

Noble gas dosimetry for non-human biota

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Reasons for noble gas dosimetry

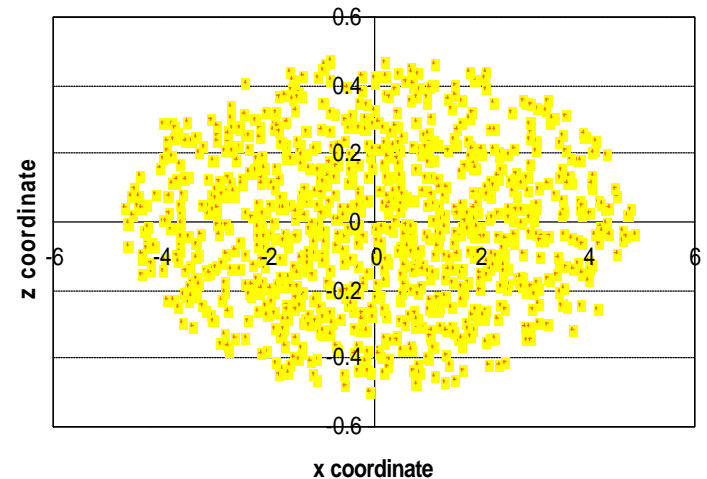
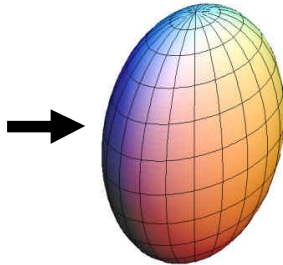
- ^{85}Kr and ^{41}Ar account for approximately 75% and 10% of airborne releases for nuclear reactors
- Smaller amounts of $^{131\text{m}}\text{Xe}$, ^{133}Xe and ^{88}Kr also released.
 - ^{41}Ar and ^{85}Kr contribute 80% from next-generation AP1000 reactors
 - ^{85}Kr the main radionuclide discharged from Sellafield ($\sim 40 \text{ PBq y}^{-1}$).
- For inert gases Ar and Kr plume immersion is the only concern.
- For Rn there is the problem of internal dose by the daughters.
- General drive to ensure that the environment is protected
 - Ongoing need to prove limited impact in new reactor designs
 - Compliance and public perception (birds roosting on stacks?)
 - The US NRC now includes the plume dose from noble gas emissions in assessing dose to biota for new reactor licensing

- In previous habitat assessments, the EA (2002) suggested ^{137}Cs as an analogue for application in assessments of ^{41}Ar and ^{85}Kr , and ^{239}Pu for ^{222}Rn and its daughters.
- However, the use of such an analogue led to highly conservative dose estimates (Beresford et al., 2004).
- Produce sufficiently robust methodology to allow the calculation of Ar, Kr and Rn doses to biota without using analogues.
- Explicit approaches developed in 2003 (Ar, Kr) and 2009 (Rn) at Westlakes Scientific Consulting, UK
- The Ar and Kr method was adopted in an updated version of the EAR&D 128 methodology.
 - To date, no such approach exists in ERICA and the method remains a valid option.

- Noble gases have a small but finite solubility in water and body fluids.
- ^{41}Ar and ^{85}Kr are inert gases and internal incorporation in animals can be neglected compared with cloud immersion.
- Noble gases are not deposited to soil (so no plant uptake, etc.).
- Will be exchanged within the air pore volume of surface soil (but small component).
- So we assume pore air concentration = ground level air concentration

Argon and krypton methodology

- Calculate DCC values using the EA R&D 128 ellipsoid-based Monte Carlo approach.
- Compare with human dose conv. factors (DCFs) from ICRP 72.
- Study dependence of DCC with area/volume ratio
- Incorporate methodology into R&D 128 terrestrial model.
- Update with ERICA geometries



$$DCC^{int} = 5.77 \times 10^{-4} \times \bar{F}_l \times \sum_i (p_i E_i)$$

$$DCC^{ext} = 5.77 \times 10^{-4} \times (1 - \bar{F}) \times \left(\sum_i p_i E_i \right)$$

- Inputs: Ar and Kr in air (measured or calculated with simple semi-infinite cloud model)
- Outputs: Convert DCC in $\mu\text{Gy h}^{-1}/\text{Bq m}^{-3}$ instead of kg^{-1} using 1.2 kg m^{-3} air density.
- Usual approximations: equilibrium transfer, ellipsoid geometries, uniform density between organism and media, uniform distribution in organism and doses averaged for whole body.
- Species considered: all terrestrial R&D 128 + “reference man” ellipsoid defined for comparison with ICRP-72.
- Method has been recently revamped to include:
 - DCCs for the ERICA reference organisms
 - DCCs for $^{131\text{m}}\text{Xe}$, ^{133}Xe and ^{88}Kr
- Paper submitted to STOTEN

Dose calculation – humans vs. biota

$$Dose_{ij}^{imm} = C_j \left(dcf_j^{skin} + a \times dcf_j^{imm} (1 - f_i^{ind} (1 - b)) \right) \times f_i^{occ} h_y$$

- $Dose_{ij}^{imm}$ - Plume immersion dose to age group i , nuclide j ($Sv\ y^{-1}$).
- dcf_j^{skin} - Skin dose from unit air concentration, nuclide j ($Sv\ h^{-1}/Bq\ m^{-3}$).
- C_j - Air concentration ($Bq\ m^{-3}$).
- dcf_j^{imm} - Dose from unit air concentration, nuclide j ($Sv\ h^{-1}/Bq\ m^{-3}$).
- a - Multiplier to account for departure from semi-infinite plume.
- b - Multiplier to account for shielding from a building.
- f_i^{ind} - Fraction of time spent indoors during site occupancy.
- f_i^{occ} - Fraction of time spent at site (site occupancy).
- h_y - Number of hours in one year ($8760\ h\ y^{-1}$).

- This is a modified semi-infinite cloud model for close distances from source
- For biota we adapt this to use $a = 1$, $b = 0$, $f_i^{ind} = 0$, and $f_i^{occ} = 1$.

Implementation in EA R&D 128

- Introduction of two new radionuclides (^{41}Ar and ^{85}Kr);
- Inclusion of the ^{41}Ar and ^{85}Kr DCCs converted to $\mu\text{Gy h}^{-1}/\text{Bq kg}^{-1}$ of air;
- Simple modifications to the code necessary to accommodate the fact that ^{41}Ar and ^{85}Kr (like ^3H , ^{14}C , ^{32}P and ^{35}S , already in model) derive from a concentration in air rather than in soil, and
- Small changes in coding to include doses from immersion of organisms in air in the calculation.

	A	B	C	D	E	F	G	H	I	J	K	L	M	N	O
1					Concentration factors, organisms:air or organisms:soil										
2	Nuclide	Air or soil	Soil	Bacteria	Lichen	Tree	Shrub	Herb	Seed	Fungi	Caterpillar	Ant	Bee	Woodlouse	Earthworm
3		$\text{Bq m}^{-3}/\text{Bq kg}^{-1}$	See colour key	See colour key	See colour key	See colour key	See colour key	See colour key	See colour key	See colour key	See colour key	See colour key	See colour key	See colour key	See colour key
4															
5	^3H	0.00E+00	5.4E+01	5.4E+01	1.6E+02	1.1E+02	1.5E+02								
6	^{14}C	0.00E+00	1.9E+03	1.9E+03	3.8E+01	1.3E+03	4.2E+03								
7	^{32}P	0.00E+00													
8	^{35}S	0.00E+00	5.0E+01	5.0E+01	1.5E+02	1.5E+02	1.5E+02								
9	^{41}Ar	0.00E+00	1.0E-04	1.0E-04	0.0E+00	0.0E+00	0.0E+00								
10	^{85}Kr	0.00E+00	1.0E-04	1.0E-04	0.0E+00	0.0E+00	0.0E+00								
11	^{60}Co	0.00E+00													
12	^{90}Sr	0.00E+00	7.0E-01	7.0E-01		1.0E+00	1.7E-01								
13	^{106}Ru	0.00E+00			1.5E+00										
14	^{129}I	0.00E+00	7.0E-01	7.0E-01											
15	^{131}I	0.00E+00	7.0E-01	7.0E-01											
16	^{137}Cs	0.00E+00	7.0E-01	7.0E-01	7.7E-01	4.0E-02	1.6E-01								
17	^{226}Ra	0.00E+00	7.0E-01	7.0E-01	1.0E-01	1.1E-01	2.2E-01								
18	^{234}Th	0.00E+00													
19	^{238}U	0.00E+00	7.0E-01	7.0E-01		1.4E-01	2.0E-01								
20	^{239}Pu	0.00E+00	7.0E-01	7.0E-01	6.6E-01	3.7E-01	2.0E-01								
21	^{241}Am	0.00E+00	7.0E-01	7.0E-01		1.5E-01	1.0E-01								
22															
23	Habitat factors:														
24	f soil		1.0E+00	0.0E+00	1.0E+00	1.0E+00	1.0E+00								
25	f soil surface		0.0E+00	1.0E+00	0.0E+00	0.0E+00	0.0E+00								
26	f air		0.0E+00	0.0E+00	5.0E-01	5.0E-01	5.0E-01								
27															
28	Radiation weighting factors:														
29															
30	Low energy beta	3.0E+00													
31	beta and photon	1.0E+00													
32	Alpha	2.0E+01													
33															
34															

Values in Bq m^{-3} of air:
Values in Bq kg^{-1} (dw) of soil:
Values in Bq kg^{-1} (fw) of organisms:
Values in Bq kg^{-1} (tw) of organisms:



- Internal dose negligible: default Ar and Kr CRs for all organisms set to 0.
- Although no deposition, some migration into soil pores possible leading to a transfer factor.
 - Assume pore air is at the same concentration as ground level air concentrations
 - assume a free air space of 0.15 v:v, bulk density for soil of 1500 kg m⁻³, so free air space = $0.15/1500 = 10^{-4} \text{ m}^3 \text{ kg}^{-1}$. Thus $\text{Bq m}^{-3}(\text{air}) \times 10^{-4} = \text{Bq kg}^{-1} (\text{wet soil})$.
 - A TF of 10^{-4} is therefore specified as a default for air (Bq m⁻³) to soil (Bq kg⁻¹ wet).

