

Radon as a Natural Tracer of Gas Transport Through Trees

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Summary

- Trees are sources, sinks, and conduits for gas exchange between the atmosphere and soil, and effectively link these terrestrial realms in a soil-plant-atmosphere continuum.

- 18 • We demonstrated that naturally produced ^{222}Rn gas has the potential to disentangle the
19 biotic and physical processes that regulate gas transfer between soils or plants and the
20 atmosphere in field settings where exogenous tracer applications are challenging.
21
- 22 • Patterns in stem radon emissions across tree species, seasons, and diurnal periods suggest
23 that plant transport of soil gases is controlled by plant hydraulics, whether by diffusion or
24 mass flow via transpiration.
25
- 26 • We establish for the first time that trees emit soil gases during the night when
27 transpiration rates are negligible, suggesting that axial diffusion is an important and
28 understudied mechanism of plant and soil gas transmission.

29
30 **Key Words:** plant radon emissions, plant hydraulics, plant radon tracer, tree methane emissions,
31 tree carbon dioxide emissions
32

33 **Introduction**

34 Gas exchange is a fundamental life process that governs photosynthesis, respiration, primary
35 production and many other facets of the ecophysiology of plants. It is also a primary mechanism
36 by which plants control the chemical composition of Earth's atmosphere. The factors that
37 regulate gas exchange across leaves are well understood and captured in models operating at
38 scales ranging from a single leaf to the planet (Franks *et al.*, 2018). By comparison, the factors
39 that regulate tree stem gas exchange are poorly understood and presently limit our ability to
40 partition stem gas exchange in eddy covariance studies and to scale measurements of stem gas
41 exchange from individual trees to forests. A particularly significant limitation to developing

42 conceptual and numerical models of tree stem gas exchange is isolating the physical processes
43 that govern gas flux through trees from the plant and microbial sources and sinks that also
44 regulate most gas fluxes. Progress in forest gas exchange science requires new detailed and
45 nuanced studies of tree stem gas dynamics that disentangle physical and biological processes
46 (Hölttä & Kolari, 2009; Steppe *et al.*, 2015; Barba *et al.*, 2019; Covey & Megonigal, 2019).

47 Plant-mediated transport of gases between soils and the atmosphere is well documented
48 in wetland ecosystems but is poorly understood in upland ecosystems on freely drained soils.
49 This is particularly the case for upland trees, which can both emit and remove the greenhouse
50 gases CO₂, CH₄ and N₂O, as well as volatile organic carbon compounds and other gases that
51 govern atmospheric chemistry. It has proven difficult to establish the provenance of greenhouse
52 gases emitted from trees because gases can be simultaneously produced and consumed in the tree
53 itself and in the underlying soil (Covey & Megonigal, 2019). Additionally, gases can be
54 transported by trees from their site of origin in trees or soils before being emitted to the
55 atmosphere. The potential for trees to transport soil gases to the atmosphere has been established
56 in the laboratory (Pearson and Jones, 1966; Garnet *et al.* 2005), but no study has isolated
57 physical gas transport from soils through trees in an intact forest ecosystem to our knowledge.

58 Here we demonstrate that naturally produced radon gas is a useful tracer of soil gas
59 transport through trees. Radon-222 is a radioactive noble gas with a half-life of 3.8 days. It is a
60 product of the ²³⁸U decay series generated by α -decay of ²²⁶Ra, which has a 1600-year half-life
61 and is present in most soils. Radon moves by the same mass flow and diffusion pathways that
62 transport other soil gases, but radon is neither produced nor consumed by biological processes
63 that can obscure the origin or transport rates of other gases. Collectively, these characteristics

64 suggest that radon has the potential to provide novel insights into the movement of gases through
65 trees.

66 Plants take up ^{222}Rn 's parent element, ^{226}Ra , from soils (Simon and Ibrahim 1990) and
67 ^{222}Rn emissions have been observed from herbaceous plants and crops such as corn, sunflower,
68 and fescue growing on mine tailings (Schery *et al.*, 1984). Trees are hypothesized to emit radon
69 entrained in their transpiration stream (Trumbore *et al.*, 1990; Jayaratne *et al.*, 2011), but a tree
70 radon flux has never been measured directly. Forest studies directed at constraining stand-level
71 gas CO_2 exchange rates have assumed that tree emissions are negligible compared to soil
72 emissions, and near zero at night in synchrony with transpiration rates (Trumbore *et al.*, 1990;
73 Ussler *et al.*, 1994; Martens *et al.*, 2004; Jayaratne *et al.*, 2011).

