a exhaling top boundary ($Y = 1$) is:

$$\frac{\partial \phi}{\partial Y} = +1 \quad (\text{for } 0 \le X \le 1) \tag{7}$$

Dimensionless boundary condition for negligible flux or impermeability (wherever evoked) becomes:

$$\frac{\partial \phi}{\partial X} = 0 \quad (\text{for } 0 \le Y \le 1) \tag{8}$$

at any vertical boundary (either $X = 0$ or $X = 1$) or:

$$\frac{\partial \phi}{\partial Y} = 0 \quad (\text{for } 0 \le X \le 1) \tag{9}$$

for either bottom ($Y = 0$) or top boundary ($Y = 1$).

In view of Eq. (4), scale ΔC depends on Rn-222 exhalation rates J_{Rn}, which in turn depends on the phosphogypsum content of the material [11],[12]. The lack of information about J_{Rn} does not prevent one from performing simulations. On the contrary, as discussed ahead, one may carry out numerical simulations to obtain J_{Rn}, thus characterizing a given exhaling phosphogypsum-bearing material.

3 Numerical method

Aiming at design, scale-up and/or optimization, one may rely on numerical simulation to reduce the number of tests, thus saving material and/or human resources. In order to yield detailed distributions of Rn-222 activity concentration, an existing academic simulator has been continuously adapted [13],[14]. Programmed in FORTRAN, such non-commercial simulation code has been successfully employed to investigate coupled heat and mass transfer problems in media partially or fully filled up with porous material [21]-[24].

Able to deal with time-dependent phenomena in two-dimensions, such simulator numerically solves coupled partial differential equations related to conservation principles (i.e., bulk mass, momentum, energy and species). In this work, the code was adjusted to simulate time-varying Rn-222 activity concentration in air (dimensionless formulation), in a square domain as previously described.

Following a finite-volume discretization method, governing equations are converted into an algebraic system, which is solved through tri-diagonal matrix algorithm (TDMA) [25]. Numerical simulations were performed on uniform orthogonal grids while under-relaxation factor was set at 0.9. For the sake of information (though not used in simulations here carried out), continuity and momentum equations are coupled in the code via SIMPLER algorithm while staggered grids are adopted in order to prevent pressure oscillations [25].

4 Results and discussion

By reminding that the bottom boundary (floor) is allegedly free from phosphogypsum, four distinct exhaling scenarios are investigated. As sketched in Fig. 3, differences between them refer to whether or not a given boundary contains phosphogypsum, thus leading to Rn-222 exhalation into the adjacent air.

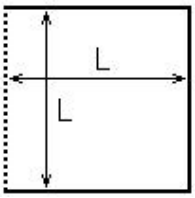
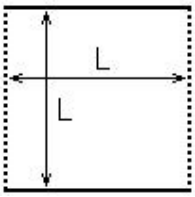
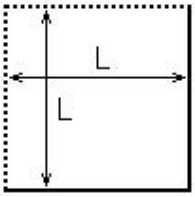
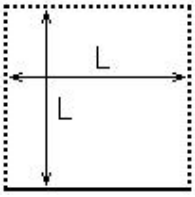
Exhaling scenario	Sketch
(*a*) Configuration "A": One vertical (left) exhaling boundary	
(*b*) Configuration "B": Two vertical (left and right) exhaling boundaries	
(*c*) Configuration "C": One vertical (left) and one horizontal (top) exhaling boundaries	
(*d*) Configuration "D": Two vertical (left and right) and one horizontal (top) exhaling boundaries	

Fig. 3. Exhaling configurations simulated (solid line = impermeable or negligible-flux boundary; dashed line = uniform Rn-222 exhaling boundary).

Radon-222 self-decay constant is $\lambda = 2.098 \times 10^{-6}$ s^{-1} whereas its diffusivity in open air has been reported as either $D = 1.2 \times 10^{-5}$ m^2·s^{-1} [7] or $D = 1.1 \times 10^{-5}$ m^2·s^{-1} [6],[18]. If $L = 3.5$ m is set as the characteristic length of the domain, the decay-to-diffusion ratio for scenarios simulated in this work results as $R \cong 2.3364$, according to Eq. (3).

