

Long-term measurements of ^{222}Rn in soil gas and at different height levels in Amazon tall tower observatory

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ABSTRACT

Emitted from soil as a radioactive noble gas, ^{222}Rn has the potential to be used as a tracer for greenhouse gases. In the atmosphere, it undergoes transport and mixing processes that depend on climatological factors. Therefore, studying factors that influence radon dispersion in hotspot areas can improve our understanding of the driving forces behind atmospheric gas behavior. This study presents data obtained from five years of soil gas measurements at various heights above ground level (18 m, 40 m, and 80 m) below and above the forest canopy in the pristine area of the Amazon Tall Tower Observatory to identify important soil parameters related to radon activity concentration. The results indicate that moisture significantly affects radon activity concentrations during dry and wet periods. The mean ^{226}Ra activity concentration in the soil is 28.3 ± 2.1 Bq/kg. In topsoil, ^{222}Rn varies from 152 to 470 Bq/m³ at ground level, from 1.2 to 10.3 Bq/m³ at 18 m, from 0.37 to 8.6 Bq/m³ at 40 m, and from 1.0 to 5.0 Bq/m³ at 80 m. Daily variations show higher activity concentrations in the early morning, while monthly variations show higher activity concentrations during the dry period.

1. Introduction

Radon (Rn) is an inert noble gas. Three radon isotopes are environmentally significant due to their continuous production in natural decay series: radon (^{222}Rn) from the ^{238}U series (half-life 3.823 days), actinon (^{219}Rn) from the ^{235}U series (half-life 3.983 s), and thoron (^{220}Rn) from the ^{232}Th series (half-life 55.83 s) (Baskaran, 2016). Radon isotopes and their short-lived decay products constitute the largest source of natural radiation exposure when inhaled outside and inside buildings (Belete and Anteneh, 2021). Consequently, most radon-related research has focused on potential radiological health impacts on humans (Al-Zoughool and Krewski, 2009; Skubacz et al., 2019; Sukanya et al., 2022; Ismail et al., 2023; Akuo-ko et al., 2024).

Soil is the primary source of continuous radon emission into the atmosphere, which originates from the decay of radium isotopes. Newly

formed radon atoms diffuse into air-filled pores and can eventually escape through mineral grains into the air. The rate at which radon moves from soil to the atmosphere is called the exhalation rate (Sharma et al., 2024). Several physical factors govern the transfer of radon from soil to the atmosphere, affecting the efficiency with which radon partitions into soil-air pore space or exchanges with the overlying atmosphere. These factors include the soil's content of the parent isotope, porosity, moisture content, temperature, and pressure gradients (Čeliković et al., 2022). Once released into the air, radon undergoes atmospheric transport and mixing processes driven by meteorological conditions (Hirsch, 2007), particularly for the ^{222}Rn isotope due to its relatively long half-life of 3.8 days.

These unique properties make radon a powerful tool in atmospheric transport models (ATMs) to investigate atmospheric mixing and transport processes (Chambers et al., 2016; Crova et al., 2021) and to

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estimate greenhouse gas fluxes using the radon tracer method (Manohar et al., 2013; Levintal et al., 2020; Liu et al., 2024). There is a consensus for all these applications that long-term observations of ^{222}Rn concentrations and fluxes must be undertaken (Levin et al., 2021). However, few studies have applied ^{222}Rn specifically to the Amazon region. Trumbore et al. (1990) measured ^{222}Rn emissions from soils in the Adolpho Ducke Forest Reserve in the Amazon rainforest, located 25 km from Manaus, Brazil. They concluded that nocturnal CO_2 production by above-ground vegetation accounted for about 20% of total soil emissions. Martens et al. (2004) used ^{222}Rn to study the transport of CO_2 between the forest canopy and the atmosphere in a tropical rainforest near the Tapajós National Forest in the state of Pará, Brazil. They demonstrated that ^{222}Rn is an effective tracer for net ecosystem exchange (NEE) of CO_2 during calm nighttime hours when eddy covariance-based NEE measurements are unreliable due to low atmospheric turbulence. Simon et al. (2005) used ^{222}Rn to study the vertical dispersion of trace gases in the Amazon rainforest, specifically in the Jaru Reserve (Rondonia) and the Cuieiras Reserve near Manaus. This study confirmed the value of this isotope as a tracer for studying vertical gas exchange within the canopy. However, unlike the present study, these studies were conducted over short time periods.

This study aims to establish a long-term baseline dataset of ^{222}Rn concentrations and soil parameters at the ATTO site. This dataset will support future applications of radon as a tracer for atmospheric transport and greenhouse gas flux estimations in the Amazon region. In this regard, this paper presents the results of five years of radon observations at the 80-m ATTO tower and one year of observations at the Instant tower at ground level and at 18 and 40 m high. Additionally, important soil characteristics affecting ^{226}Ra activity concentration were determined: soil pH, moisture, organic matter, loss on ignition, and bulk and real density.

2. Methodology

2.1. Studies area

The ATTO tower, a research station located in the central Amazon basin, is situated about 150 km northeast of Manaus (Fig. 1). Various measurement instruments were installed on two towers (Andreae et al.,

2015). The newest tower, completed in 2015, is 325 m tall (S 02°08.752', W 59°00.335'). It is referred to here as the ATTO Tower. An older tower, referred to here as the Instant Tower, is 80 m tall and was built in 2012 (S 02°08.647', W 58°59.992').

