

RESEARCH ARTICLE

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## Evaluating Radon and Thoron in surficial soil layers and groundwater from different Brazilian aquifer systems

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### ABSTRACT

This paper describes the characterization of radon ( $^{222}\text{Rn}$ ) and thoron ( $^{220}\text{Rn}$ ) in surficial soil layers (latosols) occurring at the campus of UNESP-São Paulo State University, Rio Claro city, São Paulo State, Brazil. The RAD7 equipment from Durridge Co. was successfully used for discriminating between the signals of both Rn isotopes. The analytical data indicated  $^{220}\text{Rn}$  activity concentration above the  $^{222}\text{Rn}$  activity concentration, as also found in other studies, like in caves of some Chinese Provinces. Contrarily, the dissolved  $^{222}\text{Rn}$  activity concentration in groundwater samples from several Brazilian aquifer systems surpassed the  $^{220}\text{Rn}$  activity concentration, indicating different mechanisms related to the emanation and dissolution of these Rn isotopes in the soil and water phases.

**Keywords** - natural radioactivity, radon, thoron, surficial soil layers, groundwater

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### I. Introduction

Radon ( $^{222}\text{Rn}$ , half-life = 3.83 days) is a colorless, odorless, tasteless, chemically inert and radioactive gas produced continuously in rocks and soils through  $\alpha$ -decay of  $^{226}\text{Ra}$  in the  $^{238}\text{U}$  radioactive decay series ( $4n+2$ ) (Fig. 1). Thoron ( $^{220}\text{Rn}$ , half-life = 54.5 s) is another colorless, odorless, tasteless, chemically inert and radioactive noble gas produced, on other hand, through  $\alpha$ -decay of  $^{224}\text{Ra}$  in the  $^{232}\text{Th}$  radioactive decay series ( $4n$ ) (Fig. 2). Actinon ( $^{219}\text{Rn}$ , half-life = 3.92 s) is the third natural Rn isotope, being generated through  $\alpha$ -decay of  $^{223}\text{Ra}$  in the  $^{235}\text{U}$  radioactive decay series ( $4n+3$ ); such radionuclide will not be focused in this paper due to its very short half-life.

$^{222}\text{Rn}$  and  $^{220}\text{Rn}$  are subjected to recoil at “birth” with some atoms escaping from rocks and soils to the surrounding fluid phase, such as air and groundwater. Their parents, respectively, uranium and thorium, are natural lithophile radioelements widely distributed in crustal rocks, being concentrated preferentially in acid igneous rocks compared with intermediate, basic, and ultrabasic varieties [2]. Thorium is an element ~3-4 times more abundant than uranium in crustal rocks because it is less susceptible to mobilization in the supergene

environment. It occurs predominantly as a tetravalent cation and a trace constituent in phosphates, simple and multiple oxides, and silicates, as well as in the major, rock-forming minerals such as monazite, thorianite ( $\text{ThO}_2$ ), and thorite ( $\text{ThSiO}_4$ ), among others [3, 4].

U-238				
U	U-238 4.49Ga	Pa-234 1.18m	U-234 248Ka	
Pa				
Th	Th-234 24.1 d		Th-230 75ka	
Ac				
Ra		Ra-226 1622a		
Fr				
Rn		Rn-222 3.83d		
At				
Po		Po-218 3.05m	Po-214 0.16ms	Po-210 138d
Bi			Bi-214 19.7m	Bi-210 50d
Pb		Pb-214 26.8m	Pb-210 22.2y	Pb-206
Tl				

Fig. 1. The natural uranium ( $4n+2$ ) decay series.  
Modified from [1].

