

# PHYSICAL DISTRIBUTIONS OF RADON DECAY CHAIN ACTIVITIES IN AIR

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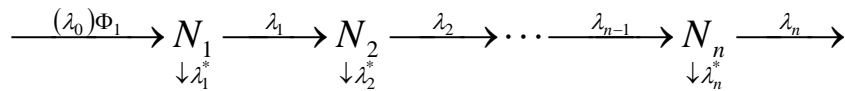
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## 1. Introduction

The distribution of short-lived radon decay chain activities in air – in time, space and on aerosols – determines their exposure potential and measurement thereof. Basic branching flow string calculations are shown below that can be applied, in combination, to quantify various distributions which arise with sources and sinks in flow compartments, including ‘internal’ flow of one chain member by decay to the next. For ‘flow’ chains where the parent nuclide has the longest half-life, and this is not separated from the progeny, successive progeny activities (concentrations) are restricted to a (transient) maximum ratio to the parent or precursors. For a compartment with a constant total source flux  $\Phi_{Rn}$  (from  $^{226}\text{Ra}$  or  $^{224}\text{Ra}$ ) the total activity of chain members in the compartment is similarly restricted. The activity ratios can be useful in evaluation of various activity distributions, which have a bearing on sources and sinks regarding remediation decisions for buildings with high exposure potential.

## 2. Flow strings for distribution calculations of string members

A basic branching flow string can be presented as



where the  $\lambda$ 's are flow variables of supply and removal of individual string members, so that at each string node  $dN_i/dt = \lambda_{i-1}N_{i-1} - (\lambda_i + \lambda_i^*)N_i$ ; any of the  $\lambda$ 's and/or  $\lambda^*$ 's may be decay constants or variables. A constant flux  $\Phi_1$  to the string can be envisaged as supply from a very long-lived (precursor) source  $\lambda_0 N_0$ . Removal at a node can be supply to the next string member or away from the string – combined removal is denoted as  $\Lambda_i = \lambda_i + \lambda_i^*$ . Complete flow networks can be assembled from, or unravelled into different flow strings, not necessarily of equal length.

For an individual flow string equation 1 gives the steady state activities to string member n, i.e. for constant flux  $\Phi_1$  and  $\lambda$ 's over a period much greater than the longest  $(\lambda_i + \lambda_i^*)^{-1}$ ;  $S_{1,n}$  may be considered the saturation coefficient of a node activity relative to the source  $\Phi_1$ , and  $S \leq 1$ :

$$S_{1,n} = \frac{\lambda_n N_{n,t}}{\Phi_{1,t \rightarrow \infty}} = \prod_{i=1}^n \frac{\lambda_i}{\Lambda_i} \tag{1}$$

For the dynamic state two string equations suffice for assembling stepwise all flow network situations; they are mere extensions of the familiar  $1 - e^{-\lambda t}$  and  $e^{-\lambda t}$  (saturation) equations for successive members with branching. The case of sampling a constant flux  $\Phi_1$  to a flow string and starting with zero activities at start time  $t=0$ . Here equation 1 has additional, time dependent components building up towards steady-state saturation:

$${}_s S_{1,n} = \frac{\lambda_n N_{n,t}}{\Phi_{1,0 \rightarrow t}} = \left( \prod_{i=1}^n \frac{\lambda_i}{\Lambda_i} \right) - \sum_{i=1}^n \frac{e^{-\Lambda_i t}}{\Lambda_i} \prod_{j=1}^n \frac{\lambda_j}{\Lambda_j - \Lambda_i + \delta_{i,j}} \tag{Kronecker } \delta_{ij} \tag{2a}$$

The second case is zero flux,  $\Phi_1 = 0$  and initial condition of all but the first activity being zero,  $N_{n>1, t=0} = 0$ , so that the additional chain activities are formed and all decay away:

$${}_d S_{1,n} = \frac{\lambda_n N_{n,t}}{\lambda_1 N_{1,t=0}} = \frac{1}{\lambda_1} \sum_{i=1}^n e^{-\Lambda_i t} \prod_{j=1}^n \frac{\lambda_j}{\Lambda_j - \Lambda_i + \delta_{j,i}} \quad 2b$$