At the ATTO site, the year is divided into two distinct seasons: a rainy season from December to May and a dry season from June to November. The average air temperature ranges from 24 to 26 °C, with minimal variation throughout the year, and the average annual precipitation is approximately 2300 mm. According to the Köppen classification, the climate in the region is defined as Am Tropical Monsoon (Paralovo et al., 2016). The spatial distribution of soil groups is highly associated with topography and parent material, with hydromorphic Oxisols predominating in uplands and Spodosols occurring at low elevations (Ríos-Villamizar et al., 2022).

2.2. Soil characteristics: pH, moisture, organic matter, loss on ignition, bulk and real density

To determine the pH, 10 g of soil was placed in a 100 mL plastic beaker. Then, 25 mL of 1 mol L⁻¹ potassium chloride (KCl) solution was added, and the mixture was stirred with a glass rod for 1 min. After settling for 1 h, the sample was stirred again for the same amount of time with a glass rod, and then pH readings were taken immediately. The same procedure was performed to determine the pH in a water medium and calculate the delta pH (Kome et al., 2018).

To determine moisture content, a soil sample was collected and its wet mass was measured using a hot air oven at 105 °C until it reached a constant weight, which yielded the dry weight. Another portion of the same soil was first air-dried at room temperature, then placed in the oven at 105 °C. The moisture content was calculated based on the difference in mass between the two weighings.

The organic matter content was determined by measuring the mass loss after calcining the sample at 550 °C for 4 h, while the loss on ignition was determined by measuring the mass loss after calcining the sample at 1000 °C. The lost mass determined by loss on ignition refers to carbonate decomposition and the dehydration of (oxy)hydroxides, such as iron and manganese (Hoogsteen et al., 2015).

Bulk density was calculated by dividing the dry soil mass by its volume, excluding the pore space volume.



Fig. 1. Location of the site of study, a) Brazil map showing Manaus location, in the state of Amazon and b) location of ATTO site, where the tower is installed, and its relative position in relation to Manaus.

2.3. Soil chemical characterization

The soil was chemically characterized using instrumental neutron activation analysis (INAA) (Szentmiklósi et al., 2023). The soil samples and reference materials were irradiated for 8 h. Two sets of counting were done for elemental determination. The first was performed one week after irradiation, and the second was performed two weeks after irradiation. Irradiation was performed at a neutron flux of approximately 1 to $5 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$ at the IEA-R1 nuclear research reactor at the Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN-SP). As reference materials, standard solutions containing certified amounts of the elements to be determined (Spec Certiprep, USA) were pipetted onto a strip of filter paper. Certified reference materials, Estuarine Sediment (STM 1646a) and Table Mountain (USG-STM-2), were also measured to ensure the quality of the results.

2.4. Soil radionuclides content

The activity concentrations of ^{226}Ra , ^{228}Ra , and ^{40}K were determined by gamma spectrometry using a high-purity germanium detector (GX2020) from Canberra with 20% relative efficiency and a resolution of 2.00 keV FWHM at 1.33 MeV. Daily performance verification was conducted using the ^{57}Co isotope with an energy of 122 keV and the ^{60}Co isotope with an energy of 1332 keV. For this measurement, the sample was stored in a sealed container for at least 30 days to allow equilibrium to be reached between ^{222}Rn and its short-lived daughters. The ^{226}Ra activities were determined by taking the mean activity of two separate photo peaks of its daughter nuclides: For ^{214}Pb , the energy was 352 keV; for ^{214}Bi , the energy was 609 keV. For ^{228}Ra , the energy was 338 keV and 911 keV. The ^{40}K activity was determined using the 1460 keV gamma-ray peak. Sample measurements were taken for 86,400 s. Background was counted for 172,800 s, and certified reference materials (CRMs) RG-U, RG-Th, and RG-K from the IAEA were counted for 7200 s.

To ensure quality assurance and certainty, the IAEA-327 reference material (Radionuclides in Soil) from the IAEA was used. The obtained values were $626 \pm 43 \text{ Bq/kg}$ for ^{40}K , $36 \pm 3 \text{ Bq/kg}$ for ^{226}Ra , and $38 \pm 7 \text{ Bq/kg}$ for ^{228}Ra . All of the results are within the 95% confidence interval of the reference material: 612–630, 32.7–35.5, and 37.2–39.2 Bq/kg for ^{40}K , ^{226}Ra , and ^{228}Ra , respectively.

2.5. Radon measurements

The first long-term ^{222}Rn measurement at the ATTO site began in December 2018 at the ATTO tower, which is 80 m above ground level (a. g.l.) and approximately 50 m above the canopy. A Heidelberg Radon Monitor (HRM) was used for the measurements. Designed in the 1990s, it is described in detail in Levin et al. (2002). The HRM is a one-loop filter that measures the polonium-214 (^{214}Po) isotope, which is an alpha (α) emitter and a daughter product formed during the decay of ^{222}Rn (Grossi et al., 2020). In short, the HRM consists of a Whatman filter (QMA, 47 mm diameter), on which aerosols are actively deposited from an ambient airflow of approximately $1 \text{ m}^3/\text{h}$ (Levin et al., 2002). Alpha activity is measured by a surface barrier detector (Canberra CAM 900 mm^2 active surface) located approximately 5 mm from the surface of the loaded filter. Readings are recorded as the mean integrated signal every half hour. The final measurement uses a disequilibrium factor of $^{214}\text{Po}/^{222}\text{Rn} = 0.704 \pm 0.081$ to correct for the discrepancy between the activity concentrations of ^{214}Po and ^{222}Rn (Levin et al., 2002, 2017). Data from the HRM detector from 2018 to 2024 is available at <https://www.attodata.org/ddm/data/Showdata/487>.