4.1 Mesh sensitivity analysis

As far as the mesh size is concerned, a compromise between the number of grid points and numerical accuracy was examined by solving a steady-state simplification of Eq. (3), namely:

$$\frac{1}{R}\left(\frac{\partial^2\phi}{\partial X^2}+\frac{\partial^2\phi}{\partial Y^2}\right)-\phi=0 \qquad (10)$$

subjected to impermeable boundaries (i.e., to either $\partial\phi/\partial X = 0$ or $\partial\phi/\partial Y = 0$) except for the left vertical one (at $X = 0$), for which an exhaling condition is imposed (i.e., $\partial\phi/\partial X = -1$). In line with Fig. 3(*a*), such scenario is referred to as configuration "A".

The variation of the simulated value for ϕ at the midpoint $X = Y = 0.5$ as a function of the number N of grid points along each coordinate x and y (so that the total number of points is $N \times N = N^2$) is shown in Fig. 4. As a compromise between convergence and computational effort, the 100×100 mesh was chosen for the subsequent sensitivity analysis with respect to the dimensionless advancing time step $\delta\tau$.

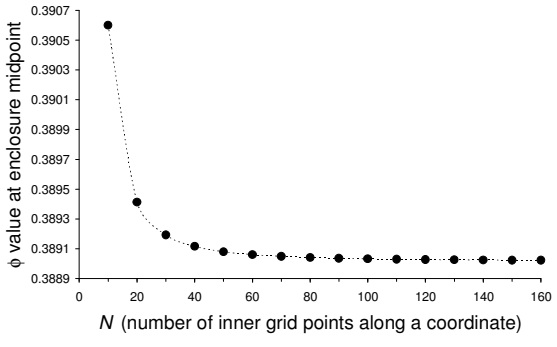


Fig. 4. Mesh sensitivity analysis with respect to grid spacing: ϕ simulated at the midpoint for different number of inner grid points (steady-state scenario).

In order to set $\delta\tau$ for time-varying simulations, the complete differential equation, Eq. (3), was solved for the same scenario (namely, configuration "A"), using the 100×100 mesh in the time interval $0 \le \tau \le 1$ (which refers to a simulated period of about $1/\lambda = 5.5$ days). The variation of value ϕ simulated at the midpoint and at the final instant $\tau = 1$ as a function of distinct $\delta\tau$ values is shown in Fig. 5. Again aiming at a compromise between accuracy and computational effort, $\delta\tau = 0.001$ was chosen so that numerical results shown hereafter were carried out using the 100×100 mesh and such advancing dimensionless time step.

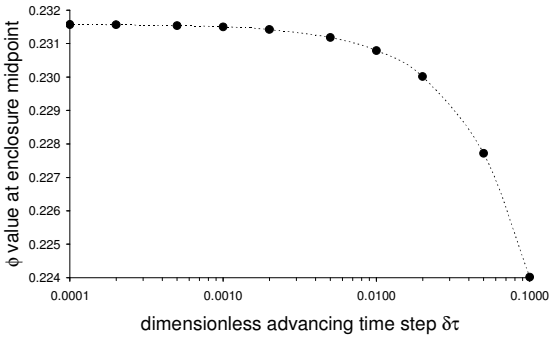


Fig. 5. Mesh sensitivity analysis with respect to advancing time steps: ϕ simulated at the midpoint at the final instant $\tau = 1$ (time-varying scenario).

4.2 Simulation of Rn-222 accumulation

Dimensionless Rn-222 activity concentration $\phi = \phi(X,Y,\tau)$ are numerically simulated for each exhaling scenario considered in this work (Fig. 3). Resulting concentration isolines at increasing dimensionless time instants are presented for configuration "A" in Fig. 6, configuration "B" in Fig. 7, configuration "C" in Fig. 8 and configuration "D" in Fig. 9. Time instants considered were $\tau = 0.25$, 0.50, 0.75 and 1.00 (which refer to approximately after 33 h, 66 h, 99 h and 132 h, respectively).

An interesting issue to ponder has to do with the location of the symmetry axis with reference to the ϕ distribution within the domain, for each exhaling configuration. As one would expect by examining the scenarios shown in Fig. 3, configuration "A" is symmetric about the horizontal axis through $Y = 0.5$, configuration "D" is symmetric about the vertical axis through $X = 0.5$ while those two symmetry axes are present in configuration "B". Configuration "C" is symmetric about the diagonal linking the upper left and lower right corners. It is worth noting that all those symmetry axes are retained throughout the time evolution of the concentration distribution. In other words, all time-dependent distributions retain their own symmetry patterns.