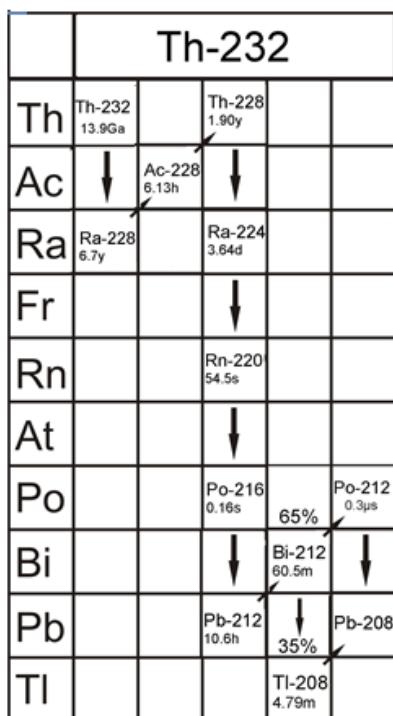


Fig. 2. The natural thorium (4n) decay series.  
 Modified from [1].

The total surface area of minerals, rocks, soils, sediments, as well as the concentration and distribution of  $^{238}\text{U}$  ( $^{226}\text{Ra}$ ) and  $^{232}\text{Th}$  ( $^{224}\text{Ra}$ ) in them, including the microscopic properties like the variable  $^{226}\text{Ra}$  and  $^{224}\text{Ra}$  distribution, surface roughness and/or nanopores network are chiefly the factors responsible by the radon and thoron emanation relatively to their production in the solid phases [5].

$^{226}\text{Ra}$  and  $^{224}\text{Ra}$  enrichment in grain boundaries would be able to control the radon and thoron supply to the surrounding fluid phases, additionally to an extensive network of 100-200 Å wide nanopores occurring in the grain surfaces and causing an increase of the internal surface area of the solid matrices, thus, constituting favorable paths for the radon and thoron diffusion into the atmosphere or water [6, 7].

$^{222}\text{Rn}$  is a radionuclide of great concern to humans. It is one of the most important sources of ionizing radiation from natural origin into which people are exposed, being considered by the World Health Organization (WHO) as the second largest cause of lung cancer (the first is smoking) [8].

In addition to  $^{222}\text{Rn}$  concerns, the  $^{220}\text{Rn}$  (thoron) contribution to human health has also been

recognized. It was unvalued for a long time due to its short half-life (about 55 seconds), as well as its availability in the environment. However, further studies have shown that in certain regions, exposure to thoron gas and its progeny can match or even exceed that of radon [9]. Because radon and thoron are often found together in the environment, for their analysis, it is necessary to use a methodology that discriminates them, avoiding mutual interferences [10].

Different approaches have focused on the thoron in environmental studies. For instance, Nuccetelli and Bochicchio [11] pointed out that the indoor concentration of thoron ( $^{220}\text{Rn}$ ) decay products is a relevant quantity from the health point of view, because, in certain not uncommon situations, such as when thorium-rich building materials are used, thoron may represent a significant source of radioactive exposure. These authors analyzed dose-exposure conversion factors often used for thoron decay products, highlighting the poorer basis of such factors when compared to those for radon ( $^{222}\text{Rn}$ ).

Porstendörfer [12] employed a lung dose model based on the structure of the ICRP 66 respiratory tract model to calculate the dose per exposure unit of the short-lived radon and thoron decay products. The dose conversion factors (DCF) of the radon and thoron decay products were estimated by [12], taking into account the unattached fraction of the decay product clusters and size distribution of the unattached and aerosol-attached decay products for different living and working places.

Tokonami et al. [13] reported measurements of natural radiation in cave dwellings located in Shanxi and Shaanxi provinces in the Chinese loess plateau. Among 193 dwellings, indoor radon concentrations ranged from 19 to 195 Bq/m<sup>3</sup> with a geometric mean of 57 Bq/m<sup>3</sup>, whereas indoor thoron concentrations ranged from 10 to 856 Bq/m<sup>3</sup> with a geometric mean of 153 Bq/m<sup>3</sup>, revealing that the presence of thoron and its decay products is not negligible, requiring special attention for accurate dose assessment in such an environment.