In October 2023, three RAD7 devices were installed on the Instant Tower. The RAD7 (model RAD7, DurrIDGE Co., Billerica, MA) is a solid-state, ion-implanted silicon alpha particle detector. The detector directly converts alpha radiation to an electric signal, which allows for the discrimination of alpha particles by energy. Radon activity concentrations are determined by the decay of ^{218}Po and ^{214}Po trapped in a 0.7 L

chamber cell operating at a flow rate of 2.5 L/min. We used the manufacturer's calibration for our analyses. The manufacturer guarantees the instrument's calibration accuracy with a precision better than 5% and an intrinsic background of 0.2 Bq/m^3 for the life of the instrument (DurrIDGE, 2024).

Three Rad7 instruments were installed inside a container. Each one was connected to a tube. One tube collected ^{222}Rn exhaled from the soil. For this measurement, a 25-cm-high PVC pipe with an internal diameter of 7.2 cm was buried approximately 5 cm deep in the ground. This pipe is connected to the RAD7 inlet through a 5-m tube. The second tube is approximately 23 m long and collects air from 18 m above ground level (around 12 m below the canopy). The third tube is approximately 45 m long and collects air from 40 m above ground level (around 10 m above the canopy). For the two tubes installed on the Instant Tower, a filter was installed to prevent dust from entering. Both tubes were mounted with the opening facing the ground to prevent rainwater from entering. Before entering the RAD7, all air samples pass through a desiccant vessel to prevent humidity in the equipment. Readings are recorded as an integrated hourly mean.

3. Results

3.1. Soil parameters

Table 1 shows the results of moisture, organic matter, and carbonate loss, together with the pH values measured in H_2O and KCl, as well as the ΔpH value. Soil organic matter formation is determined by the decomposition and transformation of plant litter inputs, which are highly variable in the Amazon forest. According to Souza et al. (2018), the Amazon region has high rates of organic matter mineralization due to high temperatures and consistently high moisture levels. Although the LOI value at 1000°C is not specific to carbonate, since calcination at this temperature also eliminates chemically bound water and hydroxyl groups that are part of some minerals, the LOI values found at 1000°C are in the range reported by Cailleau et al. (2014) for the Amazon region, varying from 1.4 to 6%. This mass loss is assumed to be mostly due to organic matter oxidation.

The ΔpH , which is the numerical difference between the pH values measured in 1 M KCl and H_2O ($\text{pH}_{\text{KCl}} - \text{pH}_{\text{H}_2\text{O}}$), showed a negative value, which implies that negative charge predominates in this soil and acts as a cation exchange layer. Bulk density is an important factor affecting soil gas exhalation and, consequently, radon activity concentration at the soil level. This is because an increase in bulk density results in decreased soil porosity (Benavente et al., 2019).

3.2. Soil chemical composition and radionuclides

To determine the elemental composition of the soil by INAA, 34 soil samples were collected from the nearby ATTO and Instant towers, and

Table 1
pH and ΔpH ($\text{pH}_{\text{KCl}} - \text{pH}_{\text{H}_2\text{O}}$), relative and real density, moisture, organic matter and loss on ignition in soil samples collected at ATTO region.

$\text{pH}_{\text{H}_2\text{O}}$	pH_{KCl}	ΔpH	BD (g cm^{-3})	RD (g cm^{-3})	Temperature	% loss $\pm$ S. D.
4.05	3.78	-0.27	0.93	2.28	Oven at 105°C (after 24h)	24.5 ± 0.1
					Air dried and oven at 105°C (after 24h)	1.27 ± 0.15
					550°C (after 4h)	12.7 ± 0.64
					1000°C (after 2h)	3.35 ± 0.08

BD, relative density; RD, real density; SD, Standard deviation.

105°C (Moisture); 550°C (Organic matter); 1000°C (Loss on ignition).

the results are shown in Table 2. The alkaline elements K, Rb, and Cs were only measured in six samples, with values lower than the UCC average of 23,068, 82, and 4.1 µg/g, respectively (Gaschnig et al., 2016). Calcium was below the detection limit (2200 µg/g) in all samples, and the amount of iron is greater than that of the UCC (3.9). These results are consistent with those presented by Matschullat et al. (2020) for thirteen locations in the state of Amazonas, covering an area of approximately 740,000 km² in the central and southern regions. The Th/U ratio of 7.24, the Zr/Th ratio of 38.2, and the La/Lu ratio of 34 are in agreement with the results shown by Costa et al. (2014). Soil characterization shows intense scavenging of more soluble elements and enrichment of heavy minerals. The results also suggest that Th and U are likely associated with different soil-forming components.

The same geochemical characteristics are evident in the activity concentrations of the radionuclides ²²⁶Ra, ²²⁸Ra, and ⁴⁰K, as shown in Table 3. The thorium series presents values higher than the global average, and there is a depletion of ⁴⁰K (UNSCEAR - UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION, 2010).

3.3. Radon activity concentrations

3.3.1. Radon in soil

Fig. 2 shows the mean diurnal cycle of radon activity concentration at four vertical levels (soil, 18 m, 40 m, and 80 m) and the observed ranges during the rainy (December–May) and dry (June–November) seasons at the ATTO site. The solid black line represents the mean diurnal cycle computed from all available monthly data (see Supplementary Material 1–12), and the shaded envelopes correspond to the minimum and maximum values observed each hour within each season. These values are aggregated across one year (August 2023 to June 2024) of measurements for the soil, 18-m, and 40-m levels. For the 80-m level, the measurements cover a period of five years (January 2019 to April 2023). Grouping observations by the regional seasonal regime allows the envelopes to capture the full range of variability associated with dry and rainy conditions while preserving the underlying diurnal structure. Comparing heights reveals a progressive reduction in radon activity and

Table 2

Basic statistics for the element concentration of soil samples from Plateau region, at ATTO site, in µg g⁻¹, except Fe in %.

