***New Phytologist* Supporting Information**

Article title: Radon as a Natural Tracer of Gas Transport Through Trees
Authors: J. Patrick Megonigal, Paul E. Brewer, and Karen L. Knee
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The following Supporting Information is available for this article:

Table S1 Diameter at breast height (DBH) and summer radon emission rates for tree stems as reported in Figure 1.

Species	**DBH (cm)**	**^{222}Rn Flux (Bq m^{-2} stem s^{-1})**
Fagus grandifolia	25.8	1.38
F. grandifolia	34.5	1.00
F. grandifolia	32.5	1.57
F. grandifolia	29.0	1.10
F. grandifolia	38.5	2.38
Liriodendron tulipfera	102.5	3.53
L. tulipfera	66.2	2.60
L. tulipfera	74.0	7.85
L. tulipfera	55.5	1.10
Quercus rubra	111.8	1.29
Q. rubra	80.0	3.60

Table S2 Spreadsheet model of radon flux underestimate due to radioactive decay (see separate file).

Methods S1 Methods for model of radon flux underestimate due to radioactive decay.

The loss of ^{222}Rn activity due to decay over the course of a closed-chamber flux measurement was estimated by calculating the decline in activity of individual 15-minute cohorts, matching the 15-minute interval programmed into the RAD7 for accumulating disintegrations in a single activity measurement. The loss of ^{222}Rn activity in an individual cohort emitted to the chamber over the remainder of the flux measurement was calculated as $A_t = A_0 e^{-\lambda t}$, where A_t = activity at time t, A_0 = initial activity, t = time (seconds) and λ = the ^{222}Rn decay constant (2.10 x 10^{-6} decays per second). The model tracked 20 cohorts emitted over a 5-hour period during which the ^{222}Rn flux was assumed to be linear at 10 Bq per cohort (Table a). The fraction of emitted radon activity that was counted was independent of the ^{222}Rn emission rate (Table S1, data not shown).

As modeled, the fraction of expected ^{222}Rn activity observed after accounting for decay was a linear function of the incubation time (Figure a) according to the equation $Y = -6\ x\ 10^{-5}\ X + 0.9996$ (R^2=0.9993), where Y = the fraction counted and X = time in minutes. The 0-15 minute cohort was excluded because no meaningful decay occurred during the period (i.e. the fraction counted was 1.000). Although the decline in ^{222}Rn activity for a given cohort is exponential, the loss of ^{222}Rn activity over the course of a given incubation was approximately linear because new cohorts were being added at a linear rate. Radon-222 flux in our study was underestimated by 1.2% (3 hours) to 2.3% (6 hours), except for 4 fall measurements that were underestimated by 3.1% (8 hours). The incubation time for each of the six rates measured during the diurnal flux campaign were the same (4 hours), so each interval was underestimated equally and ^{222}Rn decay does not account for the decline in rates over night.

Methods S2 Radon produced and emitted from wood.

One aim of our study was to determine if upland trees emit soil-produced radon emitted whether by uptake of Rn-enriched groundwater or vadose zone water, or by diffusion. However, we recognize that some fraction of tree Rn emissions may also result from the decay of ^{226}Ra (half-life 1600 years) in the wood, and would thus occur in the absence of any soil source. Plants take up ^{226}Ra (half-life 1600 years) from soils where it accumulates in tissues (Simon & Ibrahim, 1990) and decays to ^{222}Rn. We estimated the amount of ^{222}Rn emitted from wood samples of the two focal tree species in our study – *Liriodendron tulipfera* and *Fagus grandifolia* – by assuming that ^{222}Rn emitted into the stem flux chambers was produced within a wedge-shaped volume of wood, the oven-dry weight (DW) of which was calculated as:

$$DW = \frac{rwhd}{2}$$

where r is the tree radius (m), w=chamber width (m), h=chamber height (m), and d=wood density, defined as dry weight of wood per green volume of wood (Kg/m^3). Wood density on a green volume basis was 560 Kg/m^3 for *L. tulipfera* and 400 Kg/m^3 for *F. grandifolia* as reported in Miles & Smith (2009).

