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Radon 222 tracing of soil and forest canopy trace gas exchange in an open canopy boreal forest

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A set of continuous, high-resolution atmospheric radon (^{222}Rn) concentration time series and radon soil flux measurements were acquired during the summer of 1990 at a micrometeorological tower site 13 km northwest of Schefferville, Quebec, Canada. The tower was located in a dry upland, open-canopy lichen-spruce woodland. For the period July 23 to August 1, 1990, the mean radon soil flux was 41.1 ± 4.8 Bq m^{-2} h^{-1}. Radon surface flux from the two end-member forest floor cover types (lichen mat and bare soil) were 38.8 ± 5.1 and 61.8 ± 15.6 Bq m^{-2} h^{-1}, respectively. Average total forest canopy resistances computed using a simple "flux box" model for radon exchange between the forest canopy and the overlying atmosphere range from 0.47 ± 0.24 s cm^{-1} to 2.65 ± 1.61 s cm^{-1} for daytime hours (0900–1700 LT) and from 3.44 ± 0.91 s cm^{-1} to 10.55 ± 7.16 s cm^{-1} for nighttime hours (2000–0600) for the period July 23 to August 6, 1990. Continuous radon profiling of canopy atmospheres is a suitable approach for determining rates of biosphere/atmosphere trace gas exchange for remote field sites where daily equipment maintenance is not possible.

Introduction

The rate of trace gas exchange between the terrestrial biosphere and the overlying atmosphere controls important aspects of tropospheric chemistry and the oxidative balance of the atmosphere [e.g., *Mooney et al.*, 1987]. Accordingly, a series of recent field experiments (e.g., Global Tropospheric Experiment/Arctic Boundary Layer Expedition or GTE/ABLE [*Harriss et al.*, 1990]) have included an improved understanding of surface exchange processes as part of their goals. In this paper we present the results of a forest canopy radon transport experiment conducted during the GTE/ABLE 3B field experiment near Schefferville, Quebec, Canada, in the summer of 1990 that was designed to directly measure biosphere/atmosphere trace gas exchange rates.

Because most trace gases emitted by the biosphere are either biogenically or photochemically reactive, they are potentially unsuitable tracers for trace gas transport within and out of a vegetated land surface. However, ^{222}Rn, a radioactive noble gas, is ideally suited as a tracer of gas transport in the lower atmosphere for the following reasons: (1) it is emitted almost exclusively by the soil, and the land surface can be modeled as a planar source for radon; (2) the only sink for radon is its radioactive decay, which can be accurately quantified using its radioactive decay constant; (3) it is a chemically inert gas, making it suitable for tracing physical exchange of reactive trace gases between the biosphere and the overlying atmosphere; and (4) near-surface concentrations are easily measured by existing technology.

A large effort to employ radon as a tracer of near-surface meteorological processes was made during the 1960s and early 1970s [e.g., *Moses et al.*, 1960; *Pearson and Jones*, 1965; *Birot et al.*, 1970; *Larson and Hoppel*, 1973; *Li*, 1974]. Turbulent eddy diffusion coefficients computed by these and other investigators have formed the basis for a better parameterization of surface exchange rates used by three-dimensional general circulation models [e.g., *Liu and McAfee*, 1984; *Jacob and Prather*, 1990]. However, application of radon to the measurement of biosphere/atmosphere trace gas exchange was not made until the work by *Trumbore et al.* [1990]. They showed for a closed-canopy tropical rain forest that radon-derived trace gas exchange rates determined for five brief nocturnal periods compared favorably with simultaneous, independent estimates obtained by eddy correlation techniques [*Fan et al.*, 1990; *Fitzjarrald et al.*, 1990]. Thus radon was shown to have the potential to provide an independent and reliable estimate of the rates at which trace gases are exchanged between a forest canopy and the overlying atmosphere. In this paper we present a radon data set for an open-canopy boreal forest which was collected continuously over a period of 2 weeks at a time resolution of 30 min. This data set makes it possible to observe directly how biosphere/atmosphere trace gas exchange rates and forest canopy resistance values vary over daily as well as longer time periods.

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METHODS

Site Characteristics

The radon measurements were conducted at the NASA ABLE 3B forest site 13 km northwest of Schefferville, Quebec, Canada (54°50'N, 66°40'W, 500 m above sea level), during the summer of 1990. The study site was located on a dry upland in a mature, open-canopy lichen-spruce woodland comprised almost exclusively of black spruce (*Picea mariana*). Mean canopy height was 5 to 6 m above the forest floor and canopy density was approximately 600 trees ha^{-1} (P.S. Bakwin, personal communication, 1993). The forest floor was carpeted with a 5- to 10-cm-thick lichen mat dominated by *Cladina stellaris*, *Cladonia* spp. Unvegetated soil occurred either as bare patches or as narrow, polygonal networks of cracks that separated large patches of the lichen mat. This bare soil surface was approximately 10% of the forest floor surface area. Woody understory shrubs (e.g., *Betula glandulosa*, *Ledum groenlandicum*, *Vaccinium* spp.) and immature black spruce were sparsely distributed and were often localized along the network of cracks developed in the dry lichen mat.

Soils at the study site are developed on a thin glacial till veneer deposited during the Wisconsin glaciation and are generally acidic and nutrient poor. The region around Schefferville was one of the last to become ice free, about 7000 years B.P. [*Prest*, 1973; *Mayewski et al.*, 1981]. Thus the soils are relatively young compared to the rest of North America.

Atmospheric Radon Concentrations

Atmospheric gas was sampled continuously at four heights above the local land surface. Two gas intakes were located on a 31-m micrometeorological tower (9.5-m and 18.2-m heights) and the remaining gas intakes were in close proximity to the tower base (1.0-m and at three 0.05-m heights located over bare soil and lichen cover). The 0.05-m sites were selected to represent the range of ground cover type present on the forest floor. Gas samples were pumped continuously at 1 L min^{-1} through 0.95-cm OD polyethylene tubes from the sample points to individual flow-through radon scintillation detectors located in the main instrument tent 20 m southeast of the tower base. Entrainment of raindrops was prevented by attaching 8-cm-diameter plastic funnels to the sample inlet tubes and hanging them in an inverted position. Individual gas streams were dried to constant dew point (-73 °C) using an in-line drying column containing approximately 0.6-kg 8-mesh drierite (changed daily) and passed through a 25-mm in-line filter (5-μm PVC membrane filter supported by a coarse nitrocellulose filter) just before entering a scintillation detector. This in-line filter removed unwanted radon (^{222}Rn) and thoron (^{220}Rn) daughter contamination from the sample stream. Bias of near-surface ^{222}Rn activity measurements by accumulation of the ^{220}Rn ($t_{1/2}$ = 55 s) daughters in the flow-through scintillation detectors is unlikely for two reasons: (1) sampling delay between the nearest sample location and the scintillation detector would have resulted in at least a 75% reduction in any thoron activity by the time the gas sample reached the detector and (2) repeated field site measurements of detector background activity begun immediately after removing the detector from the gas sample stream have shown that initial background readings are within the range of random variability for the entire background experiment and for laboratory background measurements made after allowing the detector to be sealed for a period exceeding 1 month.