- For plants and fungi occupancy factors are changed to 1.0 soil, 0.5 air (instead of 0).
- Soil, bacteria and earthworms are assumed to reside only in the subsurface soil and are exposed only to ^{41}Ar and ^{85}Kr in the air pore spaces.
- External DCCs for fungi are those calculated for bacteria (i.e. infinite medium DCCs). Internal DCCs are those calculated for the dimensions of the fruiting body.

Dose calculation formulae

- The R&D 128 spreadsheet uses the following formulae for all radionuclides whose concentration is referenced to air: ^3H , ^{14}C , ^{32}P , ^{35}S , ^{41}Ar and ^{85}Kr .

$$(\text{Soil conc})_{\text{nuclide}} = (\text{Air conc, Bq m}^{-3})_{\text{nuclide}} \times CR_{\text{nuclide}}^{\text{soil}}$$

$$(\text{Air conc, Bq kg}^{-1})_{\text{nuclide}} = (\text{Air conc, Bq m}^{-3})_{\text{nuclide}} / 1.2$$

$$(\text{Internal dose})_{\text{nuclide, organism}} = (\text{Air conc})_{\text{nuclide}} \times CR_{\text{nuclide}}^{\text{organism}} \times DCC_{\text{nuclide, organism}}^{\text{internal}}$$

$$(\text{External dose})_{\text{nuclide, organism}} = (\text{Soil dose} + \text{Immersion dose})$$

$$(\text{Soil dose}) = DCC_{\text{nuclide, organism}}^{\text{external}} \times \left[(\text{Soil conc})_{\text{nuclide}} \times \left(\frac{f_{\text{soil organism}} + f_{\text{soilsur organism}}}{2} + f_{\text{air organism}} \times (\text{reduction factor})_{\text{radiation type}} \right) \right]$$

$$(\text{Immersion dose}) = DCC_{\text{nuclide, organism}}^{\text{external}} \times (\text{Air conc, Bq kg}^{-1})_{\text{nuclide}} \times \left(\frac{f_{\text{air organism}} + f_{\text{soilsur organism}}}{2} \right)$$

- Where the reduction factor is the modifier for dose to organisms in air is received from exposure to soil: 0 for α and low-energy β radiation and 0.25 for high energy $\beta+\gamma$ radiation.

Comparison with human dosimetry (ICRP 72)

Source	Description	DCC (mSv h ⁻¹ per Bq m ⁻³)	
		⁴¹ Ar	⁸⁵ Kr
Human dosimetry	External γ DCF ¹	2.20E-04	4.73E-07
	Skin (β) DCF ²	8.69E-07	4.44E-07
	Total ($\beta+\gamma$) DCF	2.21E-04	9.17E-07
Monte Carlo calculation	External DCC ³	2.03E-04	3.89E-07

- ¹Data from ICRP 72 (1996). ²Data from Simmonds *et al.* (1995).
³Geometry-corrected as $\frac{1}{2} \times$ external DCC (beta plus photon) values for the "human geometry" above.
- ICRP 72 ⁸⁵Kr immersion dose rate is 9.2×10^{-13} Sv h⁻¹ per unit air conc.
- This is an effective dose (includes skin dose with a weighted by 1%).
From the EU methodology (Simmonds *et al.* 1995), the weighted beta dose rate to skin is given as 4.44×10^{-13} Sv h⁻¹.
- Assuming that only the skin receives beta dose, the dose rate due to gamma is therefore $9.17 \times 10^{-13} - 4.4 \times 10^{-13} = 4.73 \times 10^{-13}$ Sv h⁻¹.

Conceptual difference human vs. biota

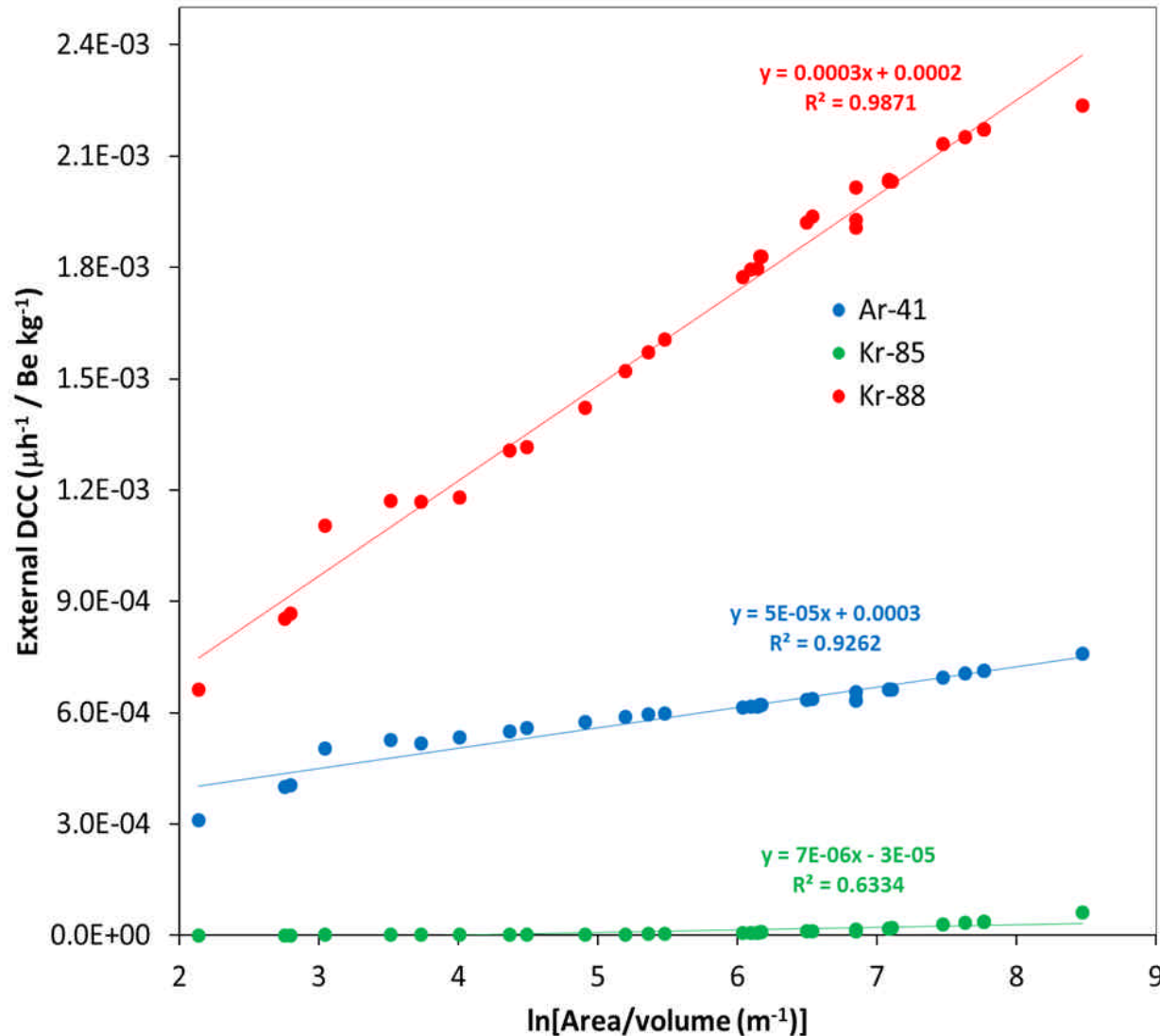
- Human DCFs are calculated for a plume emitting "from above", i.e., over a 2π (semi-infinite) geometry.
- Biota DCCs, are calculated assuming that the medium envelops the organism (infinite) geometry.
- This is not an inconsistency in so far as it is recognised that the human DCFs and biota DCCs refer to two different geometry definitions.
- Having corrected biota DCCs by a factor of 0.5, we should obtain identical results for ^{41}Ar and about a half different for ^{85}Kr .

Comparison results

- For ^{41}Ar , only 0.25% of the total biota DCC arises from β -radiation.
- For Kr, the β - component is significantly higher.
- For γ -rays, both methods (DCF and DCC) give the same result for Kr.
- However, for β -radiation, the DCC method undershoots for Kr.
- Explanation: The DCC method averages the external dose over the whole volume, whereas the DCF method averages over an outer layer.
- The averaging method makes little difference for small organisms and significant difference for large organisms like a human.
- Hence, the "1% of skin dose" component of the human DCF ($4.7 \times 10^{-5} \mu\text{Sv h}^{-1}$ per Bq m^{-3}) should resemble the DCC for a very small organism (divided by geometry factor of 2).
- Examination of a suitable small organism ("germinating seed") confirms this. β DCC = $6.1 \times 10^{-5} / 2 = 3.1 \times 10^{-5} \mu\text{Sv h}^{-1}$ per Bq m^{-3} .
- The two values are less than 35% different; this provides assurance that ignoring density differences has not introduced significant errors.

Relationship between DCC and organism size

- Select area/volume as the sizing variable.
- DCCs increase with larger area (more surface to absorb) and decrease with larger volume (averaging into larger organism).
- Predicted Ar and Kr DCCs of 2.6×10^{-4} and $6.4 \times 10^{-7} \mu\text{Gy h}^{-1} / \text{Bq m}^{-3}$, similar to explicitly calculated values.