Yamada et al. [14] reported indoor radon and thoron measurements in cave dwellings of the Chinese loess plateau in Gansu province, where previously the Laboratory of Industrial Hygiene (LIH), China, and the U.S. National Cancer Institute

(NCI) had conducted an international collaborative epidemiological study. The LIH-NCI study showed an increased lung cancer due to the high residential indoor radon and thoron levels, which exhibited arithmetic mean concentrations of 91 and 351 Bq/m<sup>3</sup>, respectively. Similar to the results reported by [13], the presence of high thoron concentration was confirmed, and thoron was predominant over radon in the cave dwellings.

Nikezic et al. [15] reported DCF calculations based on the doses in the human lung per unit exposure to thoron progeny as estimated considering the model related to the ICRP 66 recommendations, as well as the DCF dependence on various environmental and subject-linked parameters. Also, the human lungs were considered as the source of beta and gamma radiation which target the other organs of the human body [15]. The DCF to other organs was obtained as 20  $\mu\text{Sv} \cdot \text{WLM}^{-1}$  for thoron progeny, which is larger than the DCF for radon progeny, corresponding to 13  $\mu\text{Sv} \cdot \text{WLM}^{-1}$  [15].

Thus, this short review highlighted the relevance of monitoring the thoron presence in certain environments due to the concerns associated with human health, in addition to the already well-developed studies focusing on radon. This should be done despite the analytical difficulties for conducting the measurements of the  $^{220}\text{Rn}$  activity concentration due to its shorter half-life compared to that of radon, i.e., about 6070 times lower. This paper describes an investigation focusing on the radon and thoron emanated from a soil cover occurring in the Campus of UNESP, Rio Claro city, Brazil. The data obtained were compared with other reports for groundwater; consequently, increasing the dataset of these noble gas isotopes in different environmental matrices.

## II. Study area

The experiment described in this paper was realized in a soil cover developed in the Campus of UNESP-São Paulo State University, Rio Claro city, São Paulo State, Brazil. UNESP spreads across 24 cities within São Paulo State, with two institutes settled at Rio Claro Campus: Geosciences and Exact Sciences Institute (IGCE), and Biosciences Institute (IB). Rio Claro city is located between the following coordinates (Fig. 3): parallels 22°14'37" and 22°33'16" S; meridians 47°27'57" and 47°46'00"

W. The investigated site is located in the Geology Department at IGCE-UNESP's Rio Claro Campus (Fig. 4).



Fig. 3. Location of Rio Claro city, São Paulo State, Brazil.

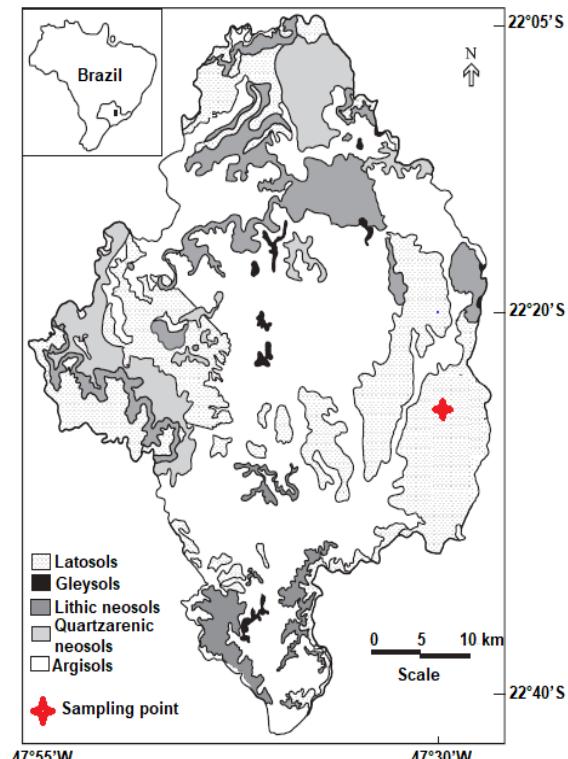


Fig. 4. Simplified pedological map of the Corumbataí River basin, São Paulo State, Brazil. Modified from [16].