Raw counts resulting from radioactive decay of radon and its daughters within the flow-through scintillation counter were collected using a dedicated portable computer and were processed further upon completion of the experiment. Raw counts were lumped into 30-min blocks, background corrected, and converted to Bq m^{-3} using a calibration factor specific to each scintillation counter. Scintillation counter calibration was obtained using a ^{222}Rn gas source of known activity in our laboratories at the University of North Carolina before and after the experiment. Scintillation counter background measurements were obtained periodically at the field site using continuously flowing dry nitrogen gas (1 L min^{-1}) at 1 atm pressure. Radon activity is assumed to follow a Poisson distribution, and unless otherwise indicated, we report the random error associated with each measurement as a proportion of the square root of the total number of raw counts for that particular counting interval ($N^{0.5}$ [*Young*, 1962]). Geometric propagation of the random errors associated with counting statistics, background determination, and calibration factors [*Shoemaker et al.*, 1981, pp. 46–50] on selected subsamples has shown that most of the random error is a result of Poisson counting statistics. Systematic errors, based on repeated field site intercalibration of the scintillation counters using a common source of dry ambient air, are on average less than 9%.

Radon Soil Flux

Radon soil flux was measured at two sites representative of the end-member forest floor cover types (lichen mat and bare soil) in close proximity to the base of the micrometeorological tower. A chamber-enclosure technique similar to that of *Crill* [1988] was employed. A welded aluminum collar with inside dimensions of 0.66 m x 0.66 m was emplaced at each of the two sites at the beginning of the field season. The collars penetrated the soil to a depth of approximately 6 cm. At the beginning of each flux measurement a 91.5-L welded aluminum chamber was placed over the flux collar and clamped tight. An airtight seal was made with a 6-mm closed-cell foam gasket attached to the chamber. Heating effects caused by direct sunlight were minimized by covering the chamber and associated sampling equipment with a highly reflective aluminized mylar blanket. Chamber air was circulated through a closed sampling loop which included, in the direction of flow, a stainless steel bellows pump operated at 4 L min^{-1}, a flowmeter, a drierite column, in-line filter, and a 120-cm^3 scintillation cell [*Lucas*, 1957]. Each scintillation cell was attached to the sampling loop for at least 10 min to ensure uniform sampling of chamber air. From three to six chamber air samples were obtained for each flux experiment. The maximum time duration for any one flux experiment did not exceed 100 min. After sampling, the scintillation cells were held for 3 hours before scintillation counting to allow radon and its daughters to come into secular equilibrium.

An estimate of the radon soil flux for each flux experiment was obtained from a linear regression of decay-corrected radon activity against elapsed time. The increase of radon activity was linear over the course of each soil flux experiment. Table 1 lists the correlation coefficient (r) of the linear regression for each flux measurement and the number of chamber air samples used in the regression (n). Except

TABLE 1. Summary of Measured Radon Soil Flux

Day	Site	Conditions	Radon Flux, Bq m^{-2} h^{-1}	r*	n†
204	bare soil	dry/sunny	67.9 ± 9.9‡	0.980	4
205	lichen mat	dry/sunny	43.7 ± 5.2	0.993	3
206	lichen mat	dry/partly cloudy	45.4 ± 0.7	0.999	4
207	bare soil	windy/cool; rained previous afternoon	40.4 ± 1.8	0.998	4
207	lichen mat	windy/cool; rained previous afternoon	33.4 ± 6.3	0.965	4
211	bare soil	surface dry; drizzled after chamber was closed	77.1 ± 10.2	0.991	3
212	lichen mat	rain	38.5 ± 2.1	0.994	6
213	lichen mat	sunny; lichen wet and supple	32.9 ± 1.9	0.998	3

*Correlation coefficient of the linear regression.

†Number of samples in linear regression.

‡Standard error, estimated from linear regression of measured radon concentration as a function of time.

for two flux determinations (days 205 and 211, 93% and 92% confidence levels, respectively) the correlation coefficient of the linear regression was greater than the 95% confidence level (n=3, r=0.997; n=4, r=0.950; n=5, r=0.878; and n=6, r=0.811 [*Young*, 1962]), which is generally accepted as an adequate indication of a linear fit to the data [*Young*, 1962, p. 132. The error associated with each flux measurement is reported as the standard error of the regression coefficient for the slope. The flux values we report are not necessarily representative of the full range of spatial and temporal variability at this study site. Thus the errors reported in Table 1 represent the error associated with the individual radon flux measurement and are not a measure of the spatial variability that may exist at this site. Limited time and resources required that we devote our efforts toward quantifying the short-term temporal variability and the effect of changing moisture conditions at two well-characterized sites.

In contrast with published studies of radon accumulator methods [e.g., *Kearney and Krueger*, 1987] which have measured radon concentration increases over significantly longer timescales (up to 24 hours), we did not experience difficulty obtaining statistically acceptable linear increases of radon concentration with time. The linear fits that we obtained suggest that changes in temperature, atmospheric pressure, and bioactivity were minimal during our relatively short sampling period (<100 min). In addition, we conclude that diffusion of radon back into the soil as well as a reduction in radon soil flux caused by a decrease in the concentration difference between the upper soil layers and the chamber air are relatively unimportant factors affecting the results of our chamber flux measurements. The effect of surface wind on our flux measurements was not significant for two reasons: (1) nearly calm conditions prevailed during all of the radon flux measurements and (2) the chamber/collar seal was made with a closed-cell foam gasket and the collar penetrated approximately 6 cm into the soil. Our chamber technique has been tested recently in a series of experiments in Florida soils where this technique (as described above) has compared favorably (less than 15% difference, n=4 independent replicated comparisons) with an independent soil flux measurement technique utilizing charcoal canisters calibrated with radon emissions from a barium palmitate source (W.C. Burnett, P.E. Cable, and J.P. Chanton, unpublished manuscript, 1993). We conclude that by using a fixed collar/chamber configuration and by measuring radon concentration increase over very short time periods, reliable estimates of radon soil flux can be obtained.