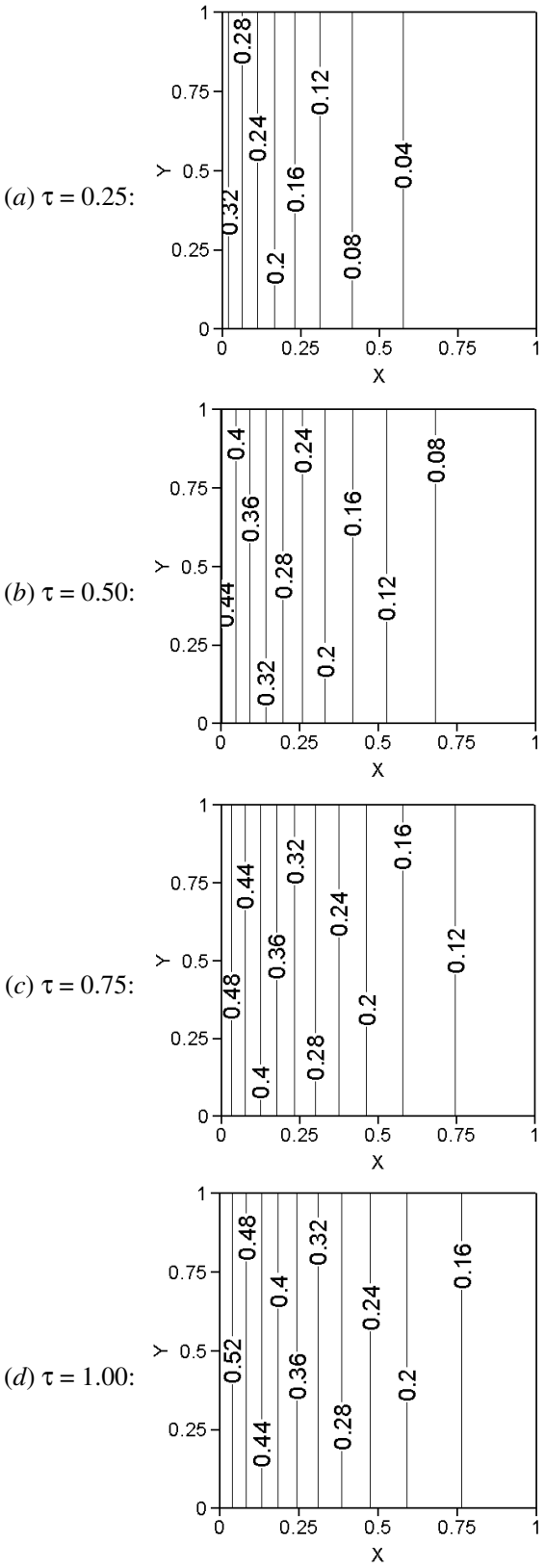


Fig. 6. Dimensionless Rn-222 concentration isolines simulated at increasing dimensionless time instants: exhaling configuration "A".