The sampling point for radon and thoron analysis was located in the soil cover developed at the UNESP's Rio Claro Campus, within the Corumbataí River watershed (CRW) (Fig. 4). The dominant soil types in the CRW are classified as argisols, latosols, and neosols, with argisols and latosols covering about 65% of the surface area of the CRW [16, 17]. They are subdivided into different types, depending on their physicochemical characteristics, formation processes, diagnostic horizons, type of horizon arrangement, and clay activity, among others, which allow them to be separated into increasingly homogeneous units [17].

Color is often considered one of the main attributes in the classification of soils, indicating the richness in organic matter and mineralogical nature of the iron oxides present. In the Rio Claro municipality, the reddish color is distinctly visible throughout the territory, including the UNESP Campus [17].

Color also allows important inferences regarding the biogeochemical history of soils and morphological and environmental characteristics related to soil fertility. To reddish tones are considered: a) Red color with low levels of  $Fe_2O_3$  - presence (domain) of hematite; poor in trace elements and total P, with good drainage (translocation of water and nutrients); b) Red color, with high levels of  $Fe_2O_3$  - presence of hematite, magnetite and maghemite; richness in trace elements and total P; good drainage [17].

The argisols are soils formed by a very heterogeneous class that, in general, has in common a substantial increase in clay content at depth. They are well structured, have variable depth and predominantly reddish or yellowish colors, texture varying from sandy to clayey in the superficial horizons and from medium to very clayey in the subsurface; their fertility is varied, and the mineralogy is predominantly kaolinitic [17].

The latosols are soils resulting from energetic transformations in the material originating from or originating from pre-weathered sediments, where they predominate, in the clay fraction, as well as in minerals in the last stages of weathering (kaolinites, iron oxides, and aluminum). The existing sand fraction is dominated by highly resistant minerals to weathering, of variable texture, medium to very clayey, generally very deep, porous, soft, and permeable, with little difference in clay content in depth, and commonly have low natural fertility [17].

The sampling point at the UNESP's Campus is located in this soil type (Fig. 4).

The neosols are poorly evolved soils, with little expression of the processes responsible for their formation, which did not lead to significant modifications of the original material. They differ largely by their source material and landscape, such as sedimentary deposits (river plains, marine sediments, sandy or not) and regions with rugged relief [17].

### III. Experimental

In this study, the measurements of radon and thoron exhaled from the surficial soil layers were performed with the RAD7 equipment from Durridge Co. A driller was used to punch the subsurface soil to a depth of 1 m to collect the samples as also described by [18]. The probe inserted into the bore was made of stainless steel with the following dimensions: a length of 1 m, an internal diameter of 2 mm (in which the pipe connectors were inserted), and an external diameter of 6 mm (Fig. 5).



Fig. 5. General view of the probe used for sampling the radon and thoron from the surficial soil layers located at the Geology Department, IGCE-UNESP-Rio Claro Campus.

Soil was added between the borehole and the probe to seal the top hole. A hose with an internal diameter of 3 mm was connected to the probe apparatus (Fig. 6), with a microprocessor-controlled pump flow rate of 1 L/min for 15 minutes, in three cycles of 5 minutes each. The pump setting was in the Auto mode, meaning that the RAD7 switches the pump on and off according to a predetermined pattern that allows for sufficient sampling of air containing radon and thoron while conserving battery charge and pump wear. In the Auto pump setting, the pump always switches on for 4 minutes at the beginning of a new test cycle to ensure a good initial sample. If the humidity remains above 10%, then the pump stays on to allow the cell to dry out. Then the pump runs for just one minute in every five, until the end of the cycle [19].