74 In all cases where radon transmission through trees has been considered, transpiration
75 was assumed to be the dominant transport mechanism. However, molecular diffusion of gases
76 occurs to some extent in all tree stems as evidenced by exchange of CO_2 (Teskey *et al.*, 2008)
77 and O_2 (Soriz and Heitz, 2006). Gases are known to move through trees by passive diffusion
78 (Rusch & Rennenberg 1998) and pressurized ventilation (Große & Schröder, 1984), a
79 mechanism that drives mass flow in trees. Thus, it is likely that all trees have the capacity to
80 transmit gases between soils and the atmosphere in both directions by transpiration-independent
81 mechanisms.

82 Mechanisms of tree stem gas exchange are important for solving a wide variety of forest-
83 atmosphere research problems. Eddy covariance and modeling studies seek to partition stem
84 emissions of CO_2 (Reichstein *et al.*, 2012); stems are a poorly quantified CH_4 source with
85 important implications for global budgets (Barba *et al.*, 2019; Covey and Megonigal, 2019);
86 stems contribute to tree emissions of volatile organic carbon (Hölttä and Kolari, 2009); O_2

87 transport regulates the metabolism of tree stems; and radon emissions influence cluster ion and
88 aerosol formation through alpha decay of ^{222}Rn in the atmosphere (Jayaratne *et al.*, 2011).

89 The goals of our study were: (i) establish whether or not radon gas is emitted from upland
90 tree stems, (ii) quantify rates of tree stem radon emissions and the fraction supported by ^{222}Rn
91 produced in wood versus soil, (iii) relate tree stem emission rates of radon, CO_2 and CH_4 , and
92 (iv) test the hypothesis that radon is useful for isolating the physical processes that regulate gas
93 transport through trees.

94

95 **Methods**

96 The study was conducted in a temperate hardwood forest at the Smithsonian Environmental
97 Research Center in Edgewater, MD, a site used for previous studies of tree stem greenhouse gas
98 fluxes (Pitz and Megonigal, 2017; Pitz *et al.*, 2018). From 12 Jun 2018 to 04 Oct 2018 we
99 measured tree stem radon emissions (n= 31 measurements), soil surface radon emissions (n=6),
100 soil profile radon concentrations (n=19), groundwater radon concentration (n=1), and air radon
101 concentrations (n=2).

102 Stem flux chambers were fit to 11 trees of three species, providing replicate stems for
103 *Fagus grandifolia* Ehrh. (n=5), *Liriodendron tulipifera* L. (n=4), and *Quercus rubra* L. (n=2).
104 Stem diameter at breast height (DBH) ranged from 26-112 cm, and the average DBH of *F.*
105 *grandifolia* stems was >50% smaller than *L. tulipifera* and *Q. rubra* (32, 75, and 96 cm DBH,
106 respectively, Table S1). Stem flux chambers were similar to those described previously (Pitz and
107 Megonigal, 2017). The chambers had a stem surface area of 0.030 m² and a volume of 1.905 L.
108 The only exception was one *F. grandifolia* stem (ID=1) which we fit with a chamber with a

109 relatively large surface area (0.048 m²) and small volume (0.448 L) to increase sensitivity. This
110 chamber was based on the design of Siegenthaler *et al.* (2016).

111 Soils at the study site are of the Collington-Annapolis series and developed from loamy
112 glauconitic fluviomarine deposits (Yesilonis *et al.*, 2016). Soil flux chambers and soil gas wells
113 were co-located in five locations across the study area. Soil flux chambers were 30 cm in
114 diameter (surface area 0.071 m²) and 10 cm in height, and they were inserted approximately 4
115 cm into the soil surface. Height varied with microtopography, so the combined volume of the
116 chamber and lid ranged from 6.224-8.062 L. The total volume of the tubing and instrument
117 (1.224 L) was added to the chamber volumes to calculate total gas dilution.