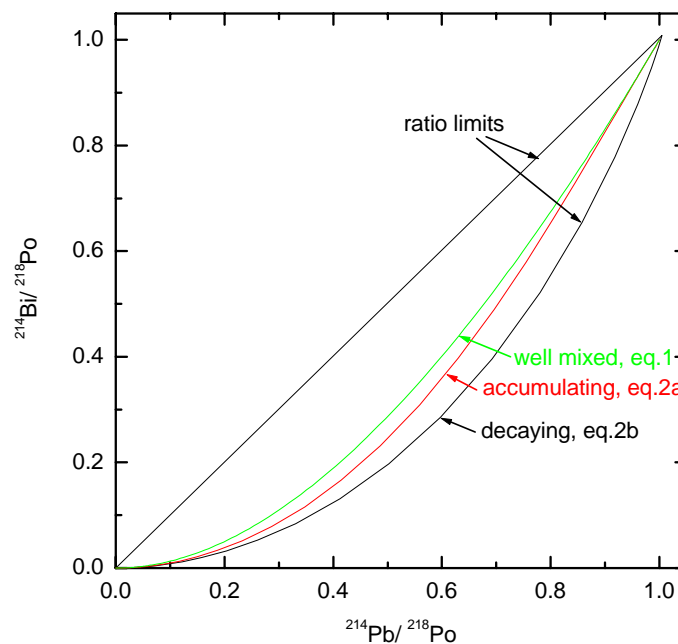
The leading subscripts to  $S$  indicate sampling a flux or decay.

The activity response of a string member to a unit flux  $\Phi_{1,0 \rightarrow t}$ , for known  $\lambda$ 's, can be evaluated over any period after the start of the period. From a measurement response record the actual  $\Phi$ 's and  $\lambda$ 's for periods, and concentrations, can be reconstructed provided there is adequate independent measurement data (degrees of freedom).

### 3. Rn-222 decay product activity ratios in air – steady state

Since individual  $^{222}\text{Rn}$  decay product activity concentrations in air can readily be determined to good accuracy (considerably better than the gas), it is appropriate to assess further information contained in their relative distributions. (As a side benefit the decay product concentrations directly furnish the exposure relevant potential alpha energy concentration (exposure) without the large uncertainty from Rn concentration via a stipulated Rn equilibrium factor  $F_{\text{PAE}}$ ).

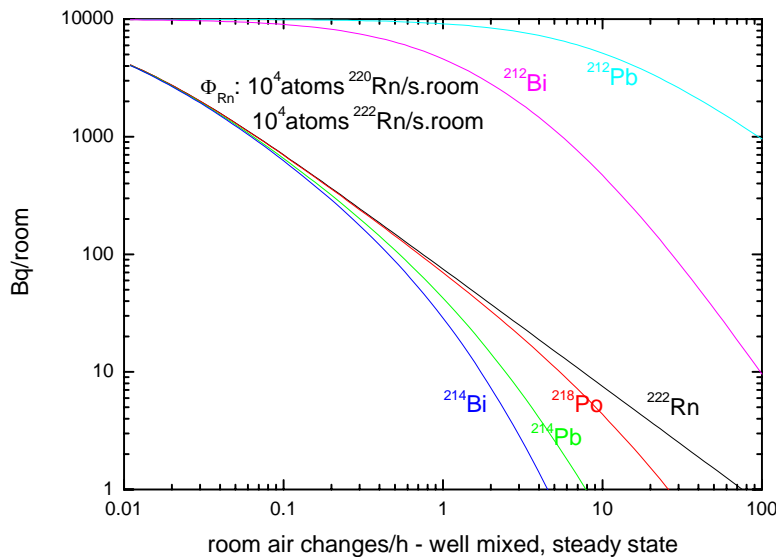
In Fig.1 the three curves show  $^{222}\text{Rn}$  decay product activity ratio relationships which can occur directly according to equations 2b, 2a and 1 respectively. The lowest curve corresponds to the ratio relationship of decay products (eq. 2b) that develop in a room with an initial quantity of  $^{222}\text{Rn}$  only, which then decays. This decay/time model is equivalent to a decay along distance model for plug flow in a tunnel with constant Rn injection at distance 0; the maximum transient decay ratio values ( $^{214}\text{Pb}/^{218}\text{Po}$  1.005 and  $^{214}\text{Bi}/^{218}\text{Po}$  1.009) are approached after several hours decay. All  $^{222}\text{Rn}$  decay product ratios are linear combinations of these ratios, so that between this (decay) relationship curve in the figure and the straight line from axis points (0,0) and (1.005,1.009) all possible pairs of activity relationship ratios in air occur. This ratio restriction had been used a few decades ago to derive optimized batch- or grab-sampling measurement methods for potential alpha energy concentration. The successive decay product ratio restriction is imposed by the far longer-lived  $^{222}\text{Rn}$  which cannot be removed from air without removing also the decay products. For the  $^{220}\text{Rn}$  chain in air there is only for  $^{212}\text{Bi}/^{212}\text{Pb}$  a maximum transient decay activity ratio value of 1.1048.