Fig. 6. Probe coupling to RAD7 equipment during sampling the radon and thoron from the surficial soil layers located at the Geology Department, IGCE-UNESP-Rio Claro Campus.

The RAD7 utilizes a solid-state alpha detector, comprising a Si semiconductor material that converts the energy of the alpha particles into an electrical signal. The accumulation of many signals results in a spectrum that is presented on a scale of alpha energy in the range of 0-10 MeV. Such energy

interval is suitable for  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  readings as data acquisition takes place between 6 and 9 MeV [19].

The spectrum is displayed in a set of 200 channels grouped into eight windows of different energy range. A, B, C and D are the main windows, whereas E, F, G and H are the diagnostic of main windows (Fig. 7). Windows A and C provide  $^{222}\text{Rn}$  activity concentration data from  $^{218}\text{Po}$  and  $^{214}\text{Po}$  decays, representing “new” ( $^{218}\text{Po}$ ) and “old” ( $^{214}\text{Po}$ )  $^{222}\text{Rn}$ , respectively [19]. Windows B and D provide  $^{220}\text{Rn}$  activity concentration data from  $^{216}\text{Po}$  and  $^{212}\text{Po}$  decays, representing “new” ( $^{216}\text{Po}$ ) and “old” ( $^{212}\text{Po}$ )  $^{220}\text{Rn}$ , respectively [19]. Thus, the RAD7 separates the radon and thoron signals by energy of the alpha particles from their progeny, becoming possible to measure both Rn isotopes simultaneously [19].

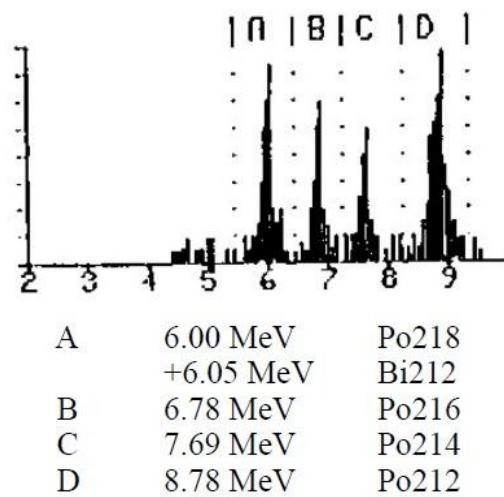


Fig. 7. Radon and thoron spectrum, according to [19]. Peaks in window A and/or C come from radon, while peaks in window B and/or D come from thoron.

Therefore, when radon decays inside the analysis chamber, it changes into  $^{218}\text{Po}$ , which adheres to the surface of the detector. When  $^{218}\text{Po}$  decays, alpha particles are released, forming  $^{214}\text{Pb}$ , and, afterwards,  $^{214}\text{Po}$  and  $^{210}\text{Pb}$  are produced. These radionuclides release particles with different energy values that are converted into electrical signals, which can be distinguished by the equipment [19].

Thus, the RAD7 uses electrostatic collection of alpha-emitting radon progeny ( $^{218}\text{Po}$ ,  $^{214}\text{Po}$ ) onto a silicon detector. The equipment draws

air at about 1 liter/minute through the detector chamber, utilizing electrostatic collection and alpha-particle counting, with the key steps including a purge to remove moisture and setting the test parameters, such as cycle time and number of cycles [19].

#### IV. Results and Discussion

Fig. 8 shows the alpha spectrum recorded by RAD7 during the data acquisition, which highlights a higher  $^{220}\text{Rn}$  peak (window B; 127 cpm) compared to the  $^{222}\text{Rn}$  peak (window A; 86.2 cpm) in the sample. From the three cycles of readings, it was possible obtain the following values for thoron: minimum = 53.9 kBq/m<sup>3</sup>; maximum = 57.5 kBq/m<sup>3</sup>; mean = 56.1 kBq/m<sup>3</sup>; standard deviation = 1.94 kBq/m<sup>3</sup>. The mean radon activity concentration corresponded to 38 kBq/m<sup>3</sup>; consequently,  $^{220}\text{Rn}$  is about 1.5 times higher than  $^{222}\text{Rn}$ , indicating its relevant emanation in the surficial soil layers studied.