118 Radon activity was measured using a RAD7 electronic detector (DurrIDGE Company, Inc.,
119 Billerica, MA), with a manufacturer-reported radon sensitivity of 9.25 Bq m⁻³. The RAD7 was
120 connected to a flux chamber in a closed loop and continuously circulated gas between the
121 chamber and instrument over the measurement period. The instrument was programmed to
122 accumulate decay counts at 15-minute intervals, which were then summed over 30-minute
123 intervals in Jun-Jul when emission rates were relatively high, or 45-minute intervals in Sep-Oct
124 when rates were relatively low. Low rates of radon emissions from trees required measurement
125 periods that ranged from 4-8 hours. After summing, the maximum number of summed counts per
126 flux measurement was 192±206 (mean±SD) with a median of 130 counts and a range of 18-758
127 counts. Flux rates were not corrected for ²²²Rn decay because the 3.8 day half-life is much longer
128 than the flux measurement period, and thus reported rates underestimate radon flux by 1.6% (4
129 hour flux) to 2.3% (6 hour flux) (Methods S1, Table S2), except for four measurements in the
130 fall with an error of 3.1% (8 hour flux). Note that the intervals for each of the six rates measured

131 during the diurnal flux campaign were the same (4 hours), so the intervals are all underestimated
132 equally.

133 Stem flux chambers for radon measurements were designed for automated CO₂ and CH₄
134 flux measurements which ran at 2-hour intervals before and after the radon flux measurement.
135 Whenever possible, radon fluxes were compared to CO₂ and CH₄ fluxes measured on the same
136 day. On two dates there were no usable CO₂ and CH₄ flux data to match the radon flux from an
137 individual tree, so CO₂ and CH₄ flux data for the tree from the day before and the day after were
138 averaged. On a single occasion, one a tree with a very high rate of radon emissions was
139 measured continuously over 24 hours (six consecutive 4-hour measurement intervals) to test for
140 diurnal variation in flux.

141 Groundwater was extracted from a groundwater well at 3.0 m depth after flushing for
142 several minutes, and the activity of dissolved ²²²Rn (Bq m⁻³ water) in a 2-L sample was
143 measured with the RAD7 attached to a RAD-H₂O Big Bottle Accessory (DurrIDGE, Billerica,
144 MA). Triplicate samples were collected from the same well at the same depth and had a relative
145 uncertainty of <5%.

146

147 *Data Reduction and Statistical Analyses*

148 Fluxes were calculated from a linear fit of the increase in radon activity over time using SAS
149 Proc Reg, and scaled to chamber volume and area with the equation: $F_{Rn} = S_{Rn} \cdot V / A$, where F_{Rn}
150 is radon flux in Bq m⁻² s⁻¹, S_{Rn} is the slope of radon activity versus time in Bq m⁻³ s⁻¹, V is
151 chamber volume in m³, and A is chamber area in m². Area refers to stem surface area covered by
152 the chamber for tree fluxes, or soil surface area covered by the chamber for soil fluxes.

153 Two of the stem flux radon regressions were not significant at $p < 0.05$, so these flux data
154 were not used for further analyses. The remaining 29 stem radon fluxes were further reduced by
155 averaging. Because radon, CO_2 , and CH_4 fluxes were measured over different time intervals
156 (hours for radon versus minutes for the other gases), all the fluxes for a given tree made in a 24-
157 hour period were averaged, yielding one flux estimate of radon, CO_2 and CH_4 in that 24-hour
158 interval. The replicated unit in the study was an individual tree or soil chamber, so in instances
159 where multiple flux measurements were made on the same tree in a season, the fluxes were
160 averaged. Most radon flux measurements were made during the summer (Jun-Jul), composed of
161 one flux for each of the 11 trees. A subset ($n=4$) of the same trees was remeasured in the fall
162 (Sep-Oct).