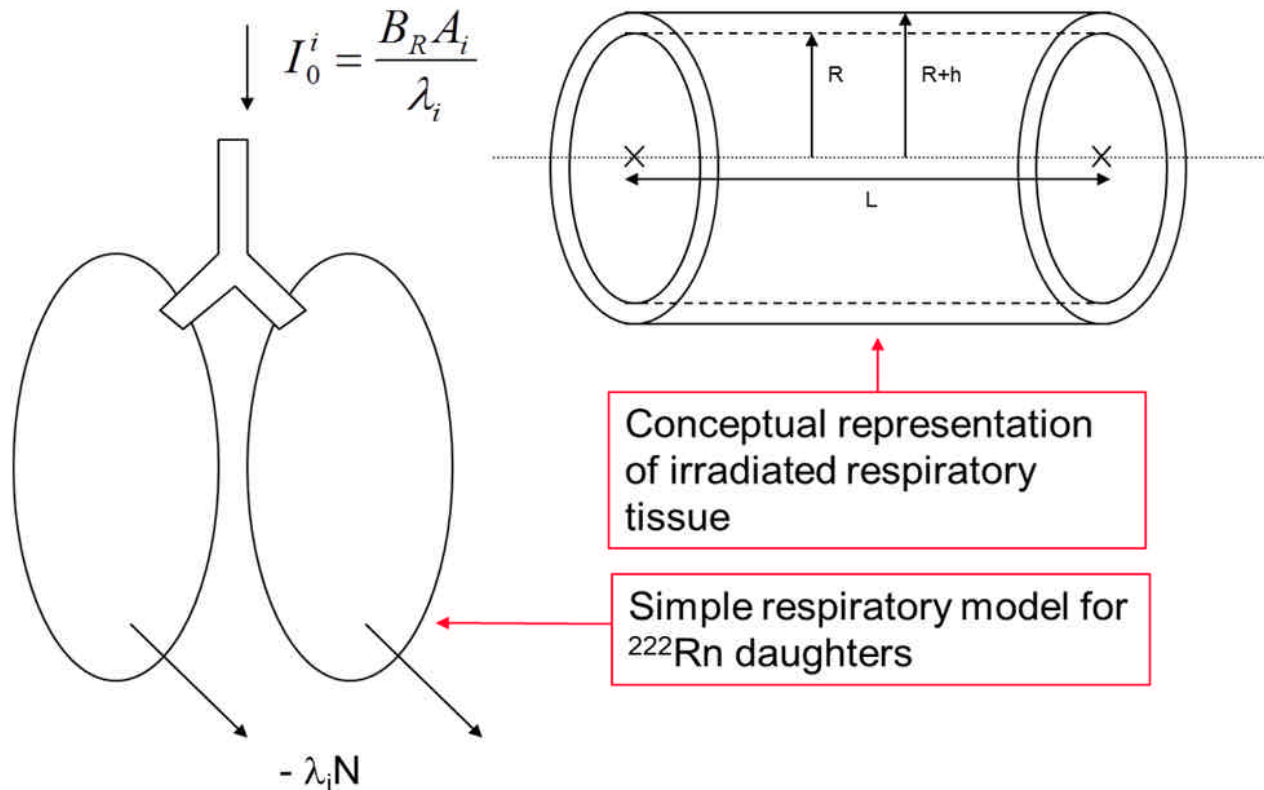
Using ^{85}Kr as an analogue for other Kr and Xe

- Previous statement that other Xe and Kr isotopes can be modelled using ^{85}Kr as a surrogate (Copplestone et al., 2010).
- We tested this assumption by calculating DCCs for $^{131\text{m}}\text{Xe}$, ^{133}Xe and $^{88}\text{Kr}(+^{88}\text{Rb})$ explicitly.
- The DCCs for ^{88}Kr can vary significantly with respect to ^{85}Kr : by a factor between 10 and 300 (β -radiation > 10 keV) and between 1200 and 1400 (γ -radiation).
- The corresponding factor ranges of variation for $^{131\text{m}}\text{Xe}$ and ^{133}Xe are 0.2 - 3800 and 4 - 20, respectively; similar for both radionuclides.
- The main sources of difference are therefore (a) the larger amount of γ -emissions of ^{88}Kr (52 vs. 1 for ^{85}Kr and 10 for the Xe isotopes), and (b) the stronger high-energy β -component of the Xe isotopes.
- The ^{85}Kr analogue approximation is therefore not valid for ^{88}Kr .
- For Xe, it is only valid for small organisms – not for birds, mammals and trees.

Radon – allometric model

Basis of the approach

- A model based on allometrically derived respiration rates and target tissue masses, designed for calculating ^{222}Rn daughter dose rates to sensitive tissues and the whole body of terrestrial animals and plants.



- Model the input of a constant flow of atoms into a compartment with continuous decay, with these two fluxes in equilibrium.
- Assume that the compartment is 100% efficient at trapping the material, i.e. no particles escape by exhalation and decay is the only source of removal.
- The input flow I_0^i equals the specific activity \times breathing rate / decay constant (in order to convert disintegrations per unit time to particles).

$$I_0^i = \frac{B_R A_i}{\lambda_i}$$

i : Index labelling the radionuclide: 1 to 5 for ^{222}Rn , ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po ;

A_i : Activity of radionuclide i [Bq m^{-3}] = A_1 (secular equilibrium)

BR : Breathing rate [$\text{m}^3 \text{s}^{-1}$] = tidal volume (V_T) \times breathing frequency (v_R)

λ_i : Decay constant of radionuclide i [s^{-1}].

- From here the DCC is:

$$DCC = \frac{\dot{D}}{A_{Rn}} = \frac{B_R D_P^a}{M_T} = 5.54 \times 10^{-9} \frac{B_R}{M_T}$$

- Where D_a^p is the potential α -energy per Bq activity of the short-lived radon daughters in secular equilibrium

- Many biological parameters relating to organism structure relate to metabolism and scale according to the Brody-Kleiber law:

$$Y = A \times M^b, \quad b = 0.75$$

- Other parameters scale on the basis of surface exchange, like radiation flux and heat transfer:

$$S \propto r^2 \text{ and } M \propto V \propto r^3 \Rightarrow S \propto M^{\frac{2}{3}} \text{ and } \frac{S}{V} \propto M^{-\frac{1}{3}}$$

- For this study we use the following relationships:

$$B_R(M) = A_{BR} M^{B_{BR}} = (8.7 \pm 4.4) \times 10^{-6} M^{0.76 \pm 0.02}$$

$$M_L(M) = A_{LM} M^{B_{LM}} = (1.28 \pm 0.72) \times 10^{-2} M^{1.02 \pm 0.03}$$

- M is the mass in kg and B_R is the ventilation rate in $\text{m}^3 \text{h}^{-1}$.

Breathing rate comparison using different allometric formulae

Organism	Reference	Measured	Predicted BR ($\text{m}^3 \text{s}^{-1}$)			
		BR ($\text{m}^3 \text{s}^{-1}$)	Ref. Man	DOE (1992)	Peters (1983)	This work
Man	Reference man	3.34E-04	3.33E-04	1.41E-04	1.88E-04	2.20E-04
Rat	Hofmann <i>et al.</i> (1992)	3.90E-06	5.58E-06	2.23E-06	2.76E-06	3.49E-06
Oryzomys	Drew & Eisenbud (1966)	6.67E-07	1.97E-06	7.78E-07	9.07E-07	1.22E-06

Base and exponent of the allometric formulae for ^{222}Rn daughter DPUCs (internal α irradiation)

Parameter	B	TB	L	WB
Base A	5.14E-04	5.55E-05	3.77E-06	4.83E-08
Exponent B	9.63E-02	9.63E-02	-2.57E-01	-2.37E-01

- Simple power functions for DPCCs in $\mu\text{Gy h}^{-1}$ per Bq m^{-3} :

$$DCC_B = F_U R_{WF}^a \left(\frac{D_P^a A_{BR}}{r_T h_T S_B^{RM}} M_{RM}^{2/3} \right) M^{B_{BR} - \frac{2}{3}}$$

$$DCC_{TB} = F_U R_{WF}^a \left(\frac{D_P^a A_{BR}}{r_T h_T S_{TB}^{RM}} M_{RM}^{2/3} \right) M^{B_{BR} - \frac{2}{3}}$$

$$DCC_L = F_U R_{WF}^a \left(\frac{D_P^a A_{BR}}{A_{LM}} \right) M^{B_{BR} - B_{LM}}$$

$$DCC_{WB} = F_U R_{WF}^a (D_P^a A_{BR}) M^{B_{BR} - 1}$$

This approach is only recommended for mammals. Applicability to other animals with structurally simpler respiratory systems (birds, reptiles, amphibians and insects) is conjectural and likely over-conservative.

F_U : Unit conversion factor ($3.6 \times 10^9 \mu\text{Gy h}^{-1}$ per Gy s^{-1});

BR: Gross extrapolation to the bronchial epithelium (airway generations 1 - 8);

TB: Full tracheobronchial epithelium (generations 1 - 15); L: Full lung; WB: Whole body;

$A_{BR}(A_{LM})$, $B_{BR}(B_{LM})$: Base and exponent of the allometric formulae for breathing rate & lung mass;

S_{TB}^{RM} and S_B^{RM} : surface area of the tracheobronchial tree or the bronchial epithelium;

R_{wf}^α : Radiation weighting factor for α -energy (default = 20).