Calibration of Lucas Scintillation Cells

A set of 120-cm^3 lucite scintillation cells coated with Ag-activated ZnS [*Lucas*, 1957] were utilized for the radon soil flux measurements. These cells were equipped with two Swagelok quick connectors, which permitted flow-through operation during radon soil flux measurements described in the previous section. Cell efficiencies were determined at 1 atm pressure by connecting all cells in series with a 20-L carboy containing a small amount of indicating drierite, a stainless steel bellows pump, a flowmeter, and a calibrated radon source. This closed system was circulated for 4 hours at 4 L min^{-1}, and ^{222}Rn activity was measured for each cell by scintillation counting. Cell backgrounds were determined at 1 atm using dry compressed air aged for at least 1 month before use. Cell efficiencies ranged between 72 and 91%, and cell backgrounds were typically between 4.9 and 8.5 mBq per cell.

RESULTS AND DISCUSSION

Radon Soil Flux

Radon soil flux measurements obtained over the period July 23 (day 204) to August 1, 1990 (day 213), are illustrated in Figure 1 and summarized in Table 1. Average values for the two end-member cover types of the forest floor, bare soil and lichen, are 61.8 ± 15.6 Bq m^{-2} h^{-1} (n=3, ±1σ) and 38.8 ± 5.1 Bq m^{-2} h^{-1} (n=5, ±1σ), respectively. Using the estimate that 10% of the forest floor is bare soil and that the remaining area is covered with a uniform lichen mat, the area-weighted average radon soil flux is 41.1 ± 4.8 Bq m^{-2} h^{-1} (n=8, ±1σ). This value is within the range of published radon soil flux measurements but is less than estimates of average global radon soil flux (Table 2).

Except for one measurement (day 207, bare soil) we did not observe any significant short-term variation in radon soil flux at either site. The one environmental factor which we believe may have contributed most to the decrease in radon flux over the bare soil site but not over the lichen site on day 207 is rainfall (Figure 1). The capping effect of mois-

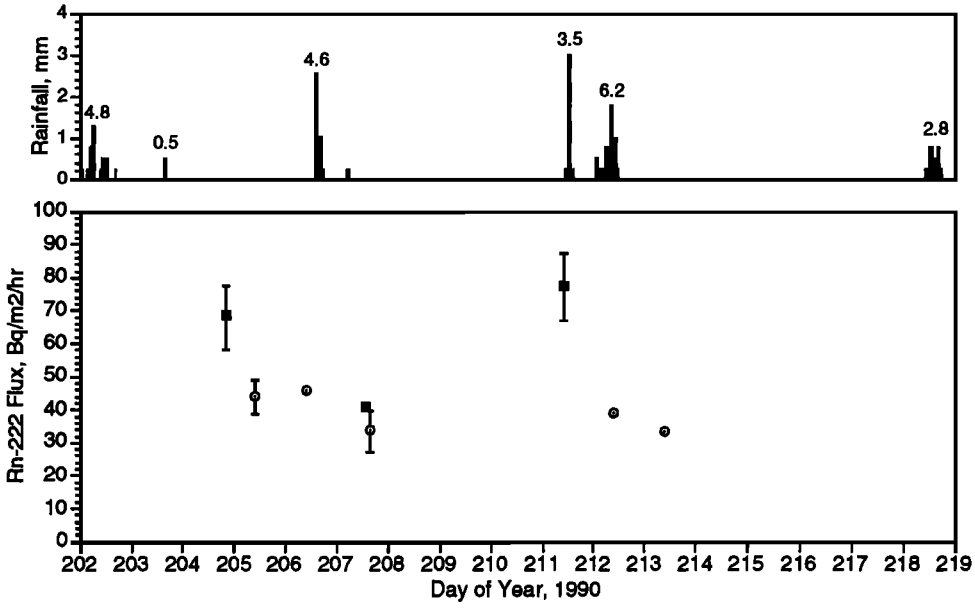

Fig. 1. Bottom panel summarizes the radon soil flux measurements obtained at two sites near the base of the micrometeorological tower. The bare soil site (solid squares) and the lichen mat site (open squares) are representative end-members of the range of forest floor cover type. The radon flux measurements are plotted at the time at which the flux chamber was sealed. Error bars represent the standard error estimated from the linear regression of measured radon concentration as a function of time. In some cases the reported error is less than the size of symbol plotted. Note that on days 206 and 211 the flux chamber was sealed just prior to the beginning of rainfall. In the top panel rainfall measurements obtained by a surface weather station (portable automated mesonet (PAM) *Brock et al.*, [1986]) are plotted in 1-hour time increments (East Valley PAM Station 1, 1 km from site [*Fitzjarrald and Moore*, this issue]). The number above each cluster of rainfall data is the total rainfall for that storm event (millimeter/event); 1 dpm ^{222}Rn = 1 disintegration per minute = 0.017 Bq = 0.45 pCi = 7900 atoms ^{222}Rn.

TABLE 2. Comparison of Published Radon Soil Flux Measurements and Estimates of Global Averages

Investigator	Location	Radon Flux, Bq m^{-2} h^{-1}
Trumbore et al. [1990]	Amazon rain forest, average of all sites	26.4 ± 3.8
Dorr and Munnich [1990]	Germany	
	sandy soil	17 to 33
	loamy and clayey soil	67 to 100
Schery and Siegel [1986]	New Mexico, gravelly sand loam	64 ± 8
Nazaroff [1992]	undisturbed soils	54 to 173
Wilkening et al. [1974]	estimate of global average	57
Turekian et al. [1977]	estimate of global average	91
This study	Quebec, subarctic forest soil	41.1 ± 4.8

ture at the soil surface has been documented with measurements of increased soil radon gas concentrations following measureable rainfall [e.g., *Kovach*, 1945; *Schery et al.*, 1984; *Asher-Bolinder et al.*, 1990]. Although none of these studies measured radon soil flux directly, the observed increases in soil radon concentration imply that radon soil flux was correspondingly reduced. We speculate that on day 207 rainfall occluded soil surface pore space enough to temporarily reduce radon soil flux at the bare soil site, whereas rainfall was insufficient to occlude porous lichen at the adjacent lichen mat site. This suggestion and the limited and sometimes conflicting observations of the effect of rainfall on radon soil flux published in the literature [e.g., *Pearson*, 1967; *Guedalia et al.*, 1970; *Tanner*, 1980; *Schery et al.*, 1984; *Stranden et al.*, 1984] indicate the need for a thorough investigation of the effects of rainfall on the exchange of radon and other trace gases between the soil and the atmosphere. In a later section we will discuss the effects that rainfall may have had on near-surface atmospheric radon concentrations.