	Valid N ^a	Mean	Minimum	Maximum	Std. Dev.
Na	34	75.4	41.1	200.9	32.0
K	6	458.8	213.4	715.7	179.5
La	34	13.6	0.6	23.2	3.8
Nd	26	10.3	6.2	15.8	2.5
Sm	34	1.8	0.5	3.3	0.7
Yb	34	2.2	0.5	3.7	0.7
Lu	34	0.4	0.1	0.6	0.1
U	34	4.1	0.6	7.1	1.4
Sc	34	5.5	0.3	7.4	2.0
Cr	34	84.1	1.5	121.1	30.2
Fe (%)	34	5.9	0.0	9.3	2.2
Co	34	0.8	0.0	1.6	0.4
Zn	25	24.0	1.4	92.7	21.6
Rb	6	8.9	2.3	26.8	9.2
Sb	34	0.5	0.0	0.7	0.1
Ba	17	33.0	6.5	72.6	16.8
Cs	6	0.2	0.0	0.3	0.1
Ce	34	17.3	0.3	23.3	6.4
Eu	31	0.3	0.0	0.7	0.2
Tb	33	0.2	0.0	0.5	0.1
Hf	34	28.1	12.4	50.2	6.7
Ta	34	2.5	0.1	3.8	0.8
Th	34	29.7	0.6	41.9	10.3
As	32	7.1	4.8	8.9	1.0
Br	34	19.3	0.5	39.7	8.2
Zr	34	1134.7	498.4	1608.3	230.3

^a Valid N means the number of measurements above detection limit.

Table 3

Radionuclides activity concentrations in soil from Plateau region, at ATTO site, in Bq kg⁻¹.

	²²⁶ Ra	²²⁸ Ra	⁴⁰ K
This work	28.3 ± 2.1	108.9 ± 13.7	11.2 ± 0.9
Global average ³⁸	32	45	412

the diurnal cycle's amplitude with increasing height.

The variation in activity concentrations (Fig. 2a) was relatively low during the day, with a coefficient of variation (CV) ranging from 28.8% to 34.9% (Brown, 1998). There was a slight tendency for lower values to be present between 10 a.m. and 1 p.m., as well as a slight increase at dusk and during the night. Considering all the measurements, the ²²²Rn activity concentration at the soil surface varied from 152 to 470 Bq/m³. The mean values for each hour during the measurement period are shown in Table 4. Higher daily activity concentrations occurred during the dry period (July to December).

The activity concentrations of ²²²Rn in the soil gas measured at the ATTO site are consistent with the values reported by Akuo-ko et al. (2024) and their references, which range from 34.7 to 1447.6 Bq/m³ near the soil surface where radon measurements were taken.

The monthly variation of soil activity concentration shown in Fig. 3a indicates that higher activity concentrations occurred from September to December. Considering the rainy and dry periods, as well as the annual average, the mean soil activity concentrations were 245 ± 16, 303 ± 18, and 274 ± 16 Bq/m³, respectively. A statistical comparison of the data obtained during the dry and rainy seasons, performed using a Student's t-test for independent samples with equivalent variances, revealed a statistically significant difference between the two periods, with a p-value <0.05.

3.3.2. Radon activity concentrations below the canopy (18 m a.g.l)

In general, the activity concentration of ²²²Rn measured at 18 m above ground (Fig. 2b) indicates that higher activity concentrations occur during the early morning hours up to around 8 p.m.. Concentrations then start to fall and increase again at sunset. Considering all the measurements, the activity concentrations varied from 1.2 to 10.3 Bq/m³. The mean values for each hour during the measurement period are shown in Table 4. It should be noted that the measurements taken at 80 m correspond to a period of five years, while the others correspond to a period of one year.

Higher activity concentrations arise in the drier months (Fig. 3b), from September to December, and follow the same soil pattern. The mean value for the dry season is 4.8 ± 1.1 Bq/m³, for the rainy season it is 3.8 ± 0.9 Bq/m³, and the annual mean value is 4.3 ± 1.0 Bq/m³. A Student's t-test for independent samples with equivalent variances revealed a statistically significant difference in the means of the dry and wet periods, with a p-value of less than 0.05.

3.3.3. Radon activity concentrations above the canopy (40 m a.g.l)

The same pattern of diurnal radon variation was observed in the measurements taken at 40 m a.g.l., as shown in Fig. 2c. The ²²²Rn activity concentration varied from 0.37 to 8.6 Bq/m³ at this height when all the measurements from this period were considered. The mean values at each hour are shown in Table 4. The mean values were 4.0 ± 0.9 Bq/m³ in the dry season, 3.9 ± 0.8 Bq/m³ in the rainy season, and 3.9 ± 0.8 Bq/m³ annually. A Student's t-test for independent samples with equivalent variances revealed no statistically significant difference in means between the dry and wet periods, with a p-value >0.05. Fig. 3b shows the mean monthly activity concentrations of ²²²Rn at a height of 40 m, presenting the same distribution pattern observed in the soil and air below the canopy.

