

# Radon as a Natural Tracer of Gas Transport Through Trees

J. Patrick Megonigal<sup>1,\*</sup> (ORCID: 0000-0002-2018-7883), Paul E. Brewer<sup>1</sup> (ORCID: 0000-0002-2715-5286), and

Karen L. Knee<sup>1,2</sup> (ORCID: 0000-0002-5784-3434)

<sup>1</sup> Smithsonian Environmental Research Center, Edgewater, MD 20754

<sup>2</sup> Department of Environmental Science, American University, Washington, DC 20016

\* Corresponding author

Author for Correspondence:

J. Patrick Megonigal

Voice: 443-482-2346

Email: [megonigalp@si.edu](mailto:megonigalp@si.edu)

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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  $^{222}\text{Rn}$  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 CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O, 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 <sup>238</sup>U decay series generated by  $\alpha$ -decay of <sup>226</sup>Ra, 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  $^{222}\text{Rn}$ 's parent element,  $^{226}\text{Ra}$ , from soils (Simon and Ibrahim 1990) and  
67  $^{222}\text{Rn}$  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  $\text{CO}_2$  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  $\text{CO}_2$  (Teskey *et al.*, 2008)  
77 and  $\text{O}_2$  (Sorz 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  $\text{CO}_2$  (Reichstein *et al.*, 2012); stems are a poorly quantified  $\text{CH}_4$  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);  $\text{O}_2$

87 transport regulates the metabolism of tree stems; and radon emissions influence cluster ion and  
88 aerosol formation through alpha decay of  $^{222}\text{Rn}$  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  $^{222}\text{Rn}$   
91 produced in wood versus soil, (iii) relate tree stem emission rates of radon,  $\text{CO}_2$  and  $\text{CH}_4$ , 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 m<sup>2</sup> 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 m<sup>2</sup>) 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 m<sup>2</sup>) 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<sup>-3</sup>. 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 <sup>222</sup>Rn 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 CO<sub>2</sub> and CH<sub>4</sub>  
134 flux measurements which ran at 2-hour intervals before and after the radon flux measurement.  
135 Whenever possible, radon fluxes were compared to CO<sub>2</sub> and CH<sub>4</sub> fluxes measured on the same  
136 day. On two dates there were no usable CO<sub>2</sub> and CH<sub>4</sub> flux data to match the radon flux from an  
137 individual tree, so CO<sub>2</sub> and CH<sub>4</sub> 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 <sup>222</sup>Rn (Bq m<sup>-3</sup> water) in a 2-L sample was  
143 measured with the RAD7 attached to a RAD-H<sub>2</sub>O 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<sup>-2</sup> s<sup>-1</sup>,  $S_{Rn}$  is the slope of radon activity versus time in Bq m<sup>-3</sup> s<sup>-1</sup>,  $V$  is  
151 chamber volume in m<sup>3</sup>, and  $A$  is chamber area in m<sup>2</sup>. 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,  $\text{CO}_2$ , and  $\text{CH}_4$  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,  $\text{CO}_2$  and  $\text{CH}_4$  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,  $\text{CH}_4$ , and  $\text{CO}_2$  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  $^{222}\text{Rn}$  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  $^{222}\text{Rn}$   
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  $^{226}\text{Ra}$  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 ( $2.5 \pm 2.0$  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 O<sub>2</sub> (Soriz and Hietz, 2006; Mugnai and Mancuso,  
226 2010) and CO<sub>2</sub> (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; Soriz and Hietz, 2006). For example, O<sub>2</sub> diffusion rates through wood decrease  
243 in the order: diffuse-porous hardwoods > ring-porous hardwoods > gymnosperms (Soriz 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 CO<sub>2</sub> and CH<sub>4</sub>.

250 Radon emissions from *L. tulipifera* stems were significantly related to emissions of CO<sub>2</sub>  
251 ( $P=0.037$ ) and CH<sub>4</sub> ( $P=0.048$ ) (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 CO<sub>2</sub> and CH<sub>4</sub> 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 CO<sub>2</sub> and CH<sub>4</sub> 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 CO<sub>2</sub> or  
260 CH<sub>4</sub> 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<sup>-2</sup> soil surface s<sup>-1</sup>, 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. KLK provided  
292 the RAD7 instrument, radon measurement expertise, and groundwater radon measurements. PEB  
293 and KLK contributed to the experimental design and edited the manuscript.