- The rate of resource use in plants = $A \times M^{3/4}$, though isometric respiration rates have also been suggested.
- We calculated a breathing rate relationship for plants from whole plant respiration:
 - Use respiration rate (net CO₂ efflux in nmol CO₂ s⁻¹) = $1.19 \times M^{1.02}$ from Reich et al. (2005).
 - Apply conversion factor of 2.5×10^3 mols of air per mols of CO₂
 - Apply a generic wet: dry mass ratio of 5 and a molar volume of 22.4 l STP.
 - This is the largest potential source of uncertainty in this calculation.

$$BR_{PLANT} (m^3 s^{-1}) = 1.95 \times 10^{-4} M (kg)^{1.02} = A_{PL} M^{B_{PL}}$$

- Justification: dry air contains approximately 0.04% carbon dioxide. The partial pressure of carbon dioxide in dry air at sea level is, therefore, 4×10^{-4} Pa. One mol of air will have 4×10^{-4} mols of CO₂ in it; hence there are $1 / 4 \times 10^{-4} = 2.5 \times 10^3$ mols of air per mol of CO₂.

- Ellipsoid with axes L, a, a : $V_{\text{ellipsoid}} = \frac{1}{6} \pi L a^2$
- 'Equivalent' cylinder radius: $R = \frac{a}{\sqrt{6}}$
- The target tissue is the space between the two interlocking cylinders of radii R and $R + h_T$ and length L , with mass:

$$m_T \approx \sqrt{\frac{2}{3}} \pi R a L h_T = 2\sqrt{6} \frac{h_T}{a} M$$

- Where M is the total mass of the organism.
- Assume that assume that the whole plant is a surface exchanging gases with the atmosphere.
- Give doses a factor of < 5 of what would have been obtained using the allometric formulae for animals.

- Simple power functions for DPCCs in $\mu\text{Gy h}^{-1}$ per Bq m^{-3} :

$$DCC_{PLANT\ TISSUE} = F_U R_{WF}^a D_P^a \frac{A_{PL} a M^{B_{PL}-1}}{2\sqrt{6}h_T}$$

$$DCC_{WHOLE\ PLANT} = F_U R_{WF}^a D_P^a A_{PL} M^{B_{PL}-1}$$

D_P^a : Potential α -energy factor $5.54 \times 10^{-9} \text{ J Bq}^{-1}$;

A_{PL} : Allometric base for breathing rate in plants, $1.95 \times 10^{-4} \text{ m}^3 \text{ s}^{-1}$ calculated by Vives i Batlle et al. (2012) based on previous data (Reich et al., 2005) and based on net CO_2 efflux data;

a : Minor axis of the ellipsoid representing the plant in m (if the two minor axes of the geometry are dissimilar then the average is taken);

h_T : Depth of sensitive tissue = $5.5 \times 10^{-5} \text{ m}$;

F_U : Unit conversion factor ($3.6 \times 10^9 \mu\text{Gy h}^{-1}$ per Gy s^{-1});

R_{wf}^a : Radiation weighting factor for α -energy (default = 10).

Dose calculation formulae (again)

- Applicable to all radionuclides whose concentration is referenced to air - that is, ^3H , ^{14}C , ^{32}P , ^{35}S , ^{41}Ar , ^{85}Kr and ^{222}Rn :

$$(\text{Internal dose, } \mathbf{mGy} \text{ h}^{-1})_{\text{nuclide, organism}} = (\text{Air conc, } \text{Bq m}^{-3})_{\text{nuclide}} \times \\ \times (DCC, \mathbf{mGy} \text{ Bq}^{-1} \text{ h}^{-1} \text{ m}^3)_{\text{nuclide, organism}}^{\text{internal}}$$

$$(\text{External dose})_{\text{nuclide, organism}} = (\text{Soil dose} + \text{Immersion dose})$$

$$(\text{Soil dose}) = (\text{Air conc, } \text{Bq m}^{-3})_{\text{nuclide}} \times (CR, \text{m}^3 \text{ kg}^{-1})_{\text{nuclide}}^{\text{soil}} \times \\ \times (DCC, \mathbf{mGy} \text{ Bq}^{-1} \text{ h}^{-1} \text{ m}^3)_{\text{nuclide, organism}}^{\text{external}} \times \\ 1.2 \text{ kg m}^{-3} \times \left[\left(\frac{f_{\text{soil}_{\text{organism}}} + f_{\text{soilsur}_{\text{organism}}}}{2} \right) \right. \\ \left. + f_{\text{air}_{\text{organism}}} \times (\text{reduction factor})_{\text{radiation type}} \right]$$

$$(\text{Immersion dose}) = (\text{Air conc, } \text{Bq m}^{-3})_{\text{nuclide}} \times (DCC, \mathbf{mGy} \text{ Bq}^{-1} \text{ h}^{-1} \text{ m}^3)_{\text{nuclide, organism}}^{\text{external}} \times \\ \times (f_{\text{air}_{\text{organism}}} + f_{\text{soilsur}_{\text{organism}}} / 2)$$

- Where the reduction factor is the modifier for dose to organisms in air is received from exposure to soil: 0 for α and low-energy β radiation and 0.25 for high energy β + γ radiation

Internal DCCs for Rn (ERICA organisms)

Animals

Organism	M (kg)	a (m)	b (m)	c (m)	f (m ³ s ⁻¹)	DCC _B	DCC _{TB}	DCC _L ^a	DCC _{WB}
Amphibian (ICRP Frog)	3.1E-02	8.0E-02	3.0E-02	2.5E-02	5.9E-07	1.3E+01	1.4E+00	3.1E-01	3.7E-03
Reptile (FASSET snake)	7.4E-01	1.2E+00	3.5E-02	3.5E-02	6.3E-06	1.6E+01	1.7E+00	1.3E-01	1.7E-03
Mammal (ICRP Rat)	3.1E-01	2.0E-01	6.0E-02	5.0E-02	3.2E-06	1.5E+01	1.6E+00	1.6E-01	2.1E-03
Mammal (ICRP Deer)	2.5E+02	1.3E+00	6.0E-01	6.0E-01	6.9E-04	3.8E+01	4.1E+00	4.0E-02	5.6E-04
Bird (ICRP Duck)	1.3E+00	3.0E-01	1.0E-01	8.0E-02	9.4E-06	1.7E+01	1.9E+00	1.2E-01	1.5E-03
Mammal (FASSET Marine)	1.8E+02	1.8E+00	4.4E-01	4.4E-01	5.4E-04	3.6E+01	3.8E+00	4.2E-02	5.9E-04
Reptile (ICRP Marine Turtle)	1.4E+02	8.5E-01	3.9E-01	8.0E-01	4.3E-04	3.4E+01	3.7E+00	4.4E-02	6.2E-04
Mammal (FASSET Freshw.)	3.9E+00	3.3E-01	1.5E-01	1.5E-01	2.3E-05	2.0E+01	2.1E+00	8.9E-02	1.2E-03

Plants

Organism	M (kg)	a (m)	b (m)	c (m)	f (m ³ s ⁻¹)	DCC _{TISS}	DCC _{WB}
Lichen & bryophytes (ICRP Bryophyte)	1.1E-04	4.0E-02	2.3E-03	2.3E-03	1.8E-08	2.8E-01	3.2E-02
Grasses & Herbs (ICRP Wild grass)	2.6E-03	5.0E-02	1.0E-02	1.0E-02	4.5E-07	1.3E+00	3.5E-02
Tree (ICRP Pine tree)	4.7E+02	1.0E+01	3.0E-01	3.0E-01	1.0E-01	4.9E+01	4.4E-02

External DCCs for Rn (ERICA organisms)

Organism	DCC ext (mGy h ⁻¹ per Bq m ⁻³)		
	b < 10 keV	b > 10 keV + g	a
Amphibian (ICRP Frog)	4.6E-11	7.8E-04	0.0E+00
Reptile (FASSET snake)	3.3E-11	7.6E-04	0.0E+00
Mammal (ICRP Rat)	4.2E-11	7.3E-04	0.0E+00
Mammal (ICRP Deer)	4.0E-12	3.8E-04	0.0E+00
Bird (ICRP Duck)	2.6E-11	6.9E-04	0.0E+00
Mammal (FASSET Marine)	4.0E-13	4.3E-04	0.0E+00
Reptile (ICRP Marine Turtle)	9.3E-13	4.2E-04	0.0E+00
Mammal (FASSET Freshwater)	3.5E-12	6.4E-04	0.0E+00
Lichen & bryophytes (ICRP Bryophyte)	1.2E-09	9.9E-04	0.0E+00
Grasses & Herbs (ICRP Wild grass)	1.7E-10	8.5E-04	0.0E+00
Tree (ICRP Pine tree)	3.6E-12	5.1E-04	0.0E+00

Model validation with data from MacDonald and Laverock (1998)

Organism	Mass (kg)	DCC (mGy Bq ⁻¹ s ⁻¹ m ³)				Dose rate (mGy h ⁻¹)		% diff.
		B	TB	L	WB	Calculated	From paper	
Mole	4.00E-02	3.77E-04	1.85E-05	8.63E-06	1.04E-07	596	451	32
Pocket gopher	2.00E-01	4.41E-04	2.05E-05	5.71E-06	7.07E-08	854	702	22
Ground squirrel	5.00E-01	4.81E-04	2.17E-05	4.51E-06	5.69E-08	311	268	16
Ground hog	3.00E+00	5.72E-04	2.44E-05	2.85E-06	3.72E-08	132	125	6
Badger	8.00E+00	6.29E-04	2.59E-05	2.21E-06	2.95E-08	90	89	1

Additional comparison with Hofmann *et al.* (2006) and Harley (1988)