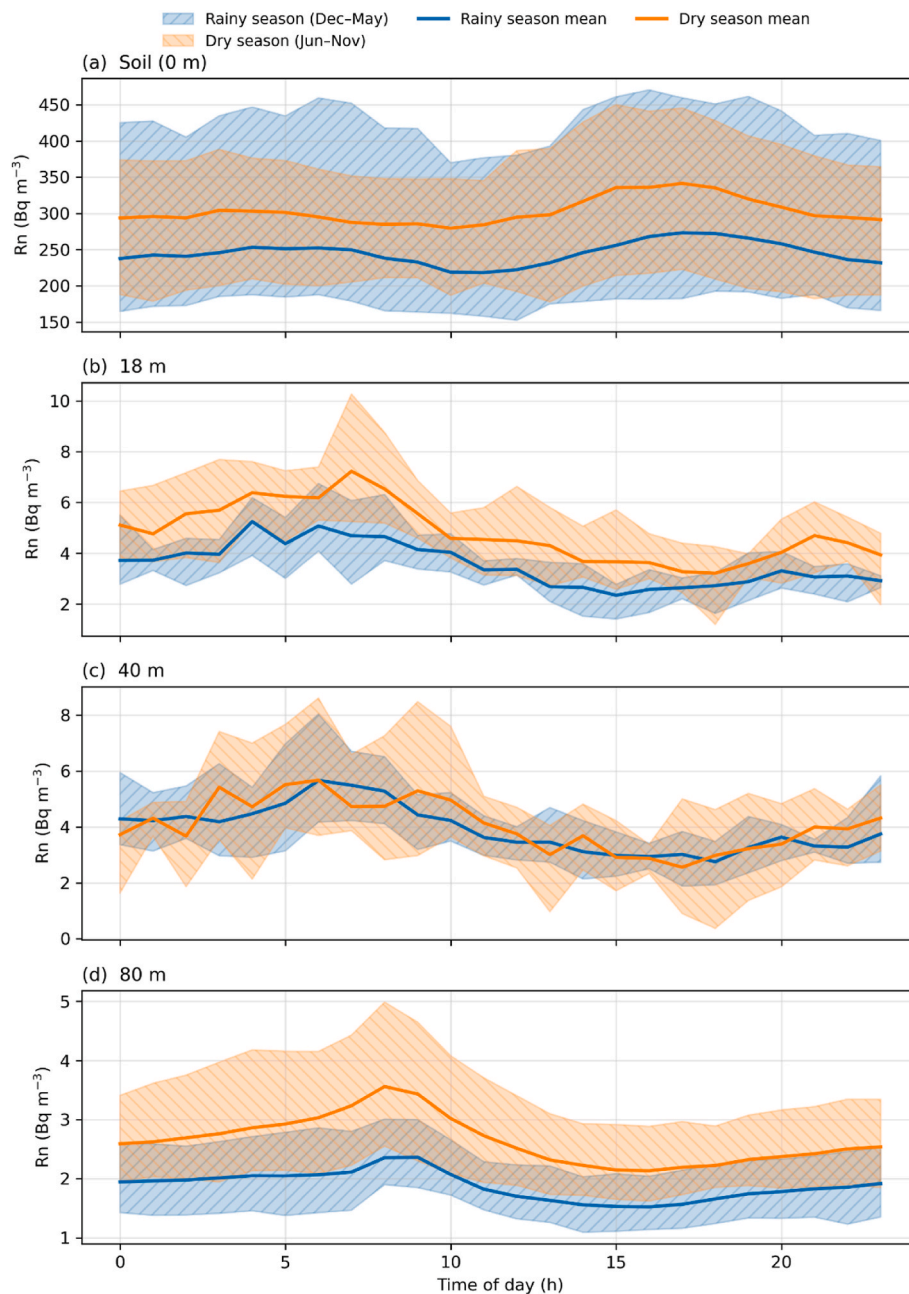


Fig. 2. Solid lines represent the mean diurnal cycle for the rainy and dry seasons, while the shaded areas indicate the corresponding observed ranges. a) soil, b) 18 m, c) 40 m, corresponding to one year of observation, and d) 80 m (corresponding to five years of observation).

3.3.4. Radon activity concentrations above the canopy (80 m above ground)

Fig. 2d shows the pattern of radon activity concentration. The higher activity concentrations at this height also occur during the early morning hours and continue until around 8 p.m., decreasing during the afternoon and increasing again at sunset. Considering all the measurements, the activity concentrations varied from 1.0 to 5.0 Bq/m³. The mean values for the measurement period are shown in Table 4.

Fig. 4 shows the annual variation of ²²²Rn activity concentration at the ATTO tower. From 2019 to 2021, the concentrations remained relatively constant, with minimal variation and a peak in July. In 2022, higher values were observed from July to September. In 2023, measurements were taken for only four months (January to April), presenting higher mean concentrations than in previous years. Fig. 3b shows the activity concentrations for the rainy and dry periods. The ²²²Rn distribution is similar to that observed below the canopy. The dry

season presents higher mean values ranging from 1.7 ± 0.2 to 2.1 ± 0.2 Bq/m³ in the rainy season and from 2.4 ± 0.3 to 2.7 ± 0.4 Bq/m³ in the dry season. A Student's t-test for independent samples with equivalent variances revealed a statistically significant difference in the means of the dry and wet periods, with a p-value <0.001.