163 SAS Proc GLM was used to test for species differences in summer radon emissions.
164 Differences between individual stems across seasons was determined with a paired T-test using
165 SAS Proc TTest. Levene's test in Proc GLM was used to test for homogeneity of variance across
166 species and seasons, and variance was homogeneous in these groups in all cases ($P > 0.05$).
167 Regression relationships between radon, CH_4 , and CO_2 were performed in SigmaPlot separately
168 by tree species, except for *Q. rubra* ($n=2$) for which the sample size was insufficient for the test.
169

170 **Results and Discussion**

171 Gas transport rates through porous media are a function of the source concentration whether the
172 mechanism is diffusion or mass flow. Radon-222 activity in soil pore space increased four orders
173 of magnitude from the soil surface to 80 cm depth, and soil radon activity at 80 cm was similar to
174 groundwater (Fig. 1). The maximum soil pore space activity of $38.4 \pm 55.1 \text{ kBq m}^{-3}$ (mean \pm SD)
175 was higher than in most other studies (e.g. Trumbore *et al.*, 1990; Davidson *et al.*, 2006; de Faria

176 *et al.*, 2016), suggesting that our study site was particularly well suited for detecting ^{222}Rn flux
177 through tree stems because of a strong concentration gradient between the soil rooting zone and
178 the atmosphere. The soil surface radon emission rate at our site of $48.4 \pm 8.8 \text{ mBq m}^{-2} \text{ soil s}^{-1}$
179 exceeds soil emission rates from many other sites (Jayaratne *et al.*, 2011), acknowledging that
180 such comparisons are limited by the short-term variation introduced by factors such as soil water
181 content (Ota *et al.*, 2007) and soil temperature gradients (Ota and Yamazawa, 2010). The reasons
182 for high radon activity are uncertain but include geologic factors such as U content of the parent
183 material and geochemical factors such as radium solubility and mobility that affect ^{222}Rn
184 emanation rates (Wanty *et al.*, 1991).

185 Summer radon emissions from tree stems averaged $2.5 \pm 2.0 \text{ mBq m}^{-2} \text{ stem s}^{-1}$ (Fig. 2,
186 Table S1). We estimate that 86-98% of the radon emitted from tree stems had a soil source,
187 averaging $97 \pm 1\%$ in *F. grandifolia* and $92 \pm 4\%$ in *L. tulipifera* (Methods S2). The remainder (2-
188 14%) came from the decay of ^{226}Ra incorporated into wood. These are the first radon fluxes
189 reported for trees or any other plant in an intact ecosystem to our knowledge. However, leaf
190 radon emissions from crops grown in uranium mill tailings under greenhouse conditions ranged
191 from 8 to $28 \text{ mBq m}^{-2} \text{ leaf s}^{-1}$ (Lewis and MacDonnel, 1990).

192 Summer stem radon emissions did not vary significantly across the three species, but
193 mean rates decreased in the order *L. tulipifera* > *Q. rubra* > *F. grandiflora* (Fig. 2). Rates did not
194 differ significantly between summer and fall, but summer rates tended to be higher than fall rates
195 (2.5 ± 2.0 and $1.1 \pm 0.2 \text{ mBq m}^{-2} \text{ stem s}^{-1}$, respectively). Evidence of temporal dynamics in stem
196 radon emissions were observed in six consecutive measurements made on a single tree over a 24-
197 hour period, whereby the flux declined monotonically by 38% during the night, then rose again
198 in daylight (Fig. 3). Collectively, these patterns suggest that meaningful variation in plant radon

199 emission rates exist in the field, but that large sample sizes are required to confidently isolate the
200 role of physical processes in regulating gas transport through trees.

201 Mechanisms of gas transport in trees include mass flux driven by pressure gradients and
202 molecular diffusion driven by concentration gradients. Trumbore *et al.* (1990) estimated an upper
203 limit to radon emissions from trees in a tropical forest by assuming transpiration as the sole
204 transport mechanism, calculating an ecosystem rate equivalent to 5% of the soil flux. Because
205 transpiration is negligible at night, tree radon emissions were assumed to be as well. The same
206 assumptions were used in another tropical forest in Brazil (Martens *et al.*, 2004), a boreal forest
207 dominated by *Picea marianana* in Canada (Ussler *et al.*, 1994), and a *Picea*- and *Betula*-
208 dominated forest in Russia (Langendörfer *et al.*, 2002). Our data are consistent with the concept
209 that trees transport soil gas via transpiration because rates peaked during the day and were higher
210 in summer than fall, but it is also clear that radon can be emitted from tree stems at night (Fig. 4).
211 Minimum emissions from one *L. tulipifera* tree occurred at the night-to-day transition and were
212 about 60% of maximum daytime emissions. Transpiration could be the sole mechanism driving
213 night emissions if there are time lags between sap flux transport of radon aboveground and radial
214 diffusion out of the stem (Hölttä & Kolari, 2009). Alternatively, the temporal patterns of stem
215 radon emissions we observed are also consistent with gaseous diffusion as the sole mechanism
216 driving diurnal variation, with higher rates during daylight when tree stem water content is
217 relatively low. It is quite likely that the two mechanisms were operating simultaneously to some
218 extent.