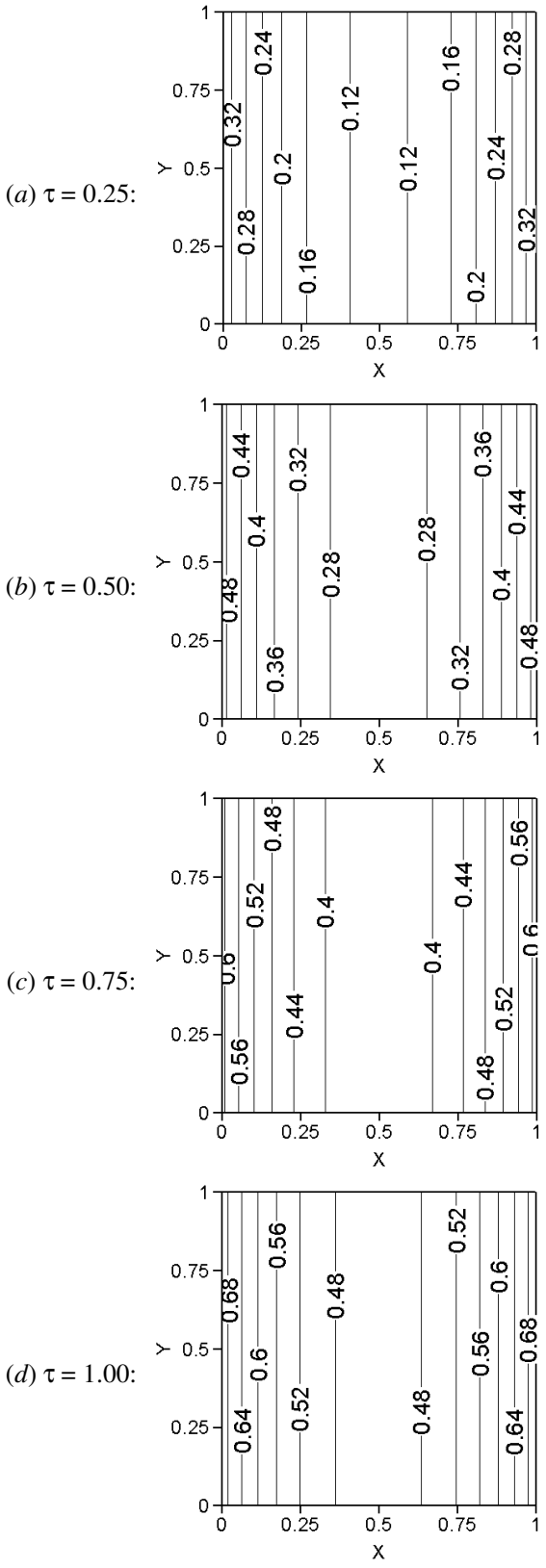
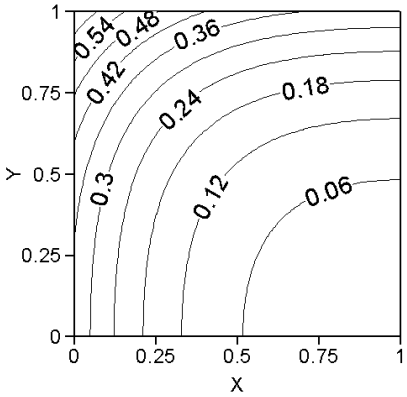
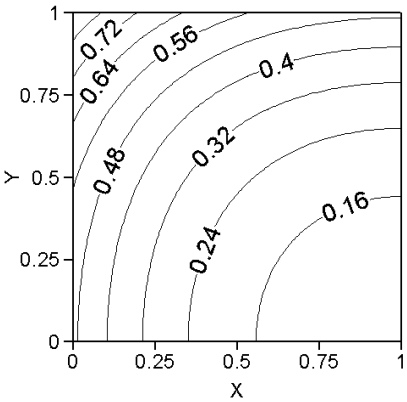


Fig. 7. Dimensionless Rn-222 concentration isolines simulated at increasing dimensionless time instants: exhaling configuration "B".

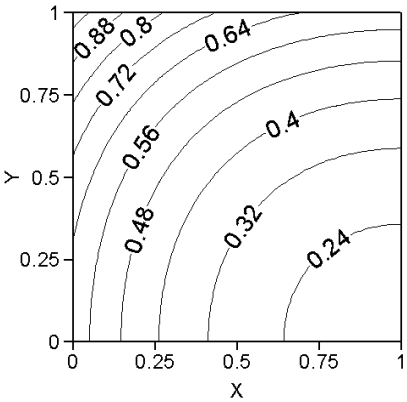
(*a*) $\tau = 0.25$:



(*b*) $\tau = 0.50$:



(*c*) $\tau = 0.75$:



(*d*) $\tau = 1.00$:

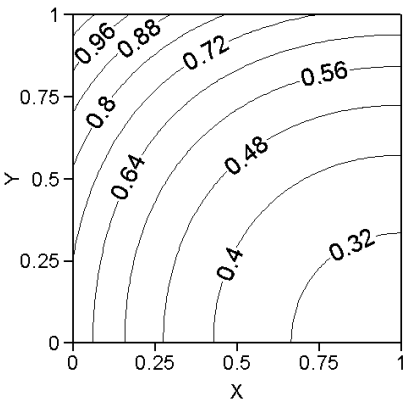
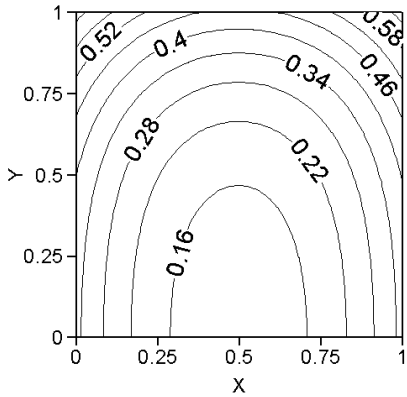
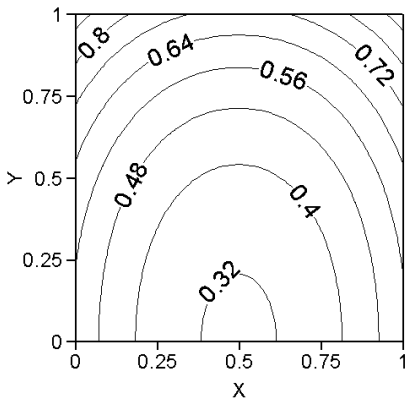


Fig. 8. Dimensionless Rn-222 concentration isolines simulated at increasing dimensionless time instants: exhaling configuration “C”.

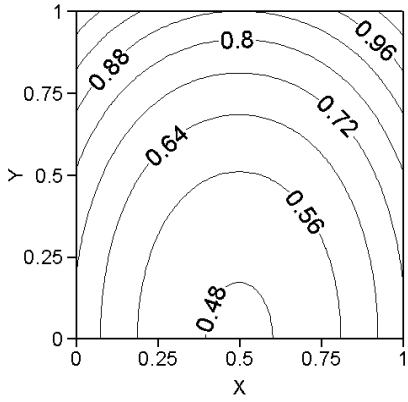
(*a*) $\tau = 0.25$:



(*b*) $\tau = 0.50$:



(*c*) $\tau = 0.75$:



(*d*) $\tau = 1.00$:

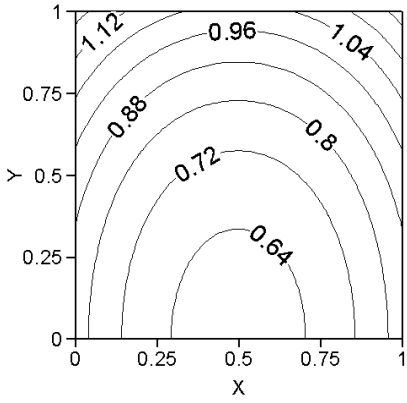


Fig. 9. Dimensionless Rn-222 concentration isolines simulated at increasing dimensionless time instants: exhaling configuration “D”.

It is also important to note that no homogeneous Rn-222 distribution is verified whatsoever inside the solution domain. As dimensionless time advances, increasing concentration levels are obtained since no air exchange mechanism is accounted for in the model. Such fact explains why air renewal is indeed necessary to make a dwelling habitable by humans and acceptable according to Rn-222 standards.

The dimensionless time range here considered ($0 \leq \tau \leq 1$) was not long enough for steady-state to be reached by any exhaling scenario. In view of Eq. (3) or (10), steady-state is reached as soon as $\partial^2\phi/\partial X^2 + \partial^2\phi/\partial Y^2. = R\cdot\phi$, throughout the solution domain.

One may use numerical simulations as means to characterize phosphogypsum-bearing materials with respect to Rn-222 exhalation rates. For instance, by evoking the proportionality between ϕ and C as given by Eq. (2), one may estimate scale ΔC_{est} by fitting numerically simulated values ϕ_{num} for the dimensionless Rn-222 activity concentration at a particular position (e.g., midpoint of the solution domain) against experimentally measured values C_{exp} for Rn-222 activity concentration at the same position (e.g., midpoint of a geometrically similar prototype). One may then manipulate Eq. (2) in order to obtain:

$$\phi = \frac{C - C_0}{\Delta C} \quad \Rightarrow \quad \Delta C_{est} = \frac{C_{exp} - C_0}{\phi_{num}} \qquad (11)$$

Once scale ΔC_{est} is known together with Rn-222 diffusivity D and characteristic length L, one may estimate related Rn-222 exhalation rates $J_{Rn,est}$ with the help of Eq. (4), namely:

$$\Delta C = \frac{J_{Rn} L}{D} \quad \Rightarrow \quad J_{Rn,est} = \frac{\Delta C_{est} D}{L} \qquad (12)$$

As already pointed out, Rn-222 exhalation rates are key parameters for radiological protection design and/or radiation exposure assessment. For the sake of comparison, those rates from phosphogypsum-bearing walls have been reported to range as $0.0014 \leq J_{Rn} \leq 0.0111$ Bq·m^{-2}·s^{-1} [9].