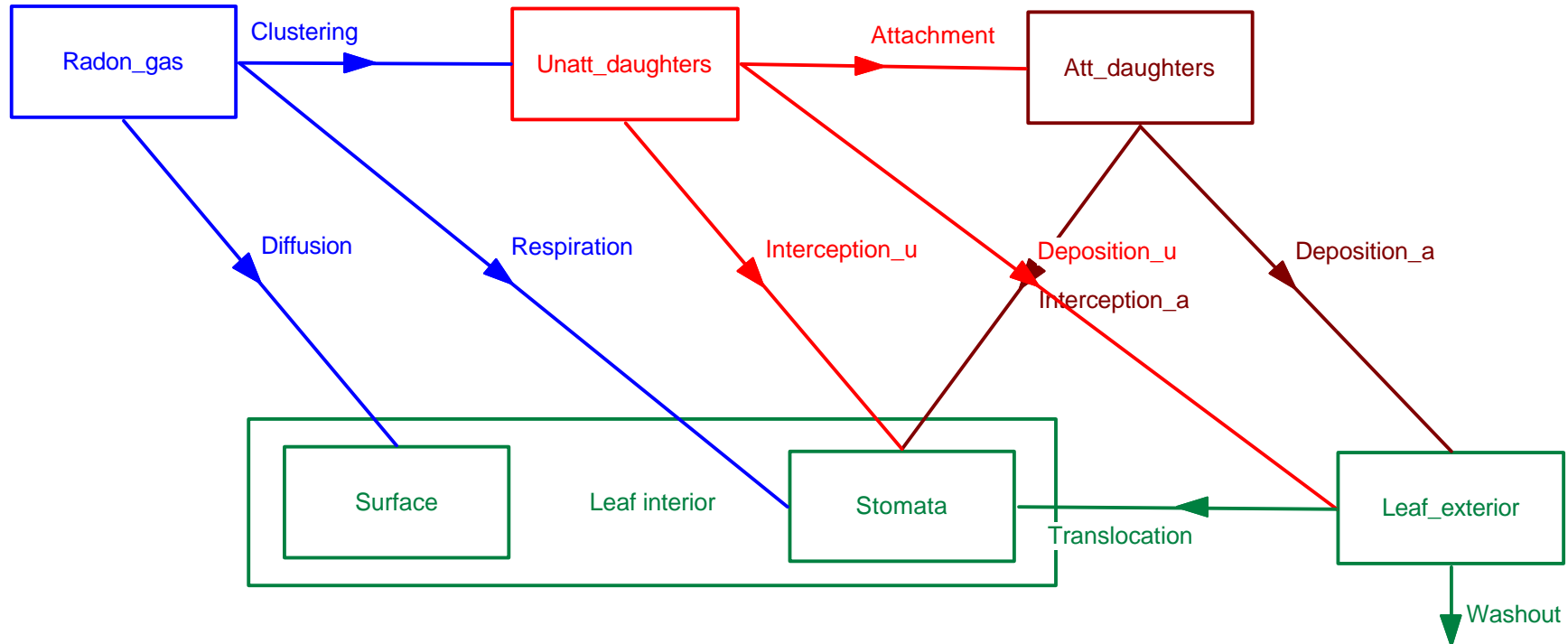
Organism	M (kg)	Source	DCC (nGy Bq ⁻¹ h ⁻¹ m ³) ^a	
			Reported	Calculated
Rat	0.3	Hofmann <i>et al.</i> (2006)	13.5 ± 12.5	76
Rats	0.35	Harley (1988)	10.3 ± 2.5	76

^aUsing the conversion 1 WLM = 6.3 × 10⁵ Bq h m⁻³ (ICRP, 1978)

- Good agreement with McDonald and Laverock (1998).
- Additional comparison with rat DCCs for the tracheobronchial tree by is problematic as reported sources they use a full respiratory model:
 - Predicting significant fractions of the radon daughters removed by the nasal passages.
 - Including lung clearance processes, resulting in transport from the alveolar region to the bronchial area, with associated decay included in transit.
 - The models consider atmospheres with various assumptions of equilibrium resulting in varying particle size, $F < 1$ and f_p values.
 - As a result, ours is a conservative approximation.

Radon – advanced plant model

- We developed (using ModelMaker) a compartment model representing:
 - Aerosol: free, unattached and attached fractions of ^{222}Rn , ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po
 - Plant uptake: surface interception of unattached and attached daughters, diffusion of radon through stomata, permeation of radon through plant epidermis.
 - Plant turnover: translocation of deposited activity from plant surface to plant interior
- We derived DCCs for internal, surface and external exposure as a function of plant surface area and steady-state concentration at ground level.



- Each sub-model contains the decay chain of radon: $^{222}\text{Rn} \Rightarrow ^{218}\text{Po} \Rightarrow ^{214}\text{Pb} \Rightarrow ^{214}\text{Bi} \Rightarrow ^{214}\text{Po}$.
- Exchange rates link individual compartments across sub-models, with rate constants linked to the parameter set.

- ^{222}Rn diffusion via stomata and permeation via epidermis (Free fraction to plant interior sub-models).
- ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po interception through stomata (Unattached and attached fractions to plant interior sub-models).
- ^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po aerosol deposition (Unattached and attached fractions to plant surface sub-models).
- Translocation of deposited ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po (plant surface to plant interior)

Basic equations and parameters

Deposition flux: $f = A_{Rn} (Bq m^{-3}) \times \frac{1}{I (s^{-1})} \times v_d (m s^{-1})$

Deposition rate: $= \frac{v_d S_A}{V_G}$

Deposition velocity:

$$v_d^{unatt} = A_{unatt} e^{I_L B_{unatt}} \quad A_{unatt} = 2.1 \times 10^{-3} m.s^{-1}, B_{unatt} = 1.04, r^2 = 0.998$$

$$v_d^{att} = A_{att} e^{I_L B_{att}} \quad A_{att} = 1.89 \times 10^{-5} m.s^{-1}, B_{att} = 1.18, r^2 = 0.997$$

Diffusion through stomata: $-\frac{dN}{dt} = \frac{DA}{l_s} (c_{out} - c_{in})$ with D the diffusion coefficient

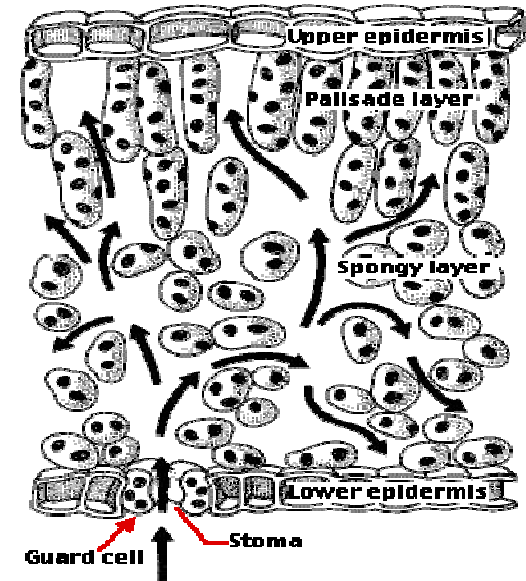
Permeation through leaf walls: $\frac{dN}{dt} = \frac{-KS}{w} (c_{in} - c_{out})$ with K the permeability

Transfer factor: $TF = \frac{a_i^{int}}{a_{Rn}^{ext}} = a_i^{int} \left(\frac{N_{Rn}^{ext}}{hS_A} \right)^{-1}$

Dosimetry:

$$H_i^j \Big|_{int} = a_i^{int} E_i AF_i(E_i) \equiv a_i^{int} DCC_i^{int}; H_i^j \Big|_{ext} = \frac{a_i^{ext}}{r_{air}} E_i (1 - AF_i(E_i)) \equiv \frac{a_i^{ext}}{r_{air}} DCC_i^{ext}$$

Parameter	Value
Duration of day	$8.6 \times 10^4 \text{ s}$
Activity of ^{222}Rn in atmosphere, A_{Rn}	1 Bq m^{-3}
Air column surface area, S_A	1 m^2
Height of radon mixing layer above soil, h	2 m
Air density, ρ_{air}	1.2 kg m^{-3}
Radon diffusion coefficient, D	$1.1 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$
Clustering rate, r_{clust}	2.3 s^{-1}
Attachment rate, r_{att}	$2 \times 10^{-2} \text{ s}^{-1}$
Internal exposure DCCs for ^{222}Rn , ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po (total, unweighted)	$3.2 \times 10^{-3}, 3.5 \times 10^{-3}, 7.9 \times 10^{-5}, 6.6 \times 10^{-5}, 4.4 \times 10^{-3} \mu\text{Gy h}^{-1} / \text{Bq kg}^{-1}$
Surface deposition DCCs for ^{222}Rn , ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po (total, unweighted)	$3.2 \times 10^{-3}, 3.5 \times 10^{-3}, 7.9 \times 10^{-5}, 6.6 \times 10^{-5}, 4.4 \times 10^{-3} \mu\text{Gy h}^{-1} / \text{Bq kg}^{-1}$
External exposure DCCs for ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po (total, unweighted)	$1.7 \times 10^{-3}, 8.1 \times 10^{-5}, 7.9 \times 10^{-5}, 2.2 \times 10^{-3} \mu\text{Gy h}^{-1} / \text{Bq kg}^{-1}$
Decay constants for ^{222}Rn , ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po , λ_i	$2.1 \times 10^{-6}, 3.7 \times 10^{-3}, 4.3 \times 10^{-4}, 5.9 \times 10^{-4}, 4.2 \times 10^3 \text{ s}^{-1}$
Potential alpha energy per unit activity weighting factors for ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po	$1.05 \times 10^{-1}, 5.16 \times 10^{-1}, 3.79 \times 10^{-1}, 6 \times 10^{-8}$
Permeability constant for Rn in plant epidermis, K	$1.25 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$
Leaf area index, I_L	2
Porosity of substomatal cavity, g	0.35
Leaf thickness, L	$2.5 \times 10^{-4} \text{ m}$
Plant density, ρ_P	10^3 kg m^{-3}
Stomata density, n_s	$3 \times 10^8 \text{ m}^{-2}$
Width of epidermis \approx stomatal length, l_s	$2.5 \times 10^{-5} \text{ m}$
Stomata surface area (maximum)	$4.9 \times 10^{-10} \text{ m}^2$
Exponential fit parameters for unattached deposition velocity, $A_{\text{unatt}}, B_{\text{unatt}}$	$2.1 \times 10^{-3} \text{ m s}^{-1}$
Exponential fit parameters for attached deposition velocity, $A_{\text{att}}, B_{\text{att}}$	1.04
	$1.89 \times 10^{-5} \text{ m s}^{-1}$
	1.18