219 The physical processes that govern radon emissions from soils are expected to apply to
220 plants. Temporal variation in soil radon emissions are strongly related to soil water content,
221 responding rapidly to precipitation events (Ussler *et al.*, 1994) and seasonally to variation in

222 water table depth (Levin *et al.*, 2002). We hypothesize that stem moisture content is an important
223 control on radon diffusion through tree stems, which are also porous bodies in which gas-filled
224 space is 18-50% of the stem volume (Gartner *et al.*, 2004). Stem water content is a dominant
225 physical factor that regulates radial diffusion of O₂ (Sorz and Hietz, 2006; Mugnai and Mancuso,
226 2010) and CO₂ (Teskey *et al.*, 2008) in tree stems, and is a likely explanation for the radon
227 emissions patterns observed here. Stem water content is known to decline during the day as
228 transpiration withdraws stem water, and then increase at night to a new maximum (reviewed by
229 Steppe *et al.*, 2015). This diurnal pattern in stem hydraulics can be observed as a daylight decline
230 in tree stem diameter, which was documented through high frequency sap flow and diameter
231 variation at the present study site (Herrmann *et al.*, 2016). Although stem water content was not
232 measured at our study site, it is expected to be lower during day versus night and in summer
233 versus fall, following patterns of transpiration. Provided that relatively low stem water content
234 coincides with high gas diffusion rates as expected from physical principles, this pattern matches
235 periods of relatively high radon flux from tree stems and suggests that soil gases can be
236 transmitted from upland trees by molecular diffusion. We cannot distinguish the relative
237 contributions of molecular diffusion and mass flow to stem radon emissions in the present study,
238 but such partitioning should be possible by combining radon flux measurements with data on sap
239 flow, stem water content, stem gas volume, and other stem ecophysiological measurements in
240 highly instrumented trees such as those described by Steppe *et al.* (2015).

241 Stem gas fluxes are constrained by the anatomy of water- and air-conducting elements
242 (Carlquist, 2001; Sorz and Hietz, 2006). For example, O₂ diffusion rates through wood decrease
243 in the order: diffuse-porous hardwoods > ring-porous hardwoods > gymnosperms (Sorz and
244 Hietz, 2006). *L. tulipifera* than *F. grandifolia* are diffuse-porous hardwoods but other aspects of

245 their wood anatomy may explain why radon emissions tended to be higher in *L. tulipifera*.
246 Alternatively, the difference may relate to the fact that the average *F. grandifolia* stem was 50%
247 smaller in DBH than other two species (Table S1). Radon may prove useful for identifying plant
248 traits that determine species-specific stem gas diffusion rates, which in turn may be used to
249 quantify stem diffusion rates for other gases of interest such as CO₂ and CH₄.

250 Radon emissions from *L. tulipifera* stems were significantly related to emissions of CO₂
251 ($P=0.037$) and CH₄ ($P=0.048$) (Fig. 4). These relationships suggest that the same physical
252 processes that regulate radon emissions also regulate emissions of gases subject to biological
253 sources and sinks. These relationships do not mean the three gases necessarily share a common
254 plant or soil source, only that their axial emissions have some physical constraint in common.
255 The fact that CO₂ and CH₄ emissions were related ($P=0.060$) despite having different soil depth
256 profiles suggests that instantaneous emissions were regulated by variation in axial diffusion from
257 the stem interior regardless of the mix of plant and soil sources. By comparison, *F. grandifolia*
258 stem emissions of CO₂ and CH₄ were not correlated with radon (data not shown), a difference
259 that may relate to wood morphology, differences in average tree size, or differences in CO₂ or
260 CH₄ sources and sinks.