Atmospheric Radon Concentration Time Series

Figure 2 summarizes a subset of the atmospheric radon concentration data obtained for the five sample locations on and adjacent to the micrometeorological tower which we will use in the next section for the purpose of characterizing how spatially averaged forest canopy/atmosphere exchange rates vary with time of day. In a subsequent paper (W. Ussler et al., manuscript in preparation, 1993) we will examine how vertical radon profiles and exchange rates within the canopy vary over the growing season. The time series in Figure 2 display a distinctive 24-hour periodicity in atmospheric radon concentration which typically reaches a maximum at approximately midnight local time. The maxima are shifted slightly forward in time at increasing heights above the ground, reflecting the upward increasing stability of the growing nocturnal boundary layer. Nocturnal increases in radon concentration are most pronounced near the forest floor and result from the accumulation of radon

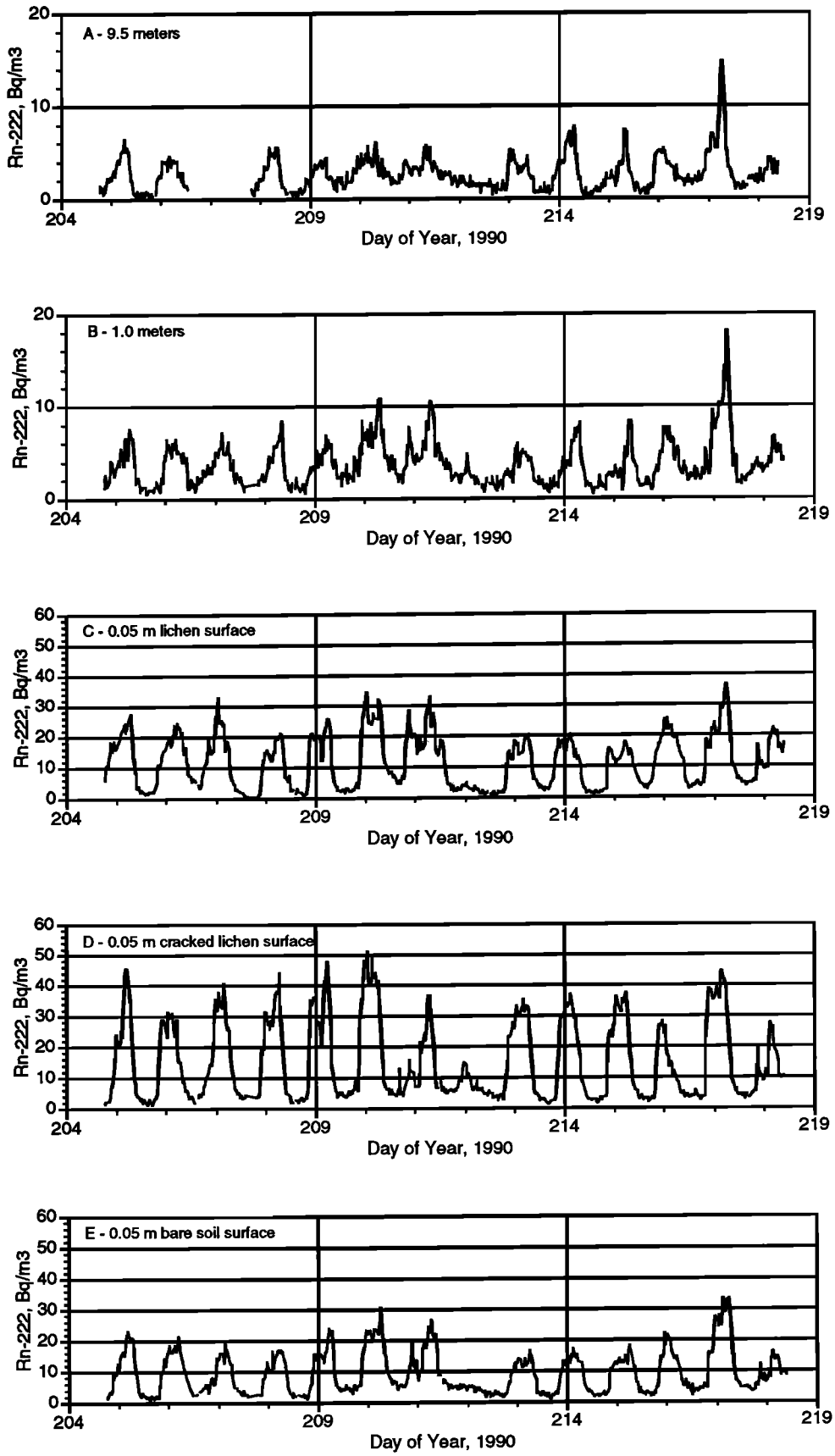

Fig. 2. Subset of the atmospheric radon concentration time series obtained at specific heights above the forest floor: (*a*) 9.5 m, (*b*) 1 m, (*c*) 0.05 m above a lichen surface, (*d*) 0.05 m above a cracked lichen surface, and (*e*) 0.05 m above a bare soil surface. Distinct nocturnal peaks in radon concentration occur regularly at all levels. The occasional absence of a large accumulation of radon at ground level (*c*, *d*, and *e*) is most often the result of a rainfall event. Data are reported on the basis of local time, which is 4 hours behind Greenwich mean time.

emitted by the soil into the base of the nocturnal boundary layer. The sharp decrease in radon concentration at all canopy heights is nearly simultaneous and is the result of the onset of thermal convection during early morning hours. Sunrise occurs at approximately 0430 LT during this time of year. Numerous studies of near-surface atmospheric radon concentrations have noted similar diel variations which result from the growth and decay of the nocturnal boundary layer [e.g., *Moses et al.*, 1960; *Fontan et al.*, 1966; *Israel et al.*, 1966; *Servant*, 1966; *Pearson*, 1967; *Li*, 1974].

Spatial variability in near-surface radon concentration (0.05-m sites, Figure 2) cannot be explained simply on the basis of differences in surface flux. Nocturnal radon concentrations are systematically higher over the lichen surface (Figure 2*c*) compared to the bare soil surface (Figure 2*e*). The highest radon concentrations occur over cracks in the lichen surface (Figure 2*d*). If flux controlled near-surface concentrations, then the lichen surface should have systematically lower radon concentrations compared to the bare soil surface, and the cracked lichen surface should have intermediate values. One possibility is that surface roughness may play a dominant role in determining near-surface radon concentration patterns by generating a roughness sublayer [*Monteith and Unsworth*, 1990]. Although the roughness sublayer is within the constant flux layer, modification of the turbulent structure by surface element drag creates the conditions necessary for storage of trace gases. Surfaces with greater roughness can cause storage of larger amounts of trace gases immediately above the surface. Qualitatively, this is what we see in Figures 2*c*, 2*d*, and 2*e*. Increasing near-surface radon concentration is correlated with increasing surface roughness (bare soil < lichen < cracked lichen).