4. Discussion

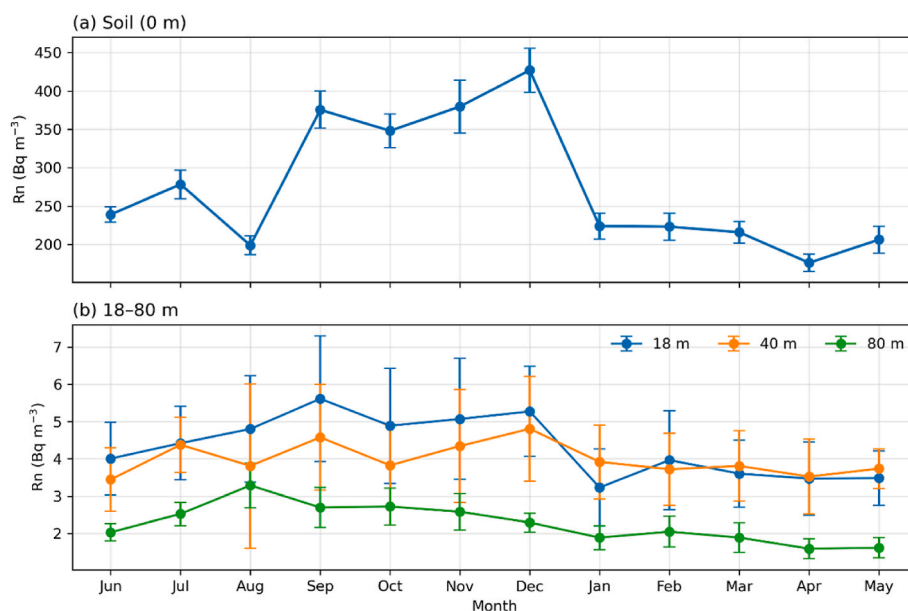
4.1. Physical and geochemical characteristics of the ATTO soil

The rate at which ²²²Rn is released from the soil depends on the ²²⁶Ra activity, as well as the efficiency with which the produced ²²²Rn enters the soil air pore space. This efficiency is moisture dependent. The flux to the atmosphere, in turn, depends on factors that regulate the rate of gas exchange between the soil air pore space and the overlying atmosphere. These factors include soil structure, soil humidity, and soil surface

Table 4Mean activity concentration, in Bq m^{-3} , measured at soil surface, 18 m, 40 and 80 above ground (data presented in the Supplementary Materials 1 – 12).

ToD* (h)	Mean		SD	Mean		SD	Mean		SD	Mean		SD
	Soil			18m			40m			80 m		
20	283	±	88	3.8	±	0.9	3.5	±	0.8	2.1	±	0.4
21	272	±	80	4.0	±	1.2	3.7	±	0.7	2.1	±	0.5
22	265	±	83	4.0	±	1.0	3.6	±	0.7	2.2	±	0.5
23	262	±	81	3.6	±	1.0	4.0	±	1.0	2.2	±	0.5
0	266	±	86	4.7	±	1.4	4.0	±	1.0	2.2	±	0.5
1	269	±	87	4.5	±	1.2	4.3	±	0.6	2.3	±	0.5
2	267	±	81	5.0	±	1.4	4.0	±	1.0	2.3	±	0.6
3	275	±	87	5.0	±	1.3	4.8	±	1.5	2.3	±	0.6
4	278	±	84	5.9	±	1.0	4.6	±	1.3	2.4	±	0.6
5	276	±	82	5.5	±	1.3	5.2	±	1.3	2.4	±	0.6
6	274	±	85	5.7	±	1.0	5.7	±	1.5	2.5	±	0.7
7	269	±	81	6.2	±	2.0	5.1	±	0.9	2.7	±	0.8
8	262	±	75	5.7	±	1.5	5.0	±	1.3	2.9	±	0.7
9	259	±	79	5.0	±	1.0	4.9	±	1.5	2.6	±	0.6
10	249	±	76	4.3	±	0.7	4.6	±	1.1	2.3	±	0.6
11	251	±	77	4.1	±	1.1	3.9	±	0.7	2.1	±	0.5
12	259	±	84	4.0	±	1.1	3.6	±	0.6	2.0	±	0.5
13	265	±	87	3.5	±	1.2	3.2	±	1.0	1.9	±	0.5
14	281	±	97	3.2	±	0.9	3.4	±	0.8	1.8	±	0.5
15	296	±	103	3.1	±	1.1	2.9	±	0.9	1.8	±	0.4
16	302	±	100	3.2	±	0.8	2.9	±	0.4	1.8	±	0.4
17	307	±	96	3.0	±	0.7	2.8	±	1.2	1.8	±	0.4
18	304	±	92	3.1	±	1.0	2.9	±	1.1	1.9	±	0.4
19	293	±	91	3.3	±	0.6	3.2	±	1.0	2.0	±	0.4

ToD* - Local time of the day.

**Fig. 3.** Monthly concentration of ^{222}Rn activity in the soil (3a), at 18 m and 40 m (one year of observation) and 80 m (five years of observation) above the ground in the ATTO region.

temperature, among others. These factors lead to different soil activity concentrations under wet and dry conditions.

Soil bulk density is an important parameter that controls radon diffusion. In soils with higher bulk density, reduced porosity affects the distribution and connectivity of air phases in pore space (Kaur et al., 2019). For the soil analyzed in this study, the porosity present at the surface (approximately 20 cm) occupies 59.5% of the soil volume, classifying it as soil with very high porosity.

Soil pH is a vital property that significantly influences solute concentration and element adsorption. In the case of radium, adsorption depends on the net surface charge, which is a function of pH (Sposito, 2008). The author stated that, among the alkaline earth elements,

radium is one of the most strongly adsorbed onto clay minerals via ion exchange at $\text{pH} \geq 7$. Therefore, the low pH values observed in the analyzed samples may favor radium (Ra) scavenging from the soil. Maity et al. (2015) made the same observation, indicating lower Ra concentrations in soils with lower pH values. Nathwani and Phillips (1979) also found that, when the concentration of calcium is low, radium is preferentially adsorbed onto organic matter besides clay minerals. In highly weathered soils at ATTO, organic matter content reached 12.7%. The influence of radium associated with organic matter on radon soil concentration will be further investigated, as shown in Noskova and Shuktomova (2015) and Kaur et al. (2019).