261 Scaling tree stem gas emissions in forest gas budgets is difficult because rates vary with
262 stem height, component (stem, branch, leaf), and time (Covey and Megonigal, 2019; Barba *et al.*,
263 2019). This challenge applies equally to scaling radon emissions from tree stems to whole
264 forests. In the present case, if we assume that the mean stem radon emission rate was uniform to
265 a stem height of 2 m (Table S1), radon emitted from an average stem per area of occupied soil
266 surface is 40.0 mBq m⁻² soil surface s⁻¹, a rate similar to soil surface emissions. If radon is also
267 emitted at heights above 2 m then trees may be hot spots, emitting more radon than the soil area

268 they occupy. On the other hand, stem surface area in this forest to 3 m height is just 13% of the
269 soil surface area (Pitz & Megonigal, 2017), suggesting that trees may be a relatively small radon
270 source at the forest ecosystem scale.

271 In conclusion, soils and plants are porous bodies that conduct gases and fluids according
272 to biological and physical drivers that are difficult to disentangle. Radon is a natural tracer with a
273 wide variety of applications to tree-gas research including studies of greenhouse gas emissions
274 (Fig. 4), volatile organic carbon emissions (Rissanen et al., 2018), hypoxia effects on physiology
275 and development (Spicer and Holbrook, 2005), and forest regulation of aerosol production
276 (Jayaratne *et al.*, 2011). Patterns in stem radon emissions across species, seasons, and diurnal
277 periods suggested that tree transport of soil-produced and plant-produced gases is controlled by
278 plant hydraulics, and that trees emit soil gases during the night when transpiration rates are
279 negligible. Further studies are required to establish the generality of these trends, and to refine
280 the methods for using radon to study soil-plant-atmosphere gas exchange.

281

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287

288 **Author contributions**

289 JPM conceived, designed, and performed the radon flux research, and wrote the manuscript. PEB
290 conceived, designed, and constructed the tree and soil gas flux chambers, gas wells, and

291 groundwater wells, and performed the methane and carbon dioxide flux research. KKK provided
292 the RAD7 instrument, radon measurement expertise, and groundwater radon measurements. PEB
293 and KKK contributed to the experimental design and edited the manuscript.

294

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407 **Supporting Information**

408 Additional Supporting Information may be found online in the Supporting Information tab for
409 this article:

410 **Table S1.** Size and summer ^{222}Rn emissions for trees in the stem radon flux study.

411 **Table S2.** Excel model for calculating the underestimate in radon flux due to radioactive decay.

412 **Methods S1.** Methods for calculating the underestimate in radon flux due to radioactive decay.

413 **Methods S2.** Methods for calculating the contribution of wood-produced radon to stem surface
414 radon emissions.

415

416 **Figure 1.** Depth profiles of soil ^{222}Rn activity, CO_2 concentration, and CH_4 concentration in
417 unsaturated soil to 80 cm, and ^{222}Rn in groundwater at approximately 300 cm. Points are means
418 ± 1 SD. Note the different units for the three gases and breaks in the radon X axis and Y (depth)
419 axis. Radon in groundwater is in units of Bq m^{-3} water. Groundwater concentrations of CO_2 and
420 CH_4 are not reported.

421

422 **Figure 2.** Tree stem and soil surface radon emissions in summer. The species investigated were
423 *Liriodendron tulipifera*, *Fagus grandifolia*, and *Quercus rubra*. Different letters denote
424 significant differences in flux at $P < 0.05$. Tree radon emissions averaged across the three species
425 was 2.5 ± 2.0 mBq m^{-2} stem (means ± 1 SD).

426

427 **Figure 3.** Radon emissions from a single *Liriodendron tulipifera* stem repeated at 4-hour
428 intervals over 24 hours. Colors denote daylight (white), night (black), or the dawn transition
429 from night to day (grey).

430

431 **Figure 4.** Relationships between stem emissions of ^{222}Rn and CO_2 (upper panel) and ^{222}Rn and
432 CH_4 (lower panel) for four *Liriodendron tulipifera* trees. Two trees were measured twice on
433 separate dates, and all measurements were made in the summer. The solid lines are best-fit linear
434 regression relationships, and dotted lines are 95% confidence limits.

[CO₂] (%) and [CH₄] (ppm)