The absence of a nocturnal peak in radon concentration is most often associated with measureable precipitation (wind effects were considered, but horizontal wind speed and near-surface radon concentration within the forest canopy were poorly correlated and often inconsistent). The best example of the effect of sustained rainfall on nocturnal radon accumulation which occurred earlier in the season is illustrated in Figure 3. As discussed in the section on soil radon flux, wetting of the soil surface may rapidly reduce radon soil flux by occluding soil pore space. Reduced radon soil flux will result in lower than normal nocturnal radon accumulation at ground level. A rainfall event beginning during the afternoon of day 211 and extending into day 212 also suppressed nocturnal radon accumulation (Figures 2*c*–2*e*). Suppression of nocturnal radon accumulation over the bare soil surface was more complete than over the lichen or cracked lichen surfaces. This observation is consistent with the speculation that bare soil pore spaces are more easily and uniformly occluded than the interstitial spaces in the lichen mat. In all cases, nocturnal radon concentrations return to near normal values within hours after cessation of rainfall. The rapid suppression and quick recovery of nocturnal atmospheric radon concentrations during rainfall events that we have observed are inconsistent with the conclusion obtained by *Jacob and Prather* [1990]. Using data from *George and Breslin* [1979] and *George* [1980; 1981], they concluded that elevated soil surface moisture has little effect on radon soil flux. Their conclusion is based on 3-day averages of both radon soil flux and precipitation. Although limited, our combined soil flux and near-surface atmospheric radon data suggest that substantial changes in radon soil flux may occur in response to rainfall over much shorter timescales, of the order of hours, not days.

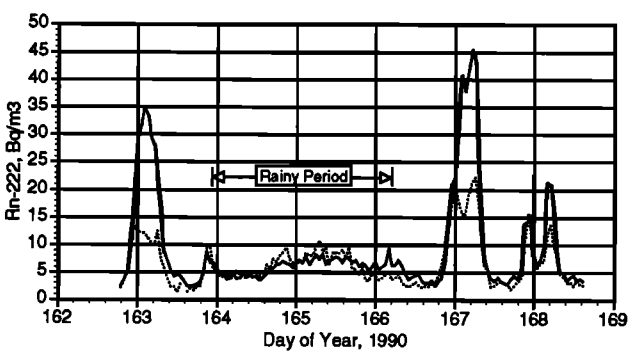

Fig. 3. An example of the effect of sustained, measureable rainfall on near-surface atmospheric radon concentrations over a cracked lichen surface (solid curve) and a lichen surface (dotted curve). Rainfall abruptly truncates the nocturnal rise in radon concentration above the lichen mat on day 163. Rain fell nearly continuously over the following 2 days before ending during the early morning hours of day 166 (total accumulation was at least 16.8 mm, measured at PAM Station 1, [*Firtzjarrald and Moore*, this issue]; missing rainfall data for days 163–164 makes this a minimum estimate). Rain has the effect of reducing radon soil flux, which results in lower than normal nocturnal radon accumulation at ground level. Note the substantially larger nocturnal radon peak on day 167. This increase is most likely due to release of radon accumulated in the soil during the rainy period.

Forest Canopy Trace Gas Exchange

The rate for trace gas exchange between a forest canopy atmosphere and the overlying atmosphere can be computed using a simple trace gas inventory model. This approach was first used by *Trumbore et al.* [1990] for quantifying radon exchange across a closed-canopy forest in the Amazon basin. This spatially averaged inventory model treats the forest as if it was enclosed by a "flux box" with a semipermeable top defined by the top of the canopy architecture (Figure 4). It assumes that net horizontal trace gas transport is negligible compared to net vertical transport. The balance between soil flux, changes in forest canopy inventory, sources and sinks within the forest canopy, and exchange between the forest canopy and the overlying atmosphere is expressed by

$$h\frac{\partial \overline{C}}{\partial t} = S - k(\overline{C} - C_t) + \int_0^h (P - L)\, dz \qquad (1)$$

where h is the canopy height, $\overline{C}$ is the spatial mean trace gas concentration within the canopy, S is the soil flux, k is a forest canopy trace gas exchange coefficient, C_t is the concentration of the trace gas in the overlying troposphere, and P and L are the production and loss, respectively, of the trace gas within the canopy. In practice, the term on the left-hand side of the equation is evaluated by comparing concentration profiles at successive time points [*Trumbore et al.*, 1990].

The importance of radon transport and emission by vascular plants has not been adequately determined and a wide range of estimates is reported in the literature [e.g., *Pearson and Jones*, 1966; *Mattsson*, 1970; *Kelley et al.*, 1991]. We assume, following the argument made by *Trumbore et al.* [1990], that evapotranspiration is not an important source for radon within the forest canopy. Loss of radon by radioactive decay is insignificant as well because the residence time