The ^{222}Rn activity produced by a given wedge-shaped volume of wood was determined by the equation (Ishimori *et al.*, 2013):

$$P = \lambda REM$$

Where P is the radon production rate (Bq s^{-1}), λ is the ^{222}Rn decay constant (2.10 x 10^{-6} decays per second), R is the ^{226}Ra activity of the wood (Bq kg^{-1}), E is the emanation coefficient (dimensionless), and M is the mass of the wood (kg). The production rate was then expressed in the same units as the tree fluxes (mBq s^{-1} per m^2 of tree surface) by converting Bq to mBq and dividing P by the surface area of the flux chamber (0.03 m^2 stem surface). This quantity was used to calculate the fraction of stem ^{222}Rn flux that could have been supported by wood-derived ^{222}Rn (Table b).

Wood ^{226}Ra activity was measured directly on samples of the two species collected within 3 km of the study site and on the same soil series. The wood sample was taken from a cross-section of the tree trunk at about 1 m height. The sample was cut into a wedge shape (wide side at the bark) to physically approximate the wedge-shape assumption used in the calculation. Thus, the sample was approximately 15 cm wide at the outer (bark) edge and extended from the bark to the center

of the tree. The wood sample was reduced to smaller pieces ranging from 1-8 cm^3 and mixed. Subsamples of the mixture weighing 171 g wet weight (85 g dry weight) for *L. tulipfera* and 149 g wet weight (113 g dry weight) for *F. grandifolia* were placed into an air-tight vessel for a period of 21 days until ^{222}Rn and ^{226}Ra were in secular equilibrium. Rn-222 activity was then determined on a RAD7 instrument from which we calculated ^{226}Ra activity. Wood dry weight was determined after the incubations ended by heating the sample to 80 °C for 4 days.
The radium activity of *Quercus rubra* was not measured, so it was estimated from the ^{226}Ra activity of *F. grandifolia* wood which has the same wood density (400 Kg/m^3) as *Q. rubra*.

A portion of the ^{222}Rn produced by ^{226}Ra decay does not escape from the site of production (i.e. organic tissue where radium is bound) into interstitial spaces through which it emanates from the tree. Emanation coefficients (*E*) for soils average about 0.2 and range from 0.05 to 0.70 (Ishimori *et al.*, 2013). We found no published estimates of *E* for wood and assumed that emanation from soil organic matter is a reasonable approximation of wood *E*. Greeman & Rose (1996) estimated an *E* of 0.46 for soil organic matter based on sequential extraction of 12 soils from five states of the eastern USA, so we adopted 0.5 as our estimate of wood *E*. We may have overestimated the contribution of ^{222}Rn from wood by ignoring the possibility that ^{222}Rn emitted into interstitial spaces decays before reaching the outside of the tree. The diffusion time of a gas increases with the square of diffusion distance, so ^{222}Rn loss to decay increases with the square of tree diameter. Because radon is mobile in wood we assumed that ^{226}Ra and ^{222}Rn are not in secular equilibrium (Lewis & MacDonell, 1980).

Supplementary References

Greeman DJ, Arthur WR. 1996. Factors controlling the emanation of radon and thoron in soils of the eastern U.S.A. *Chemical Geology* **129:** 1-14

Lewis BG, MacDonell MM. 1990. Release of Radon-222 by vascular plants: Effect of transpiration and leaf area. *Journal of Environmental Quality* **19:** 93-97.

Ishimori Y, Lange K, Martin P, Mayya YS, Phaneuf M. 2013. *Measurement and Calculation of Radon Releases from NORM Residues*, IAEA Technical Reports Series, No. 474. Vienna, Austria: International Atomic Energy Agency.

Miles PD, Smith WB. 2009. *Specific gravity and other properties of wood and bark for 156 tree species found in North America*. Res. Note NRS-38. Newtown Square, PA: U.S. Department of Agriculture, Forest Service, Northern Research Station.

Simon SL, Ibrahim SA. 1990. Biological uptake of radium by terrestrial plants. In: *The Environmental Behaviour of Radium*. IAEA Technical Report Series, No. 310., Vienna, Austria: International Atomic Energy Agency. 545–599.

Table a. Results of a spreadsheet model of loss of radon due to radioactive decay in a closed flux system over the course of a five-hour incubation. Twenty separate cohorts of ^{222}Rn are emitted at 15-minute intervals with an initial activity of 10 Bq per cohort, which then decay over the remainder of the incubation period. Emission cohort number columns show remaining ^{222}Rn activity as a function of elapsed time (rows). The total counted is less than the total emitted due to radioactive decay of ^{222}Rn.