Geochemically, the soil in the ATTO region is characterized by low

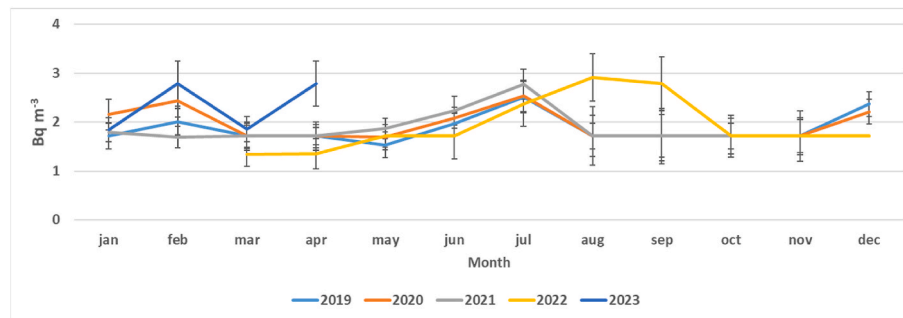


Fig. 4. Monthly ^{222}Rn activity concentration at 80 m above ground at ATTO region.

levels of alkaline and earth alkaline elements (K, Rb, Cs, and Ca). This indicates the presence of a strong scavenging process that removes these elements while retaining the heavier ones. The Th/U, Zr/Th, and La/Lu ratios suggest the accumulation of heavy elements, likely due to the adsorption of U onto the high concentrations of Fe-oxyhydroxides, where the daughter nuclide ^{226}Ra can also be retained. Fe-oxyhydroxides have a high adsorption capacity and primarily act as surface coatings on soil, although this process is more efficient at a pH of approximately 8 (Beltrán-Torres et al., 2023). Nathwani and Phillips (1979) found that organic matter absorbs approximately ten times more ^{226}Ra than clays do. Therefore, the cation adsorption capacity, as indicated by the negative ΔpH value, and the amount of organic matter in the ATTO region's soil likely play an important role in radon emanation from soil gas.

After radon emanates from grain minerals, the primary mechanisms by which this gas is transported from surface soil pore spaces to the atmosphere are molecular diffusion and "pumping" associated with the pressure gradients induced by turbulence between the soil and the overlying air. Soil moisture content strongly affects the resulting exhalation rate, as the recoil range for radon in water is much less than in air (Baskaran, 2016). Several studies have shown that radon exhalation behavior differs among soil types at different water contents (Wang et al., 2022). The maximum exhalation values occur at intermediate water contents and depend on soil properties that determine the soil's water-holding capacity. A significant decrease in exhalation occurs at water contents above 25% (Čeliković et al., 2022). The presence of this amount of water surrounding the particle surface obstructs radon escape. Additionally, water modifies the air volume in the soil and the tortuosity of the pore system. This influences radon diffusion, making it more difficult for radon atoms to reach the surface (Al-Shboul, 2023). The moisture measured in air-dried soil samples from ATTO was low ($1.27 \pm 0.15\%$), while samples measured without pre-drying in ambient air had a moisture content of 24.5%. This indicates the soil's capacity to retain capillary and free water, i.e., water strongly retained on the surface of soil particles and water filling the voids between grains. This high moisture variation affects the radon soil concentration, as seen in Fig. 3a for wet and dry periods. These results are supported by those shown in Thu et al. (2020).

4.2. Radon

4.2.1. Radon activity concentration in soil surface

The activity concentration of ^{222}Rn in soil can vary widely. It ranges from a few hundred Bq/m^3 at the surface to tens of thousands of Bq/m^3 at depths of about 1 m (Huxtable et al., 2017; D'Alessandro et al., 2018). Therefore, many external factors can influence the diffusivity and, consequently, the radon exhalation rate from the sub-surface to the surface of soil: rainfall, lower temperature, and increasing atmospheric pressure decrease the exhalation rate, while higher temperature and increasing wind speed can increase it. Additionally, these meteorological parameters can cause convective airflow in the pore space, thereby

changing the radon flux from the soil (Porstendorfer, 1994). Since most soil geochemical characteristics remain constant over long periods of time, the observed variation in soil ^{222}Rn activity concentration at the ATTO site (item 3.3.1) is mainly related to the aforementioned meteorological characteristics.

Several studies (Menetrez et al., 1997; Liang et al., 2022) have shown that radon concentration in soil air spaces increases with moisture content until it reaches a maximum. Then, it decreases once the pore spaces are saturated with water, which limits soil-atmosphere gas exchange. Due to the high porosity of the soil, rainwater penetrates the soil during the rainy period in the ATTO region, reaching a moisture content of up to 24.5%. This lowers the ^{222}Rn activity concentration at the soil surface. During the dry period, higher temperatures and long periods without rain allow the soil to dry. The decrease in soil moisture content results in higher ^{222}Rn activity concentrations in the air-filled pore spaces of the topsoil, allowing for higher surface activity concentrations.

4.2.2. Radon activity concentration in the air

The observed behavior of the monthly variation in ^{222}Rn activity concentration must be due to factors affecting soil concentrations, such as thermal gradients and wind speeds. This applies to all ^{222}Rn activity concentrations in the air at all heights.

Considering the influence of rainy and dry periods, which differ from measurements made below the canopy at 18 m a.g.l. (Fig. 3b), and given the variability, there was no significant difference in mean values between the rainy and dry seasons at 40 m a.g.l. (Fig. 3b). The amplitude of the mean values below the canopy is 1.0 Bq/m^3 , while the value for the measurements at 40 m a.g.l. is only 0.1 Bq/m^3 . This indicates that stable conditions must prevail at this height during dry and rainy periods, and more studies are necessary to understand this behavior.