**Fig. 1.** Corresponding activity ratios of  $^{222}\text{Rn}$  decay products as per equations 1, 2a and 2b.

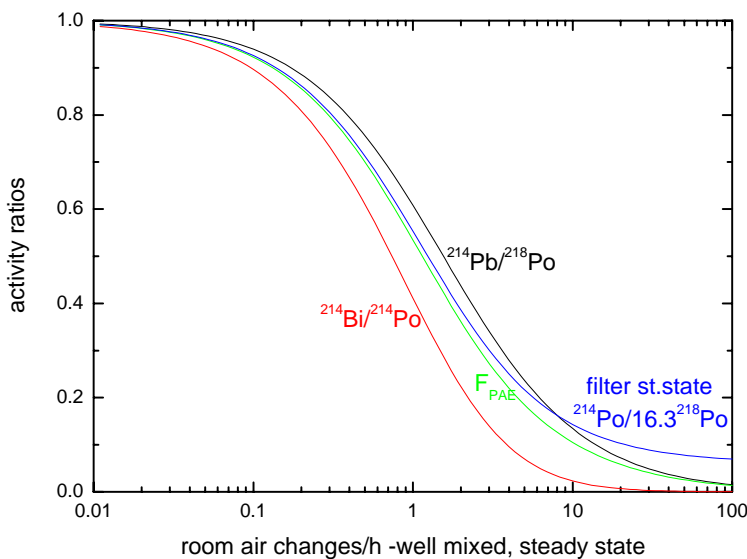
The middle curve in fig. 1 represents tunnel plug flow with constant addition of  $^{222}\text{Rn}$  along the tunnel, as per eq. 2, while the top curve per eq.1 applies to a room with air well mixed. Measurement of corresponding activity ratios and possible concentration gradients across a room thus yields the type of flow model applicable.

For the steady-state situation (eq. 1) in a room with a constant Rn flux of e.g.  $\Phi = 10\,000$  atoms Rn per second into the room, the activity concentrations (in Bq/room) of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  and of  $^{212}\text{Pb}$  and  $^{212}\text{Bi}$  are shown in Fig. 2 for various (constant) rates of air exchange,  $\lambda^*$  room volumes per hour, assuming activity free intake air and good mixing inside the room.



**Fig. 2.** Activity concentrations at a constant Rn flux and air exchange.

Figure 3 shows the corresponding air activity ratios, the  $^{222}\text{Rn}$  equilibrium factor  $F_{\text{PAE}}$ , and, for comparison, the readily measurable sampling ‘filter (+screen)’ activity ratio  $^{214}\text{Bi}/(^{218}\text{Po} \times 16.31)$ ; the steady state upper limit on a filter  $^{214}\text{Bi}/^{218}\text{Po} = 16.31 = (3.05 + 26.8 + 19.9)/3.05$  is used here to adjust to the same vertical scale.



**Fig. 3.** Air (and filter) activity ratios, Rn equilibrium factor  $F_{\text{PAE}}$  vs. air exchange

For changing Rn fluxes and air exchange, the accordingly more varying decay product concentrations can, from detailed measurement be unfolded to assess the source strengths of Rn and ventilation (sinks) – the measurements themselves require dynamic evaluation of instrument recorded data.