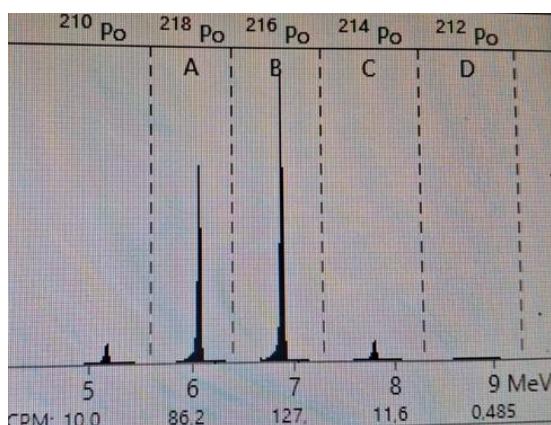


Fig. 8. Spectrum recorded by RAD7 for measuring radon (window A) and thoron (window B) emanated from the surficial soil layers located at the Geology Department, IGCE-UNESP-Rio Claro Campus.

The rock matrices responsible for the formation of the surficial soil layers play a major role in the soil chemical composition, including the radionuclides distribution. Among the lithologies occurring within the CRW, the Corumbataí Formation outcrops significantly, i.e., in 30.5% of its surface area [20]. Claystones and siltstones are the major sedimentary rock types occurring in the Corumbataí Formation. Low U/Th ratios between  $0.101 \pm 0.01$  and  $0.287 \pm 0.02$  were reported in these rocks by [21], indicating a larger preponderance of

thorium (7.10-15.21 ppm) compared to uranium (1.24-3.15 ppm) in them. This could explain the more accentuated emanation of thoron compared to radon in the surficial soil layers studied in this paper.

The mean radon and thoron levels of 38 kBq/m<sup>3</sup> and 56.1 kBq/m<sup>3</sup>, respectively, are higher than those reported by [13] and [14] in the caves of the Chinese Provinces, i.e., 57 and 91 Bq/m<sup>3</sup> for radon and 153 and 351 Bq/m<sup>3</sup> for thoron. Several factors imply that a difficult task is making the data comparison, the first one being the use of passive monitors in the surveys held in the Chinese caves, which is a methodological approach very different from that described in this paper.

Secondly, the diverse rock types and related genetic processes occurring at those sites in China and in Brazil certainly implied on variable concentrations of uranium and thorium in the minerals, drastically affecting the radon and thoron release from the solid matrices. Other possible reasons were already presented in the Introduction of this paper as pointed out by [5-7].

Anyway, the data reported by [13, 14] in the caves of the Chinese Provinces also revealed more enhanced levels of thoron compared to radon, similar to the finding of this paper, therefore, claiming special attention for thoron in dose radiation assessments.

In some groundwater studies, thoron has also been monitored in addition to radon. For instance, Salim and Bonotto [22] described a survey held in mineral waters occurring at the cities of Poços de Caldas, Águas de Lindóia, Águas de São Pedro, and Águas da Prata that are located in Minas Gerais and São Paulo States, Brazil. Twenty-three samples of water sources utilized for public consumption were analyzed by coupling the accessory RAD H<sub>2</sub>O to RAD7. The thoron and radon activity concentrations varied from 0.01 to 0.93 Bq/L and from 0.06 to 104 Bq/L, respectively. According to the Brazilian Code for Mineral Waters (BCMW), the thoron and radon activity concentration reported by [22] shows that none water source is thoriferous, whereas two are weakly radioactive. In 10 samples, the radon activity concentration was above the maximum allowed contamination level in water of 11 Bq/L as proposed by the US Environmental Protection Agency (USEPA).