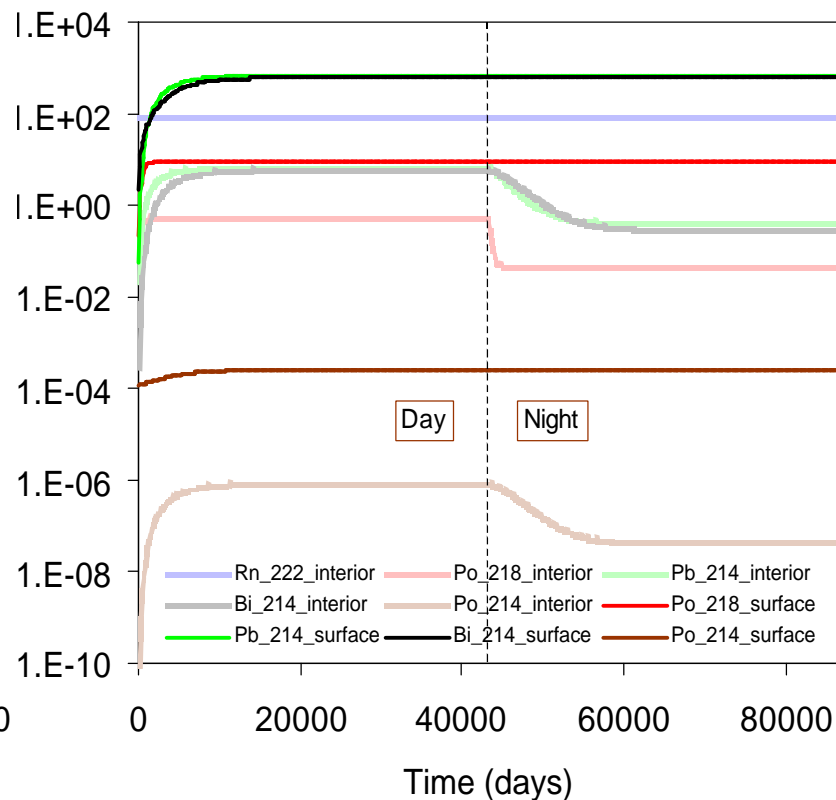
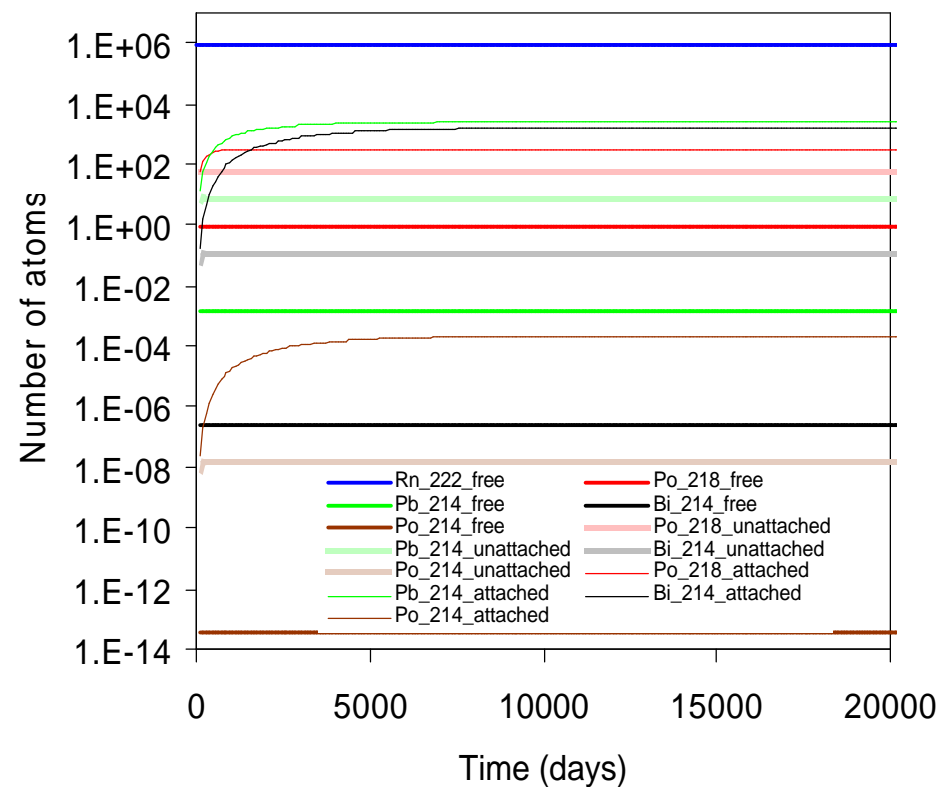


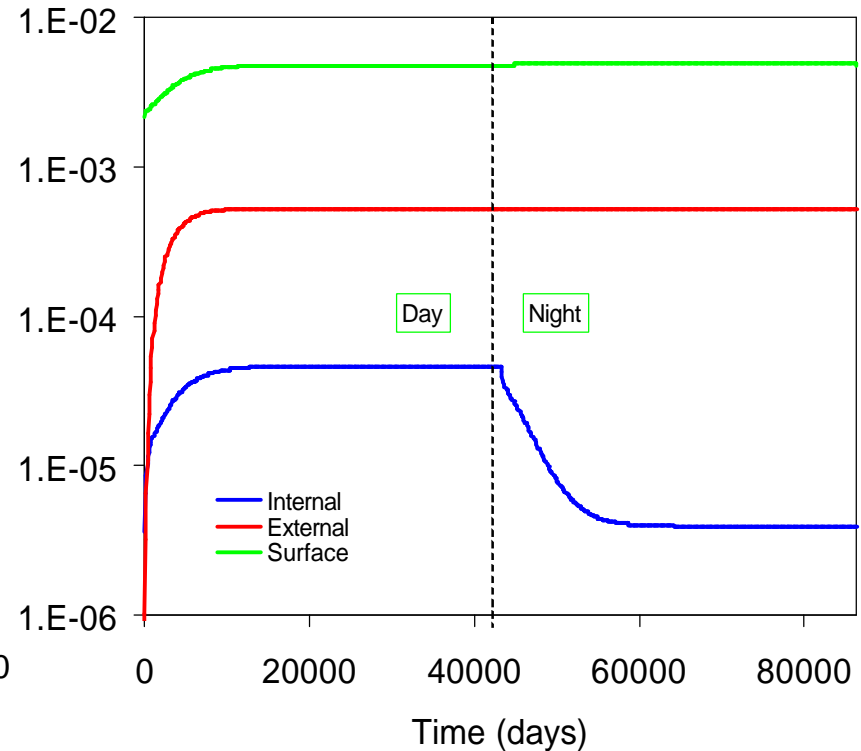
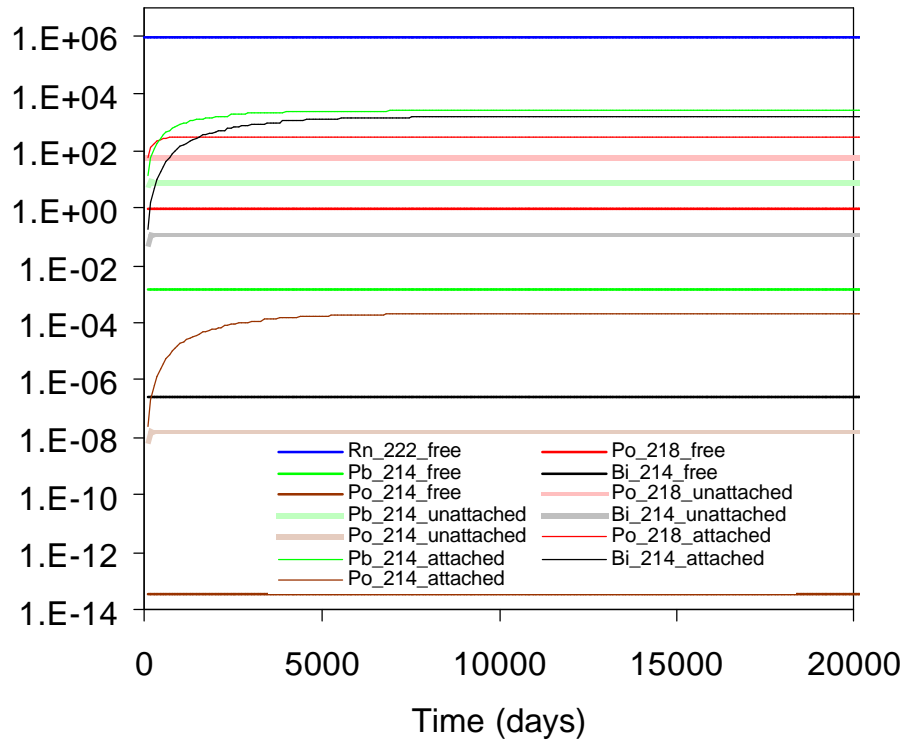
Deposition velocity values for different types of surface