#### 4. Measurement of varying decay chain member concentrations

When filtering  $^{222}\text{Rn}$  decay products from air at a given flow rate using a ‘continuous’ monitor, one deals in the instrument with three non-branching flow strings on the filter (and on the screen) and  $\lambda$  decay constants, not variables -  $\Phi_{\text{Po-218}\rightarrow\text{Po}\rightarrow\text{Pb}\rightarrow\text{Bi}}$ ,  $\Phi_{\text{Pb-214}\rightarrow\text{Pb}\rightarrow\text{Bi}}$  and  $\Phi_{\text{Bi-214}\rightarrow\text{Bi}}$ . The instrument fluxes  $\Phi$  (i.e. the concentrations sampled) vary with time, i.e. air concentrations change, but are considered constant during each ‘small’ sampling period. Applying the instrument time response functions (from eq. 2a & 2b) to used  $\Phi$  flux time intervals, all recorded counts in various time intervals and spectral ROIs can conditionally be unfolded to the original  $\Phi$  with an associated uncertainty (covariance matrix) in the various small sampling periods.

An  $\alpha$ -monitor offers only two partially overlapping  $\alpha$ -ROIs for spectrometric differentiation for the differentiation of the three strings listed above, so that contiguous sampling intervals cannot be evaluated to the individual concentrations (the data does not meet the unfolding criteria). For quasi-continuous filtering, when recording filter decays during both on and off filtering periods, the additional measurement records (degrees of freedom) allow evaluation of fluxes  $\Phi_{\text{Po}}$ ,  $\Phi_{\text{Pb}}$  and  $\Phi_{\text{Bi}}$  to the instrument, i.e. of the air activity concentrations for each filtering period. If an additional  $\beta$ -ROI is recorded, then all three concentrations can be evaluated lag-free even without ‘filtration off’ extra measuring periods.

In more accurate, longer measurements, usually the  $^{220}\text{Rn}$  flow strings  $\Phi_{\text{Pb-212}\rightarrow^{212}\text{Pb}\rightarrow^{212}\text{Bi}}$  and  $\Phi_{\text{Bi-212}\rightarrow^{212}\text{Bi}}$  also are significant (- this can be readily ascertained from ROI totals). These strings offer only one additional (degree of freedom)  $\alpha$ -ROI, so that quasi-continuous filtering is definitely required, while the additional  $\beta$ -ROI can greatly enhance the accuracy (covariance matrix) of evaluated concentrations for the various filtering periods.

Unattached Rn decay product concentrations need to be measured concurrently as, at significant contributions, they can, if neglected, distort flow evaluations substantially.

For the evaluation of time sequences of Rn-decay product concentrations, from large instrument data records and calibration tables, flexible, efficient linear modelling software has been developed. This also provides the covariance matrix for the results. Older single grab sample procedures, each lasting over the major decay of the nuclides, were very inefficient in manpower, instrumentation and evaluation – the systems were too insensitive for assessing dynamic changes in sources and air exchange. Modern computational data evaluation made continuous measurement, at much higher efficiency feasible.

#### 5. Evaluation of dynamic source-sink flow parameters from measurement data.

The further unfolding of the varying concentration data to air exchange and Rn flux involves non-linear relationships. Successive approximation simulation software with scenarios for standard type variations would probably be appropriate here. A record of additional environmental parameters, e.g. temperature, humidity, time of door opening etc., would also be useful to mark meaningful transition times. The simulation in multiple flow strings is merely enlarged if successive rooms are linked to the initial ‘Rn source’ room, or, equitably, if significant concentrations of Rn and decay products are monitored in the intake air.

The ROI data for strong diurnal cycles (e.g. factor 3 maximum concentration change per hour) in Rn decay product concentrations should allow evaluation at a time resolution under 1 hour, so that the ‘continuous change’ assumption is still reasonably applicable. When attempting to evaluate source, air exchange and levels during occupancy of a room with sometimes rapidly changing air exchange, a still better time differentiation, adjusted to expected changes in air

exchange rate, may be appropriate. At shorter time resolution of measurement the accuracy of evaluated individual results is compromised and a tradeoff should be aimed at.

## **6. Conclusions**

Test measurements and data analyses, using efficient evaluation software, have shown that variable Rn decay product concentrations can be sensitively measured (quasi-) continuously, at good time resolution. Considering extension of the evaluation to room models, using flow networks, made clear that, via physical distributions of Rn decay chain concentrations, in (room) compartments, unattached and on aerosols and individual concentration variations with time, the core parameters determining Rn exposure situations, Rn source and air exchange, can likewise be determined. An efficient software application or centralized data evaluation could make the simulation evaluation of large data sets accessible to less versed operators. Objective decisions can accordingly be made on potentially costly remediation decisions.