294

#### 295 **Literature Cited**

296

297 **Barba J, Bradford, MA, Brewer PE, Bruhn D, Covey K, van Haren J., Megonigal JP,**  
298 **Mikkelsen TN, Pangala SR, Pihlatie M, Poulter B, Rivas-Ubach A, Schadt CW, Terazawa**  
299 **K, Warner DL, Zhang Z, Vargas R. 2019.** Methane emissions from tree stems: a new frontier  
300 in the global carbon cycle. *New Phytologist* **222**: 18-28.

301

302 **Carlquist S. 2001.** *Comparative wood anatomy: Systematic, ecological and evolutionary aspects*  
303 *of dicotyledon wood.* New York, NY, USA: Springer-Verlag Berlin Heidelberg.

304

305 **Covey KR, Megonigal JP. 2019.** Methane Production and Emissions in Trees and Forests. *New*  
306 *Phytologist* **222**: 35-51.

307

308 **de Farias EEG, da Silva Neto PC, de Souza EM, De Franca EJ, Hazin CA. 2016.** Radon  
309 levels and transport parameters in Atlantic Forest soils. *Journal of Radioanalytical Nuclear*  
310 *Chemistry* **307**: 811-815.

311

312 **Franks PJ, Bonan GB, Berry JA, Lombardozzi DL, Holbrook NM, Herold N, Oleson KW.**  
313 **2018.** Comparing optimal and empirical stomatal conductance models for application in Earth  
314 system models. *Global Change Biology* **24**: 5708– 5723.

315

316 **Garnet KN, Megonigal JP, Litchfield C, Taylor GE. 2005.** Physiological control of leaf  
317 methane emission from wetland plants. *Aquatic Botany* **81**: 141-155.

318

319 **Gartner BL, Moore JR, Gardiner BA. 2004.** Gas in stems: abundance and potential  
320 consequences for tree biomechanics. *Tree Physiology* **24**: 1239-1250.

321

322 **Große W, Bauch C. 1991.** Gas transfer in floating-leaved plants. *Vegetatio* **97**: 185-192.

323

324 **Hölttä T, Kolari P. 2009.** Interpretation of stem CO<sub>2</sub> efflux measurements. *Tree Physiology*, **29**:  
325 1447-1456.

326

327 **Jayaratne ER, Ling X, Morawska L. 2011.** Role of vegetation in enhancing radon  
328 concentration and ion production in the atmosphere. *Environmental Science & Technology* **45**:  
329 6350-6355.

330

331 **Langendörfer U, Cuntz M, Ciais P, Peylin P, Bariac T, Milyukova I, Kolle O, Naegler T,**  
332 **Levin I. 2002.** Modelling of biospheric CO<sub>2</sub> gross fluxes via oxygen isotopes in a spruce forest  
333 canopy: a <sup>222</sup>Rn calibrated box model approach. *Tellus B: Chemical and Physical Meteorology*  
334 **54**: 476-496.

335

336 **Levin I, Born M, Cuntz M, Langendörfer U, Mantsch S, Naegler T, Schmidt M, Varlagin**  
337 **A, Verclas S, Wagenbach D. 2002.** Observations of atmospheric variability and soil exhalation  
338 rate of radon-222 at a Russian forest site. Technical approach and deployment for boundary layer  
339 studies. *Tellus B: Chemical and Physical Meteorology* **54**: 462-475.

340

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

343

344 **Martens CS, Shay TJ, Mendlovitz HP, Matross, DM, Saleska SR, Wofsy SC, Woodward**  
345 **WS, Menton MC, De Moura JMS, Crill PM, De Moraes OLL & Lima RL. 2004.** Radon  
346 fluxes in tropical forest ecosystems of Brazilian Amazonia: Night-time CO<sub>2</sub> net ecosystem  
347 exchange derived from radon and eddy covariance methods. *Global Change Biology* **10**: 618–  
348 629.

349

350 **Mugnai S, Mancuso S. 2010.** Oxygen transport in the sapwood of trees. In: Mancuso S, Shabala  
351 S, eds. *Waterlogging signalling and tolerance in plants*. Berlin, Heidelberg, Germany: Springer  
352 Berlin Heidelberg, 61-75.

353

354 **Pitz SA, Megonigal JP. 2017.** Temperate forest methane sink diminished by tree emissions.  
355 *New Phytologist* **214**: 1432-1439.