Surface	l_L	v_d^U (m s ⁻¹)	v_d^A (m s ⁻¹)
Soil	0	2×10^{-3}	2×10^{-5}
Grass	2.5	3.20×10^{-2}	3.10×10^{-4}
Wheat	4.2	1.57×10^{-1}	2.90×10^{-3}

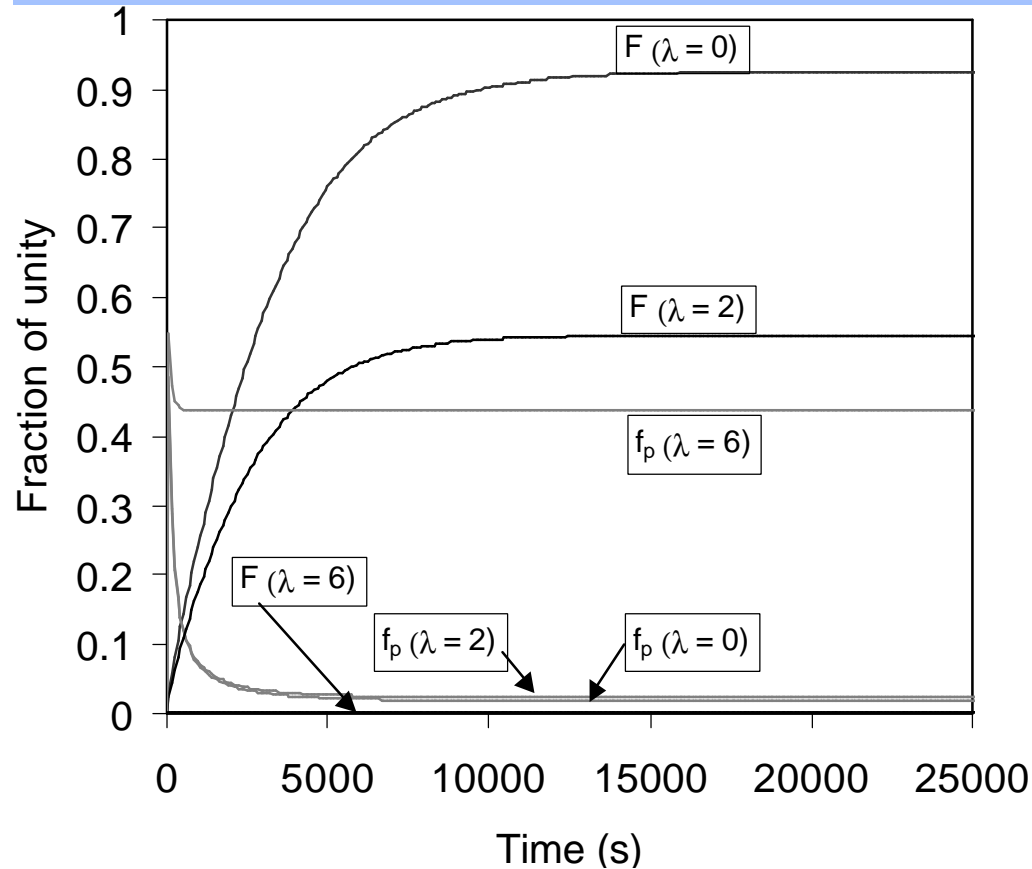
Examples of (perennial) plant leaf geometries covering a wide size range

Leaf type	Major axis a (m)	Major axis b (m)	Minor axis c (m)	Area (m ²)	Volume (m ³)	Mass (kg)
Tiny	1.5×10^{-2}	5.0×10^{-3}	2.5×10^{-4}	1.2×10^{-4}	9.8×10^{-9}	9.8×10^{-6}
Small	6.0×10^{-2}	3.5×10^{-2}	2.5×10^{-4}	3.3×10^{-3}	2.8×10^{-7}	2.8×10^{-4}
Medium	1.3×10^{-1}	5.5×10^{-2}	2.5×10^{-4}	1.1×10^{-2}	9.4×10^{-7}	9.4×10^{-4}
large	1.2×10^{-1}	1.1×10^{-1}	2.5×10^{-4}	2.0×10^{-2}	1.7×10^{-6}	1.7×10^{-3}





Equilibrium factor F and f_p



$$F = \frac{C_{Eq}}{C_0} = \frac{0.105 A(^{218}Po) + 0.516 A(^{214}Pb) + 0.379 A(^{214}Bi) + 6 \times 10^{-8} A(^{214}Po)}{A(^{222}Rn)}$$

$$f_p = \frac{0.105 A_{unatt}(^{218}Po) + 0.516 A_{unatt}(^{214}Pb) + 0.379 A_{unatt}(^{214}Bi) + 6 \times 10^{-8} A_{unatt}(^{214}Po)}{A(^{222}Rn) \times F}$$

DCCs for plant leaf geometries

Internal DCC (total), mGy h ⁻¹ /Bq kg ⁻¹						
Nuclide	Tiny leaf	Small leaf	Medium leaf	Large leaf	Average	± S.D.
²²² Rn	3.2E-03	3.2E-03	3.2E-03	3.2E-03	3.2E-03	1.6E-10
²¹⁸ Po	3.5E-03	3.5E-03	3.5E-03	3.5E-03	3.5E-03	0.0E+00
²¹⁴ Pb	7.6E-05	7.9E-05	8.0E-05	8.0E-05	7.9E-05	1.7E-06
²¹⁴ Bi	5.8E-05	6.8E-05	6.9E-05	6.9E-05	6.6E-05	5.1E-06
²¹⁴ Po	4.4E-03	4.4E-03	4.4E-03	4.4E-03	4.4E-03	0.0E+00
Surface DCC (total), mGy h ⁻¹ /Bq kg ⁻¹						
²²² Rn	1.6E-03	1.6E-03	1.6E-03	1.6E-03	1.6E-03	1.4E-09
²¹⁸ Po	1.7E-03	1.7E-03	1.7E-03	1.7E-03	1.7E-03	3.3E-09
²¹⁴ Pb	3.2E-06	1.1E-04	1.1E-04	1.1E-04	8.1E-05	5.2E-05
²¹⁴ Bi	5.0E-07	1.0E-04	1.0E-04	1.1E-04	7.9E-05	5.2E-05
²¹⁴ Po	2.2E-03	2.2E-03	2.2E-03	2.2E-03	2.2E-03	9.5E-11
External DCC (total), mGy h ⁻¹ /Bq kg ⁻¹						
²²² Rn	2.3E-07	2.3E-07	2.3E-07	2.3E-07	2.3E-07	1.6E-10
²¹⁸ Po	7.0E-09	7.0E-09	7.0E-09	7.0E-09	7.0E-09	3.2E-11
²¹⁴ Pb	2.4E-04	2.3E-04	2.3E-04	2.3E-04	2.3E-04	1.7E-06
²¹⁴ Bi	1.2E-03	1.1E-03	1.1E-03	1.1E-03	1.1E-03	5.1E-06
²¹⁴ Po	4.8E-08	4.8E-08	4.8E-08	4.8E-08	4.8E-08	1.9E-11

Transfer factors ($\text{m}^3 \text{kg}^{-1}$) for radon products in plants

Transfer factor type	Time	^{218}Po	^{214}Pb	^{214}Bi	^{214}Po
Plant surface	Day	6.68E-02	5.82E-01	7.18E-01	2.09E+00
	Night	6.70E-02	5.84E-01	7.20E-01	2.09E+00
Plant interior	Day	3.84E-03	5.89E-03	6.92E-03	6.92E-03
	Night	3.50E-04	3.50E-04	3.50E-04	3.50E-04

Comparison with allometric method

Modelling			DPURn (mGy h^{-1} per Bq m^{-3})				
approach	Organism	Time	Internal	Surface	Int + surf	External	Total
New model	Plant leaf	Night	3.9E-06	4.8E-03	4.8E-03	5.3E-04	5.3E-03
	Plant leaf	Day	4.6E-05	4.8E-03	<u>4.9E-03</u>	<u>5.3E-04</u>	<u>5.4E-03</u>
Allometric	Fungi	All	N/A	N/A	3.9E-03	8.9E-04	4.3E-03
	Herb	All	N/A	N/A	3.9E-03	1.1E-03	4.4E-03
	Lichen	All	N/A	N/A	3.9E-03	9.7E-04	4.4E-03
	Seed	All	N/A	N/A	3.9E-03	1.2E-03	4.5E-03
	Shrub	All	N/A	N/A	3.9E-03	1.1E-03	4.4E-03
	Tree	All	N/A	N/A	3.9E-03	1.1E-03	4.4E-03
	Average	All	N/A	N/A	<u>3.9E-03</u>	<u>1.0E-03</u>	<u>4.4E-03</u>