Elapsed time (minutes)	Emission Cohort Number																				Total Emitted	Total Counted	Fraction Counted
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20			
	Modeled Rn-222 Activity of Cohort (Bq)																						
15	10																				10	10.00	1.000
30	9.96	10																			20	19.96	0.998
45	9.94	9.96	10																		30	29.91	0.997
60	9.92	9.94	9.96	10																	40	39.83	0.996
75	9.91	9.92	9.94	9.96	10																50	49.74	0.995
90	9.89	9.91	9.92	9.94	9.96	10															60	59.62	0.994
105	9.87	9.89	9.91	9.92	9.94	9.96	10														70	69.49	0.993
120	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10													80	79.34	0.992
135	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10												90	89.17	0.991
150	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10											100	98.99	0.990
165	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10										110	108.78	0.989
180	9.78	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10									120	118.56	0.988
195	9.76	9.78	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10								130	128.31	0.987
210	9.74	9.76	9.78	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10							140	138.05	0.986
225	9.72	9.74	9.76	9.78	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10						150	147.77	0.985
240	9.70	9.72	9.74	9.76	9.78	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10					160	157.47	0.984
255	9.68	9.70	9.72	9.74	9.76	9.78	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10				170	167.16	0.983
270	9.67	9.68	9.70	9.72	9.74	9.76	9.78	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10			180	176.82	0.982
285	9.65	9.67	9.68	9.70	9.72	9.74	9.76	9.78	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10		190	186.47	0.981
300	9.63	9.65	9.67	9.68	9.70	9.72	9.74	9.76	9.78	9.79	9.81	9.83	9.85	9.87	9.89	9.91	9.92	9.94	9.96	10	200	196.10	0.981

Table b. Estimated contribution of wood-produced radon to radon emissions from stems of *Fagus grandifolia*, *Lirodendron tulipfera*, and *Quercus rubra*.

Species	**Tree diameter**	**Chamber width**	**Chamber height**	**Wedge volume**	**Wood density**	**Wedge mass**	**Radium activity**	**Emanation Factor**	**Radon activity**	**Wood-supported radon**	**Total stem radon flux**	**Wood-supported fraction**
	(m)	(m)	(m)	(m3)	(Kg/m^3)	(Kg)	(Bq/Kg)	(unitless)	(Bq)	(mBq/m^2 tree surface)		(%)
F. grandifolia	0.345	0.15	0.20	0.0026	560	1.45	0.74	0.5	1.13E-06	0.038	1.004	4%
F. grandifolia	0.325	0.15	0.20	0.0024	560	1.37	0.74	0.5	1.06E-06	0.035	1.569	2%
F. grandifolia	0.290	0.15	0.20	0.0022	560	1.22	0.74	0.5	9.46E-07	0.032	1.101	3%
F. grandifolia	0.385	0.15	0.20	0.0029	560	1.62	0.74	0.5	1.26E-06	0.042	2.384	2%
F. grandifolia	0.258	0.20	0.24	0.0031	560	1.73	0.74	0.5	1.35E-06	0.045	1.383	3%
L. tulipfera	0.662	0.15	0.20	0.0050	400	1.99	2.59	0.5	5.40E-06	0.180	2.596	7%
L. tulipfera	0.740	0.15	0.20	0.0056	400	2.22	2.59	0.5	6.03E-06	0.201	6.534	3%
L. tulipfera	0.555	0.15	0.20	0.0042	400	1.67	2.59	0.5	4.52E-06	0.151	1.099	14%
L. tulipfera	1.025	0.15	0.20	0.0077	400	3.08	2.59	0.5	8.36E-06	0.279	3.534	8%
Q. rubra	0.800	0.15	0.20	0.0060	560	3.36	0.74	0.5	2.61E-06	0.087	3.597	2%
Q. rubra	1.118	0.15	0.20	0.0084	560	4.70	0.74	0.5	3.65E-06	0.122	1.327	9%

Figure a. The fraction of emitted ^{222}Rn activity observed after accounting for radioactive decay according a cohort model (Table a, Methods S1). The first interval from 0-15 minutes is excluded. The relationship is approximated by the linear equation $Y = -6.4191 \, x \, 10^{-5} \, X + 0.99961$ (R^2=0.9993).