356

357 **Pitz SL, Megonigal JP, Chang C-H, Szlavecz K. 2018.** Methane fluxes from tree stems and  
358 soils along a habitat gradient. *Biogeochemistry* **137**: 307-320.

359

360 **Reichstein M, Stoy PC, Desai AR, Lasslop G, Richardson AD. 2012.** Partitioning of net  
361 fluxes. In: Aubinet M, Vesala T, Papale D, eds. *Eddy covariance: a practical guide to*  
362 *measurement and data analysis*. Dordrecht, Netherlands: Springer Atmospheric Sciences, 263-  
363 289.

364

365 **Rissanen K., Hölttä T, Bäck J. 2018.** Transpiration directly regulates the emissions of water-  
366 soluble short-chained OVOCs. *Plant, Cell & Environment*, **41**: 2288-2298.

367

368 **Rusch H, Renneberg H. 1998.** Black alder (*Alnus Glutinosa* (L.) Gaertn.) trees mediate  
369 methane and nitrous oxide emission from the soil to the atmosphere. *Plant & Soil* **201**: 1-7.

370

371 **Schery SD, Gaeddert DH & Wilkening MH. 1984.** Factors affecting exhalation of radon from  
372 a gravelly sandy loam. *Journal of Geophysical Research* **89**: 7299-7309.

373

374 **Siegenthaler A, Welch B, Pangala SR, Peacock M, Gauci V. 2016.** Technical Note: Semi-  
375 rigid chambers for methane gas flux measurements on tree stems. *Biogeosciences* **13**: 1197-  
376 1207.

377

378 **Sorz J, Hietz P. 2006.** Gas diffusion through wood: implications for oxygen supply. *Trees* **20**:  
379 34-41.

380

381 **Spicer R & Holbrook NM. 2005.** Within-stem oxygen concentration and sap flow in four  
382 temperate tree species: does long-lived xylem parenchyma experience hypoxia? *Plant, Cell &*  
383 *Environment* **28**: 192-201.

384

385 **Steppe K, Sterck F & Deslaurier A. 2015.** Diel growth dynamics in tree stems: linking  
386 anatomy and ecophysiology. *Trends in Plant Science* **20**: 1360-1385.

387

388 **Teskey RO, Saveyn A, Steppe K, McGuire MA. 2008.** Origin, fate and significance of CO<sub>2</sub> in  
389 tree stems. *New Phytologist* **177**: 7-32.

390

391 **Trumbore SE, Keller M, Wofsy SC & da Costa JM. 1990.** Measurements of soil and canopy  
392 exchange rates in the Amazon rain forest using <sup>222</sup>Rn. *Journal of Geophysical Research* **95**:  
393 16,865-16,873.

394

395 **Ussler WI, Chanton JP, Kelley CA, Martens CS. 1994.** Radon 222 tracing of soil and forest  
396 canopy trace gas exchange in an open canopy boreal forest. *Journal of Geophysical Research* **99**:  
397 1953-1963.

398

399 **Wanty RB, Johnson SL, Briggs PH. 1991.** Radon-222 and its parent radionuclides in  
400 groundwater from two study areas in New Jersey and Maryland, U.S.A.". *Applied Geochemistry*  
401 **6**: 305-318.

402

403 **Yesilonis I, Szlavecz K, Pouyat R, Whigham D, Xia L. 2016.** Historical land use and stand age  
404 effects on forest soil properties in the Mid-Atlantic U.S. *Forest Ecology and Management* **370**:  
405 83-92.

406

## 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  $^{222}\text{Rn}$  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  $^{222}\text{Rn}$  activity,  $\text{CO}_2$  concentration, and  $\text{CH}_4$  concentration in  
417 unsaturated soil to 80 cm, and  $^{222}\text{Rn}$  in groundwater at approximately 300 cm. Points are means  
418  $\pm 1$  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  $\text{Bq m}^{-3}$  water. Groundwater concentrations of  $\text{CO}_2$  and  
420  $\text{CH}_4$  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  $2.5 \pm 2.0$   $\text{mBq m}^{-2}$  stem (means  $\pm 1$  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  $^{222}\text{Rn}$  and  $\text{CO}_2$  (upper panel) and  $^{222}\text{Rn}$  and  
432  $\text{CH}_4$  (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.

[CO<sub>2</sub>] (%) and [CH<sub>4</sub>] (ppm)