In another groundwater survey, Bonotto [23] reported a wider dataset, comprising 75 water sources located in 14 municipalities of those Brazilian states (Minas Gerais and São Paulo). The  $^{222}\text{Rn}$  activity concentration range was 0.02-112.5 Bq/L, whereas the  $^{220}\text{Rn}$  activity concentration range was <0.004-0.90 Bq/L. The highest  $^{222}\text{Rn}$  levels were found in two springs located at Águas da Prata city (São Paulo State) and Dona Beja spring situated at Araxá city (Minas Gerais State), which are sites characterized by the abundant presence of natural radioelements in the rock matrices. A significant relationship was found between dissolved radon and thoron in the hypothermal and mesothermal waters, perhaps related to temperature effects on the dissolution of their parents,  $^{226}\text{Ra}$  and  $^{224}\text{Ra}$ , from the rock surfaces. According to the BCMW guidelines, none of the water sources were thoriferous (dissolved thoron levels lower than 2 Mache unit/L), whereas some waters from Águas da Prata and Araxá cities (Minas Gerais State) were weakly radioactive ( $^{222}\text{Rn}$  levels between 5 and 10 Mache unit/L) and radioactive ( $^{222}\text{Rn}$  levels between 10 and 50 Mache unit/L). Two spring waters exhibited  $^{222}\text{Rn}$  activity concentration exceeding the WHO guidance level of 100 Bq/L in drinking-water for public water supplies. The maximum  $\alpha$ -energy intake for radon progeny corresponded to  $0.4 \times 10^{-3}$  J, which is about 100 times lower than the IAEA guideline reference value. The maximum thoron progeny intake was  $5 \times 10^{-5}$  J that is almost 2600 times lower than the IAEA guideline value. Thus,  $^{220}\text{Rn}$  doses were much lower than those of  $^{222}\text{Rn}$  and the calculations described by [23] indicated that all analyzed mineral waters do not offer a health risk in terms of these radionuclides ingestion.

## V. Conclusion

The noble gas isotopes radon ( $^{222}\text{Rn}$ ) and thoron ( $^{220}\text{Rn}$ ) are radioactive, resulting from decays in the  $^{238}\text{U}$  and  $^{232}\text{Th}$  series, respectively. They have been monitored in certain environmental matrices, chiefly soils and waters, because are a subject of great concern to human health. Radon is one very important source of naturally occurring ionizing radiation that people are exposed to, being considered the second largest cause of lung cancer by the World Health Organization (WHO). On the other hand, compared to radon, only a few studies are reported in the literature focusing on thoron,

chiefly due to its short half-life of about 55 seconds and difficulties related to the analytical protocols involved. This study reported a comparative evaluation of the radioactivity due to  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  in surficial soil layers (latosols) sampled at UNESP-São Paulo State University, Rio Claro Campus, São Paulo State, Brazil, as well as in several well-known Brazilian mineral waters occurring at São Paulo and Minas Gerais states. The analytical protocols adopted allowed discriminating signals related to radon and thoron occurring in the soil and water samples. The thoron activity concentration in the soil sample surpassed the value obtained for the radon activity concentration, confirming the results of some studies reported for caves in Chinese Provinces. Contrarily, the dissolved radon activity concentration in the groundwater samples exceeded that of thoron, despite the expected higher thorium concentration compared to uranium in the rocks of the aquifer systems. The described dataset is relevant for further studies focusing on thoron in different environments, as promote new insights due to different trends related to radon occurring in soils and waters. Recent studies have also focused on using radon to track hydrogen emissions from soil covers, as bubble formations of hydrogen and methane are the only carriers of radon in the surficial soil layers and atmosphere. Under this scenario, thoron could also be utilized for such a purpose, as the results of this study indicated that its signals are even more powerful than those of radon.

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