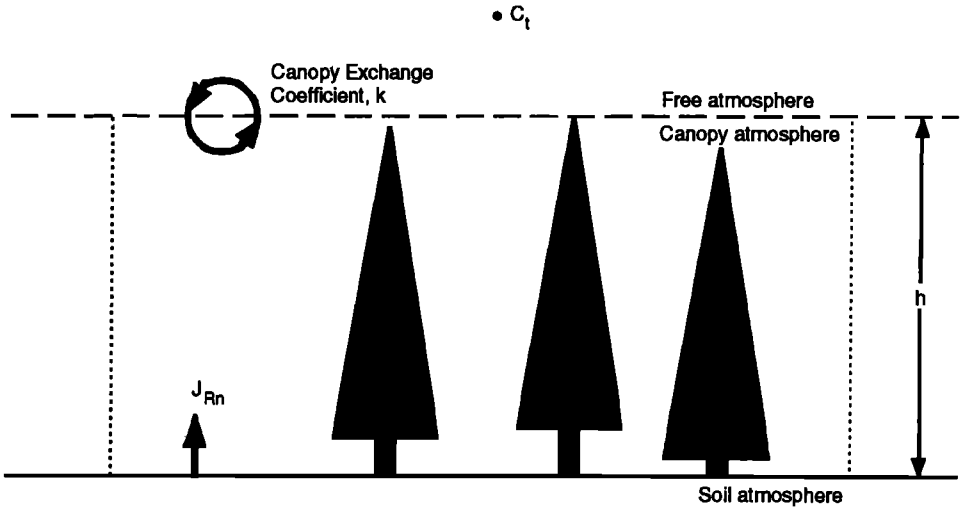

Fig. 4. A schematic representation of the canopy flux box inventory model used to compute forest canopy trace gas exchange rates. The top of the "flux box" is bounded by a plane defined by the mean height of the forest canopy (dashed line). The rate of trace gas exchange across this plane is expressed by the canopy exchange coefficient, k. Radon enters the box only from the soil surface (J_{Rn}). We assume that there is no net horizontal transport through the box walls, indicated by the dotted lines. C_t is the radon concentration measured at a fixed height above the trees. We have used the following conditions for the calculations discussed in the text: $h = 6$ m; C_t is the radon concentration time series measured at 9.5 m; the time interval between concentration profile measurements is 30 min; and J_{Rn} is an area-weighted average value computed from radon soil flux measurements (Table 1).

of radon within the canopy is much shorter than its 3.8 day half-life. For these reasons we assume that the integral term in equation (1) is negligible compared to the other terms and can be eliminated.

Implicit in the inventory model is the assumption that trace gas transport across the upper boundary of the forest canopy occurs down a concentration gradient. Although accumulating evidence suggests that countergradient transport is an important transport mechanism within forests [e.g., *Denmead and Bradley*, 1985] over relatively short timescales, of the order of minutes [*Gao et al.*, 1989], the inventory model, by virtue of its longer timescale of integration, can provide a time-averaged estimate of trace gas exchange. The inventory model records the effects of advective transport but does not require a complete physical description of the transport mechanisms.

Figure 5*a* illustrates how the total inventory of radon within the forest canopy varied through time for the period day 204 to day 218. A forest canopy trace gas exchange coefficient, k, has been computed at 30-min intervals using equation (1) and is plotted in Figure 5*b*. Nocturnal exchange rates are relatively uniform over nighttime hours and show little day-to-day variation. In contrast, daytime exchange rates are quite variable, on both hourly and day-to-day timescales. This variability is a direct consequence of the open canopy architecture of the black spruce forest. Large fluctuations in the surface heating of the open forest floor by incident solar radiation can occur as a result of changing cloud conditions. Therefore rates of convection-driven vertical exchange are highly variable.

Total Canopy Resistance of the Boreal Forest Canopy

Electrical resistance analogs have been broadly applied to the analysis of heat, moisture, momentum, and trace gas transport within and across plant canopies [e.g., *Monteith and Unsworth*, 1990; *Lhomme*, 1991; *Jones*, 1992]. The total forest canopy resistance (R_t) to the bidirectional exchange

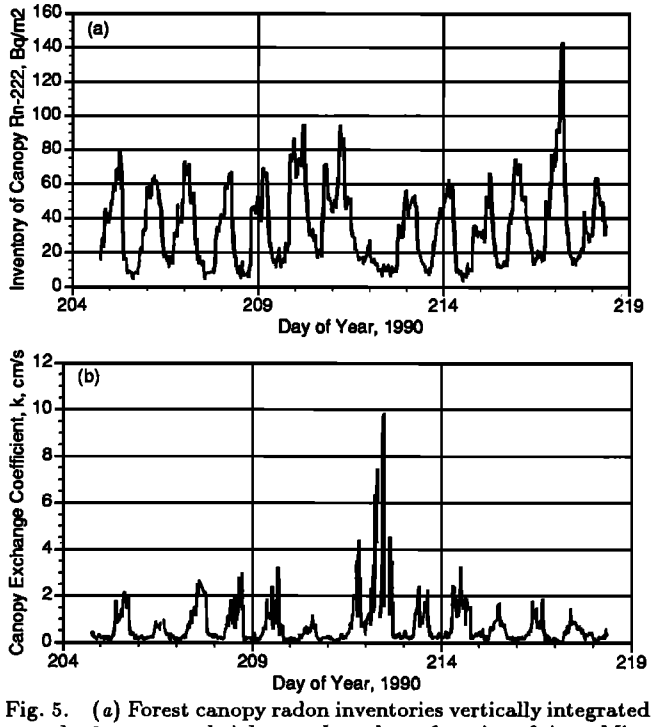

Fig. 5. (*a*) Forest canopy radon inventories vertically integrated over the 6-m canopy height are plotted as a function of time. Minimum daytime inventories are typically 10 Bq m^{-2}, while nocturnal maximum inventories can reach as high as 140 Bq m^{-2}. The radon inventory model can be extrapolated to any trace gas that is emitted at a constant rate by the soil into the forest canopy (e.g., CO_2 or N_2O). The same scale of inventory change for each gas will occur by turbulent transport alone. Additional inventory changes can result from sources or sinks of trace gas within the forest canopy. (*b*) Forest canopy trace gas exchange coefficients plotted as a function of time, computed using the "flux box" model for the forest canopy. Large short-duration spikes in the exchange coefficient during day 212 are the direct result of a significantly lower radon soil flux into the forest canopy during and shortly following a rainfall event. A mean soil radon flux value of 41.1 Bq m^{-2} h^{-1} was used to compute the exchange coefficient.

of any of these quantities between the soil and the overlying atmosphere can be computed from the sum of three key resistance elements, where

$$R_t = R_a + R_b + R_c \quad (2)$$

and R_a is the aerodynamic resistance to the transfer of momentum, R_b is the quasi-laminar boundary layer resistance, and R_c is the canopy resistance [*Hicks and Matt*, 1988]. The quasi-laminar boundary layer resistance is an "excess" resistance which accounts for the difference in how momentum, as opposed to mass and energy, is transferred to a surface [see *Baldocchi et al.*, 1987]. The canopy resistance is produced mainly by surfaces within the forest canopy and includes, among others, stomatal, cuticular, water film, and soil resistances. We will follow convention by using uppercase symbols for those resistances expressed as per unit horizontal area, as opposed to lowercase for the individual components specific to the surface elements [e.g., *Meyers and Baldocchi*, 1988].