The activity concentrations in the air measured in this study are comparable to those reported by Crova et al. (2021), who measured radon in Milan, Italy, at 6 m above ground level (m a.g.l.). According to the month of the year, median values varied from 3.5 to 11.5 becquerels per cubic meter (Bq/m^3), with higher concentrations occurring between 5 and 9 a.m. Additionally, Arnold et al. (2010) found values ranging from <1 to 10 Bq/m^3 at an elevation of 20 m a.g.l. in central Netherlands. Martens et al. (2004) found values below 4 Bq/m^3 at greater heights in measurements from 0 to 60 m a.g.l. and observed the same pattern for daily ^{222}Rn variation, with higher activity concentrations during the morning hours and lower activity concentrations in the afternoon and at night.

4.3. General behavior of radon concentrations in the air

After being released from the soil, radon is distributed in the atmosphere primarily through turbulent air mixing, which is characterized by eddy diffusivity. The activity concentration profile at different heights is influenced by the dilution of ^{222}Rn exhaled from the soil into the air, the decay constant, and the wet and dry removal of aerosols from the atmosphere. In the measurements taken for this study, the daily

concentration variations generally reach minimum values in the afternoon and maximum values at night and in the early morning hours. This behavior is explained by enhanced convective mixing in the lower layers of the atmosphere during the afternoon and by thermal inversions that occur at night, producing more stable atmospheric conditions¹⁴. As the temperature increases in the morning and the soil warms, vertical air movement occurs, allowing radon to mix with a greater volume of air (Barba-Lobo et al., 2024).

The coefficient of variation for the mean daily activity concentrations of ²²²Rn at different heights varied from 15.1% to 36.6% at 18 m a.g.l., from 14.0% to 41.6% at 40 m a.g.l., and from 17.5% to 27.8% at 80 m a.g.l. These results suggest that air mixing is less effective under the canopy than in the atmosphere immediately above it and that air mixing is more efficient at higher altitudes.

5. Conclusions

The results of five years of radon observations at the 80-m-high ATTO tower and one year of observations at the 18- and 40-m-high Instant towers, as well as soil characterization for pH, moisture, organic matter, loss on ignition, and bulk and real density, were presented. Measurements were taken in the ATTO tower region, a research station located in the central Amazon basin, approximately 150 km northeast of Manaus.

The soil in this region is characterized by its high organic matter content, high moisture content, small negative pH value, which gives it cation exchange characteristics, and high porosity. The moisture variation measured in air-dried soil (1.3%) and untreated soil (24.5%) indicates the ATTO soil's capacity to accumulate capillary and free water, resulting in the low variation in radon activity concentrations found during the dry and wet periods.

Chemically, the soil has a low content of alkaline and earth alkaline elements, as well as a high content of heavy elements. The high Th/U, Zr/Th, and La/Lu ratios indicate an intense depletion of the more soluble elements and an enrichment of the heavier ones. This also indicates that Th and U (the parent nuclide of the ²²²Rn series) are likely linked to distinct soil-forming components. ²²⁶Ra, the nuclide immediately before ²²²Rn in the ²³⁸U series, is present in the soil at activity concentrations close to the global average. However, ²²⁸Ra is present at concentrations more than twice the global average.

The ²²²Rn activity concentrations in the soil surface during the day present a coefficient of variation between measurements lower than 35%, an amplitude of coefficients of variation around 6%, a slight tendency to present lower values around noon, and a slight increase at dusk and during the night. Annual variation shows that higher activity concentrations occurred during the dry period from July to December, indicating that rainfall and soil porosity are the main driving forces controlling radon soil exhalation and, consequently, radon activity concentrations at soil level.

At all measured heights - 18 m (about 12 m below the canopy), 40 m (about 10 m above the canopy), and 80 m a.g.l. (about 50 m above the canopy) - the activity concentration of ²²²Rn was found to be highest during the early morning hours, decreasing during the day and increasing again at sunset and night. Monthly variations indicate differences in activity concentrations between the dry and rainy seasons at 18 m and 80 m a.g.l. However, no difference was observed at 40 m a.g.l., which must be investigated. Currently, it is thought that the proximity of the canopy may influence radon concentrations at this height.

The annual variation in ²²²Rn activity concentration from the 80 m a.g.l. measurements indicates fairly constant levels from 2019 to 2021, with a peak in July and higher values during the dry period in 2022. In 2023, measurements were taken for only four months of the year, showing higher mean concentrations than in previous years.

Future studies will present data on the correlation between radon activity concentration and soil moisture, organic matter, radon exhalation rates, and a model for atmospheric radon dispersion below and

above the canopy, including atmospheric properties.

CRediT authorship contribution statement

P.S.C. Silva: Writing – original draft, Resources, Formal analysis, Conceptualization. **B. Tappiz:** Formal analysis. **M.F. Maduar:** Data curation. **O.S. Lakis:** Formal analysis. **J.O.W.V. Bustillos:** Investigation. **S. Botía:** Writing – review & editing, Data curation. **S.E. Trumbore:** Writing – review & editing, Data curation. **R. Hazenfratz:** Formal analysis. **H.M.S.M.D. Linhares:** Formal analysis. **F.A.G. Silva:** Formal analysis. **M. Gachkivskyi:** Formal analysis. **S. Bulthuis:** Formal analysis. **A. Tsokankunku:** Formal analysis. **H. Harder:** Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvrad.2026.107940>.

Data availability

The data are presented as supplementary material.

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