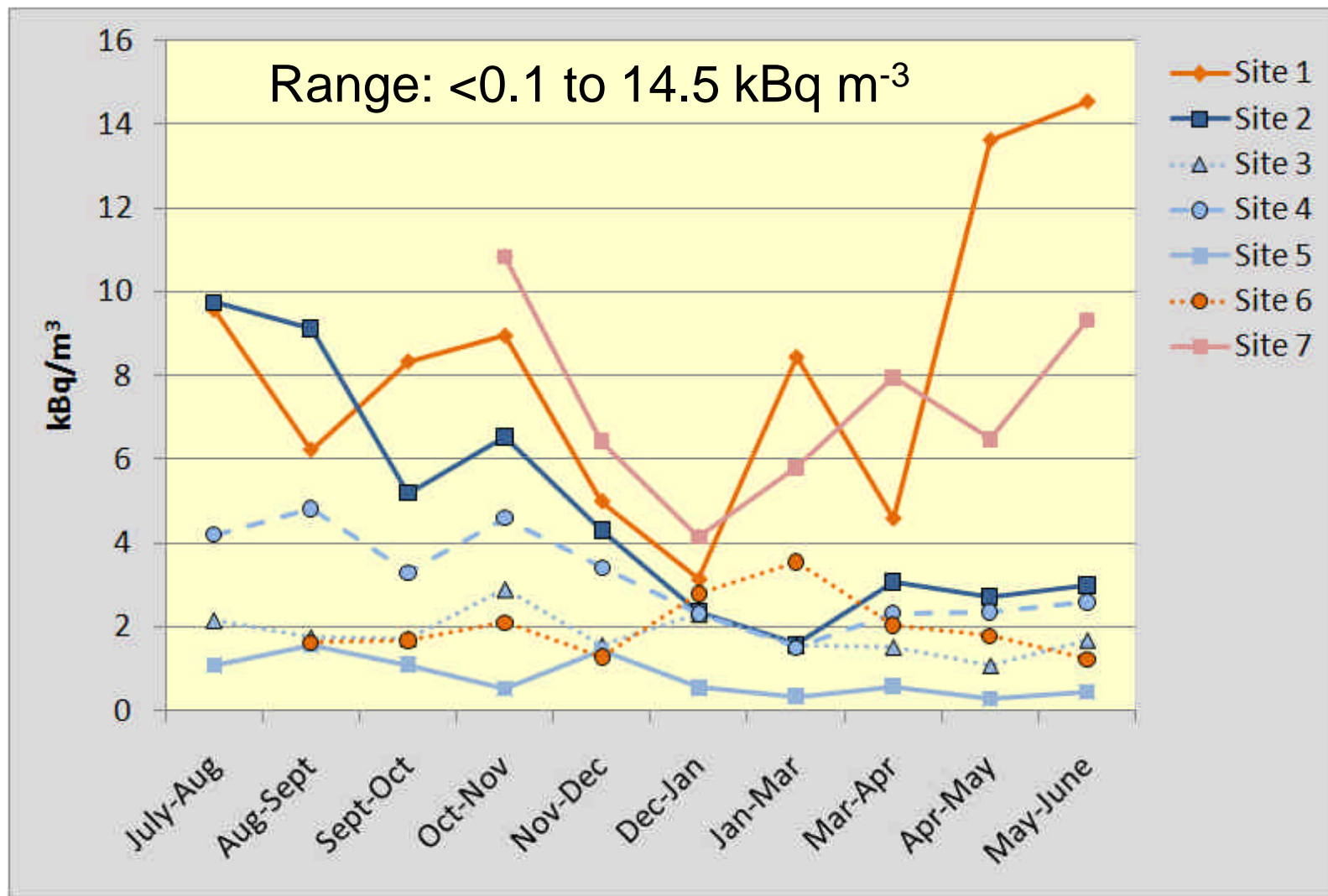
Example of application: Rn in burrowing mammals

- Available dose rate estimates for ^{222}Rn :
 - One study in area of 'Rn rich soils' in Canada
 - Whole body dose rate $>100 \text{ mGy y}^{-1}$ for small burrowing animals (c. $10\mu\text{Gy h}^{-1}$)
 - So Dose rate similar to *predicted no effect dose*
- So Beresford et al. (2012) delivered an estimation of ^{222}Rn dose rates to burrowing mammals at sites in the United Kingdom
- Dose rates calculated from measured field soil gas concentration, using the allometric methodology described previously
 - Assuming an equilibrium factor $F = 0.8$
 - Assuming an a-radiation weighting factor of 10
- 7 woodland, scrub and pasture sites selected to have range in potential Rn soil gas concentrations

- Make artificial burrows
- Use passive detectors developed by NRPB and SSI to measure soil gas ^{222}Rn activity concentration
- Sites across gradient of expected ^{222}Rn concentrations
- Detector placed in approximately 10-cm diameter perforated land-drainage tubing = 'artificial burrow'
- Tube (c.1.2 m) open ended on surface detector 50 cm below soil surface (surface length c.1 m). 3 per site
- Detector changed every 4-6 weeks (summer 2009 - summer 2010)



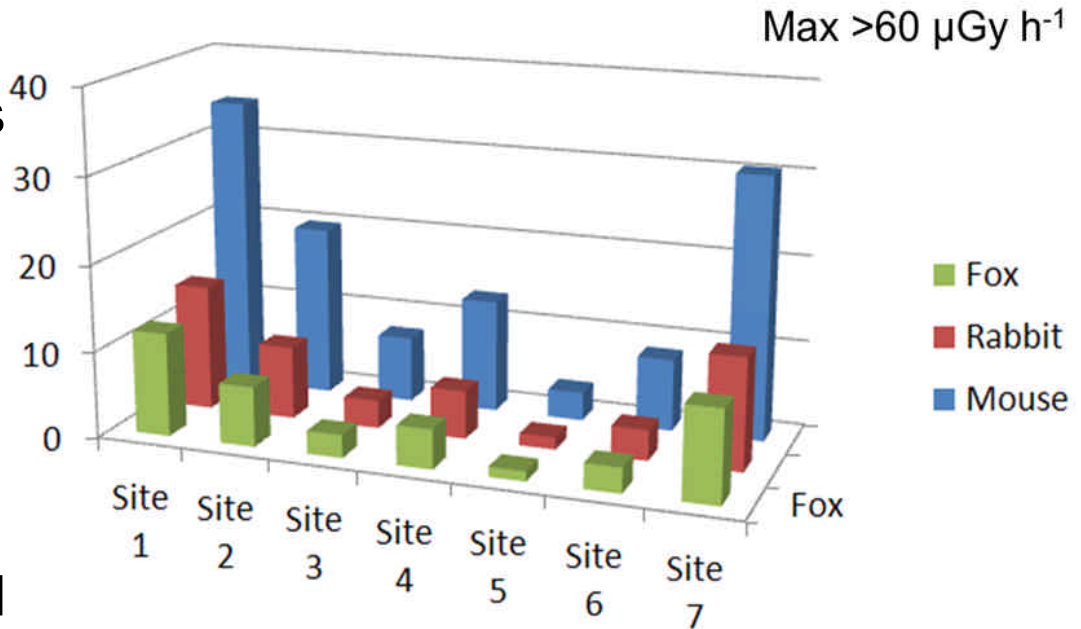
Measured ^{222}Rn concentrations in soil gas



Weighted dose rates

- Dose rate from ^{222}Rn to burrowing mammals likely to be at least 10 times higher than previously considered natural exposure sources (^{40}K , Th/U series).

- In many areas likely to considerably exceed predicted no-effect dose rate benchmarks.



Organism (geometry)	DCC ($\mu\text{Gy h}^{-1}$ per Bq m^{-3})
Mouse	2.6E-3
Rabbit	8.2E-4
Fox	9.1E-4

Conclusions

Argon and krypton (xenon)

- Ar and Kr dosimetry now codified into EA R&D 128 terrestrial spreadsheet.
- The R&D 128 DCC methodology for biota is consistent with the standard methodology for humans, the two only differences being:
 - External DCCs calculated for infinite geometry vs. DCFs for semi-infinite geometry (DCC correction factor of 0.5).
 - DCC method averages doses over whole volume - makes most difference in nucs. where external β -component predominates over γ , and (progressively) as the organism becomes larger.

Radon (allometric model)

- Radon dosimetry codified into DCCs for internal -irradiation arising from exposure of animals and plants to short-lived ^{222}Rn daughters.
- The ^{222}Rn DCCs can be used to produce an assessment in the normal way, using atmospheric radionuclide versions of the standard EA R&D 128 / ERICA formula for gaseous radionuclides.

Radon (advanced plant model)

- The predominant component of dose is surface-deposited ^{214}Po and (to a lesser extent) ^{218}Po activity.
- Less important are ^{214}Bi external exposure and ^{214}Po internal exposure.
- Doses to plant surface tissue are x 10 higher than the surface deposition dose averaged to the whole plant.
- Differences with respect to the allometric model due to combination of surface and internal dose and the equilibrium factor of 1 in the latter.

Radon exposures in mammals

- Radon levels in burrows exceeding background levels and no-effects benchmarks for non-human biota.
- Advised benchmark dose rates need to be better put into context with background dose rates, including exposure to ^{222}Rn , for credibility
- Context will be determined by the purpose of the benchmark and the assessment level.

- Integrate Ar, Kr, Rn assessment in a single tool (or incorporate into ERICA).
- Conduct assessments for new nuclear reactors including exposure to birds roosting in stacks.
- Perform additional investigations of allometric radon dosimetry for insects and plants.
- Seek evidence for dose rates that would cause stochastic effects in the lung using more detailed lung modelling (if appropriate).
- Consider how to extend the dose assessment for ^{226}Ra in soil.
- Consider developing similar approach to calculate thoron doses.
- Review benchmark values in context of background and radon levels in the natural environment.

- Vives i Batlle, J., Jones, S.R. and Copplestone, D. (2014). A methodology for the assessment of doses to terrestrial biota arising from external exposure to ^{41}Ar , $^{83,88}\text{Kr}$ and $^{132,133}\text{Xe}$. *Science of the Total Environment* (submitted).
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- Vives i Batlle, J., Copplestone, D. and Jones, S.R. (2012). Allometric methodology for the assessment of radon exposures to wildlife. *Science of the Total Environment*. **427-428**: 50–59
- Vives i Batlle, J., Smith, A., Vives-Lynch, S., Copplestone, D., Strand, T., Proehl, G. and Brown, J. (2011) Model-derived dose rates per unit concentration of radon in air in a generic plant geometry. *Radiation and Environmental Biophysics* **50**(4): 513-529.
- Vives i Batlle, J., Jones, S.R. and Copplestone, D. (2008) *Dosimetric Model for Biota Exposure to Inhaled Radon Daughters*. Environment Agency Science Report – SC060080, 34 pp.



STUDIECENTRUM VOOR KERNENERGIE
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