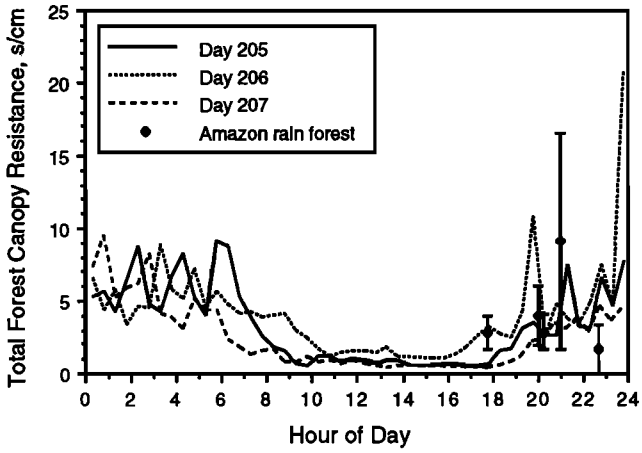

Fig. 6. Comparison of the total forest canopy resistance, R_t, computed from the reciprocal of the forest canopy trace gas exchange coefficient, k, for three sequential days (July 24 to July 26, 1990) plotted as a function of time of day (noon = 0.5). Five determinations of the total forest canopy resistance obtained by *Trumbore et al.* [1990] using an identical ^{222}Rn inventory model are included. The similarity in resistance values suggests that at least during nocturnal conditions, aerodynamic stability, rather than the vegetative canopy, is the factor controlling the rate of vertical trace gas transport in forest ecosystems. Propagation of errors associated with the computation of k using equation (1) [*Shoemaker et al.*, 1981] results in an estimate of random error of approximately 22% for both midday and nocturnal values of R_t. The largest contribution to this error results from the uncertainty associated with the measurement of atmospheric radon concentration.

The total forest canopy resistance for the transfer of radon from the soil to the overlying atmosphere can be computed from the reciprocal of the forest canopy trace gas exchange coefficient, k, obtained in the previous section. Comparison of this total forest canopy resistance (R_t) over the course of selected days with other published values for R_t are plotted in Figure 6. Mean values of R_t for selected daytime and nighttime periods for each day from the period day 204 to day 218 are summarized in Table 3. Although the total forest canopy resistance values obtained for the boreal forest and the Amazon rain forest are similar, trace gas flushing times (R_t x h) are quite different because of the differences in canopy architecture. For example, in the boreal forest with a total forest canopy resistance of 4 s cm^{-1} and canopy height of 6 m, the flushing time is 40 min. Whereas in the rain forest, where canopy height was 41 m [*Trumbore et al.*, 1990], a flushing time of roughly 270 min is obtained for the same total forest canopy resistance.

Within-Canopy Resistance to Trace Gas Transport

The within-canopy resistance ($R_b + R_c$) can be estimated from the difference between the total forest canopy resistance obtained from the radon inventory model and the aerodynamic resistance computed from simultaneous micrometeorological measurements. Aerodynamic resistances were computed from measurements made at the NASA ABLE 3B forest site by D. R. Fitzjarrald (unpublished data, 1990) using the following approach. First, hourly average values of the roughness length were computed using the Businger-Dyer stability correction [*Stull*, 1988]

$$\ln\left[\frac{z-d}{z_o}\right] = \kappa\left(\frac{U}{u^*}\right) + \Psi_M\left(\frac{z}{L}\right) \quad (3)$$

where z is the measurement height, d is the displacement height, z_o is the roughness length, κ is von Karman's constant (0.4), U is the time-averaged wind speed at height z, u^* is the friction velocity at height z, and Ψ_M is the Businger-Dyer stability correction which is a function of the Monin-

TABLE 3. Daily Mean and Median Values for the Total Forest Canopy Resistance (R_t) Derived From the Forest Canopy Flux Box Model

Days	Nighttime Resistance*, s cm^{-1}		Daytime Resistance†, s cm^{-1}	
	Median	Mean ± 1σ	Median	Mean ± 1σ
204-205	4.83	5.54 ± 1.81	0.69	0.76 ± 0.24
205-206	4.90	5.20 ± 1.80	1.45	1.58 ± 0.52
206-207	4.80	5.97 ± 3.93	0.61	0.48 ± 0.06
207-208	4.03	4.09 ± 1.10	0.69	0.72 ± 0.23
208-209	5.08	5.89 ± 3.92	0.79	0.79 ± 0.34
209-210	8.52	9.07 ± 3.19	1.60	1.81 ± 0.61
210-211	5.13	7.65 ± 7.15	2.36	2.65 ± 1.61
211-212	0.85	0.93 ± 0.57	0.60	0.47 ± 0.24
212-213	3.96	3.96 ± 1.25	0.92	0.98 ± 0.52
213-214	3.64	3.44 ± 0.91	0.57	0.65 ± 0.22
214-215	3.18	3.97 ± 2.85	1.10	1.15 ± 0.34
215-216	4.87	5.35 ± 1.76	0.90	1.00 ± 0.38
216-217	8.48	10.55 ± 7.16	1.26	1.33 ± 0.38
217-218	4.30	4.60 ± 2.01	—	—

*Spans time interval from 2000 to 0600 LT.
†Spans time interval from 0900 to 1700 LT.

Obukhov stability parameter z/L. The measurement height was at 30 m. *Fitzjarrald and Moore* [this issue] have shown that in this boreal forest the displacement height is small compared to the canopy height (0.9 m versus 6 m) and can be ignored when calculating roughness length. Next, the drag coefficient, C_D, was computed from the relationship [*Garratt*, 1992]

$$C_D = \frac{\kappa^2}{[\ln(z/z_o) - \Psi_M(z/L)]^2} \quad (4)$$

Finally, the aerodynamic resistance was obtained from the horizontal wind speed, u, measured slightly above the forest canopy and an appropriately scaled drag coefficient using the relationship

$$R_a = (C_D\, u)^{-1} \quad (5)$$

Comparison of the aerodynamic resistance computed using equations (3)–(5) with the total forest canopy resistance obtained using the radon inventory method is illustrated in Figure 7 for a representative time period. This figure shows that except for nocturnal periods of exceptionally high aerodynamic resistance, total forest canopy resistance values computed using the radon inventory method are generally 2 to 3 times larger than the aerodynamic resistances computed from the wind speed measurements. In a gross sense these resistance values mimic each other on hourly and daily timescales.