At this point, one should remember that $C_0 = 0$ was assumed in the present model. Whenever $C_{0,exp} \neq 0$, such non-zero initial concentration prevailing in the closed room (chamber) should be experimentally measured to be properly introduced in Eq. (11). Again for comparison purposes, outdoor air-borne Rn-222 activity concentration have been measured as approximately 10 Bq·m^{-3} [6].

It is worth recalling that air was here assumed to be isothermal and motionless so that thermal effects and convective (mass and/or energy) transfers were disregarded. Otherwise, continuity, momentum and energy equations should be evoked in the model and numerically solved while Rn-222 activity (species) equation should be extended so as to accommodate those additional phenomena. Such comprehensive approach is left for forthcoming developments of this work.

Indeed, other effects include not only convective transport due to air motion (related to the existence of fans, doors and/or windows) but also the presence of the phosphogypsum-bearing material itself in the solution domain (which can be extended up to two or even three dimensions). The nuclear physicist or engineer should evoke numerical methods to solve the resulting partial differential equation system.

4 Concluding remarks

Aiming at large-scale (and commercial) exploitation of phosphogypsum as alternative building material, knowledge of Rn-222 exhalation and accumulation in indoor air is fundamental for proper radiological protection design. Zero-order model frameworks with respect to spatial coordinates have been used to simulate time-dependent Rn-222 concentration in indoor air. In those models, uniform Rn-222 activity distribution is supposed throughout the domain of interest. Nevertheless, such hypothesis breaks down if point-to-point variation is desired.

Accordingly, the present work proposed a time-varying second-order model for Rn-222 activity concentration, which was implemented by adapting an existing academic (i.e., non-commercial) finite-volume simulator for transport phenomena in porous media. Numerical simulations were carried out for a square domain subjected to four distinct diffusion-dominant Rn-222 exhaling scenarios.

While expected symmetries were retained during the time evolution of Rn-222 activity distributions in simulated scenarios, numerical results confirmed that no homogeneous distribution occurs inside the solution domain. As Rn-222 activity concentrations were observed to be particularly higher near upper corners, one may overestimate activity levels (in comparison to average levels) if recordings are only accomplished in the vicinity of those corners.

Having in mind the ability to deal with more general situations, a primary objective is to widen the model framework in order to include additional phenomena (e.g., thermal effects and/or convective heat/mass transfer) while being extended to three-dimensional solution domains. The academic finite-

volume simulator must be properly adapted as well in order to cope with those model extensions.

Nomenclature

C	Rn-222 activity concentration ($Bq \cdot m^{-3}$)
D	Rn-222 diffusivity in open air ($m^2 \cdot s^{-1}$)
J	Rn-222 flux / exhalation ($Bq \cdot m^{-2} \cdot s^{-1}$)
L	Characteristic length of solution domain (m)
N	Number of inner grid points (dimensionless)
R	Decay-to-diffusion ratio (dimensionless)
t	Time (s)
X, Y	Cartesian coordinates (dimensionless)
x, y	Cartesian coordinates (m)
x_c	Thickness of phosphogypsum material (m)

Greek symbols

λ	Rn-222 decay constant (s^{-1})
τ	Time (dimensionless)
ϕ	Rn-222 activity concentration (dimensionless)
ΔC	Rn-222 activity concentration scale ($Bq \cdot m^{-3}$)
$\delta\tau$	Advancing time step (dimensionless)

Subscripts and superscripts

est	Estimated (assessed) value
exp	Experimentally measured value
num	Numerically simulated value
Rn	Absolute value for Rn-222 flux / exhalation
x, y	Directions along Cartesian coordinates
0	Initial value for Rn-222 activity concentration

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