Figure 8 illustrates the time variation of the within-canopy resistance. Removal of the contribution of the quasi-laminar boundary layer resistance to the within-canopy resistance (Figure 8) results in little change in the net canopy resistance (R_c). Because R_b is difficult to evaluate experimentally [*Baldocchi et al.*, 1987], we have assumed that R_b has the same value as the aerodynamic resistance [*Hicks and Matt*, 1988, their Figure 6]. *Fitzjarrald and Moore* [this issue] obtained estimates of stomatal resistance for this boreal forest ranging from 2 to 4 s cm^{-1}. These values are comparable to daytime within-canopy resistances illustrated in Figure 8 but are often exceeded during nocturnal periods. However, we have no way of identifying the particular sources of within-canopy resistance and their relative contributions. If stomatal resistance is a significant fraction of the within-canopy resistance, then our assumption that plant-mediated transport of radon is unimportant may not be true for this lichen-woodland forest canopy. The integral term in equation (1) would be positive, which would result in a smaller forest canopy trace gas exchange coefficient, and the corresponding total forest canopy resistance would be larger. We conclude that this is possible within the limits of the data we have presented, and that plant-mediated transport of radon via evapotranspiration may be important. Future studies of radon transport by vascular plants is warranted. It is important to note here that surface resistances computed using the radon inventory method are gas specific. Application of radon-derived resistances to within-canopy transport of trace gases exchanged by plants will depend on the bioactivity of the gas of interest.

An important source of error that may bias the total forest canopy resistance (R_t) is nonuniform mixing of the forest canopy, especially during nocturnal periods. The relatively large nocturnal within-canopy resistance values shown in Figure 8 imply that the mean canopy radon term $\overline{C}$ in equation (1) is biased by high near-surface radon concentrations and that the canopy may be strongly stratified within its lowermost meter (Figure 2). Recomputation of the total forest canopy resistance for a forest with a 1-m stagnant radon layer results in nocturnal within-canopy resistance values that are nearly one half of those shown in Figure 8. This stagnant layer is of comparable scale with the 0.9-m displacement height computed by *Fitzjarrald and Moore* [this issue] for this forest. This similarity of scale suggests that large amounts of radon and other trace gases emitted by the soil may be trapped in a shallow stagnant boundary layer on the forest floor especially during nocturnal periods.

SUMMARY AND CONCLUSIONS

Continuous, high-resolution time series measurements of atmospheric radon concentration gradients can be combined

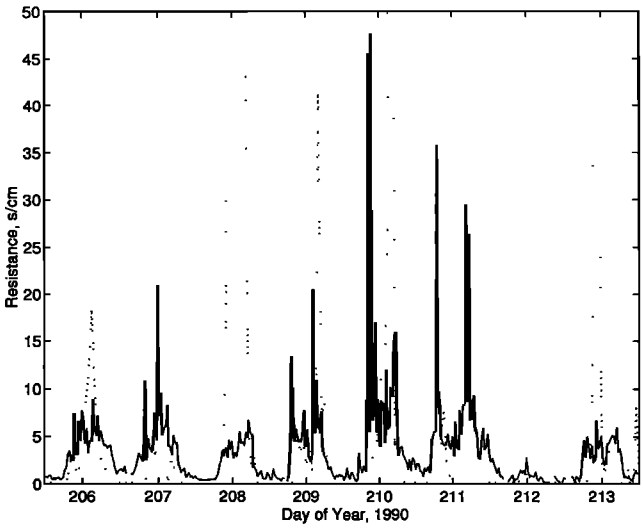

Fig. 7. Comparison of the total forest canopy resistance (R_t) computed from the canopy exchange coefficient, k (solid curve), with the aerodynamic resistance to the transfer of momentum, R_a (dotted curve), computed from micrometeorological measurements. R_a was computed using a mean wind speed measured at $z = 10$ m.

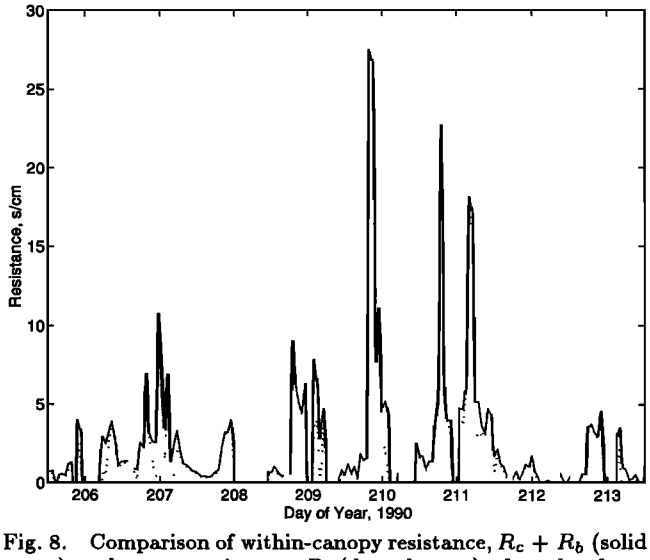

Fig. 8. Comparison of within-canopy resistance, $R_c + R_b$ (solid curve), and canopy resistance, R_c (dotted curve), plotted as function of time. Except for limited time periods, removal of R_b has little effect on the overall canopy resistance. During most nocturnal periods, aerodynamic resistance exceeded the total forest canopy resistance, resulting in negative values for the within-canopy resistance.

with simultaneous radon soil flux measurements to quantify the rate of vertical trace gas exchange between a forest canopy and the overlying atmosphere. The similarity between nocturnal total forest canopy resistance values for the open-canopy boreal forest and a closed-canopy rain forest (Figure 6) suggests that at least during nocturnal conditions, aerodynamic structure of the canopy air, rather than the resistance of the vegetation itself, is the factor controlling the rate of vertical trace gas transport in forest ecosystems.

Average total forest canopy resistances computed using the simple "flux box" model for radon exchange between the forest floor and the overlying atmosphere range from 0.47 ± 0.24 s cm^{-1} to 2.65 ± 1.61 s cm^{-1} for daytime hours (0900-1700 LT) and from 3.44 ± 0.91 s cm^{-1} to 10.55 ± 7.16 s cm^{-1} for nighttime hours (2000-0600). Atmospheric radon measurements using the automated techniques we have employed have low maintenance requirements, making them more suitable for remote field locations where daily maintenance of standard micrometeorological equipment, such as sonic anemometers, is not possible.

The mean radon soil flux from the forest floor for the period July 23 to August 1, 1990, was 41.1 ± 4.8 Bq m^{-2} h^{-1}. The absence of nocturnal peaks and anomalous changes in radon accumulation rates in near-surface atmospheric radon concentrations closely coincide with rainfall events. This correlation is consistent with previous investigations of the effects of rainfall on radon soil gas and suggests that rainfall may cause a rapid reduction in radon soil flux by soil pore space occlusion. Drying of the soil surface appears to be relatively rapid in this boreal forest environment, as evidenced by the rapid reestablishment of normal near-surface atmospheric radon concentrations following rainfall. Temporal scaling is an important issue for models quantifying soil trace gas exchange with the biosphere and atmosphere.

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