



Use of radon isotopes, gamma radiation and dye tracers to study water interactions in a small stream in Brazil

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Abstract

In this study, natural (^{222}Rn) and fluorescent (uranin) tracers were used to investigate the interactions between surface and subsurface waters in a small hydrographical basin located in the southeast region of Brazil. Levels of ^{222}Rn were measured in 117 water samples with the use of an alpha solid-state detector. After the identification of the probable discharge sections along the stream, a measurement of the natural flows, upstream and downstream of these sections, was done with the use of a fluorimeter and fluorescent tracers. Also, scanning was done to verify a correlation between the natural gamma radiation and the ^{222}Rn in the areas where its activity was higher. The results showed some sections where the ^{222}Rn activity is more significant and contributed to the growth of the flows along the stream. It was possible to confirm a correlation between the discharge sections and the natural gamma radiation, what can be used as a preliminary approach to finding these sections in scenarios similar to the one studied here.

Keywords Tracers · Hydrology · Discharge sections · Radon

Introduction

The use of stable and radioactive isotopes as tracers to understand the behaviour of hydrological systems has increased in recent years, when adequate instrumentation for the detection and measurement of these tracers has become available. Isotopes are elements that are chemically identical, but with different mass numbers which are naturally present in the hydrosphere/atmosphere and may be used as natural environmental tracers. When applied intentionally to water for specific investigations, they are called artificial isotopic tracers. In many cases, the studied processes cover water used for public supply and, therefore, it is not desirable to discharge large volumes of artificial substances into rivers, lakes, aquifers or oceans. Thus, environmental isotopic

tracers are often recommended for applications in hydrology (Chen et al. 2014; Marques et al. 2014).

A tracer is any substance or particle, which can be used on an intermittent or continuous basis to assess the behaviour of a system, such as the flow characteristics of a groundwater flow system. A tracer for the study of water should be non-toxic, have low cost, should present unambiguous detection and quantification at minimum concentrations, must move with water and not disturb the system under study (Davis et al. 1980). In addition, it must be chemically stable, it must not be present in large quantities in the water studied and it must not be absorbed or filtered by the solid medium through which the water moves. In hydrology, tracers are classified either as environmental (inherent components of the water cycle) or as artificial (added to the system) (Leibundgut et al. 2009).

A tracer that has been used prominently in hydrology is ^{222}Rn , which has a half-life of 3.82 days and is produced by the alpha decay of the ^{226}Ra in the ^{238}U series. It is a chemically inert element, which does not cause undesirable biogeochemical reactions and does not change its physical characteristics. Also, radon is enriched in groundwater when compared to surface waters (Burnett et al. 2008). This feature occurs because groundwater is in direct

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contact with mineral grains containing radium which produces radon gas by decay. By contrast, surface water is subject to turbulence promoted by environmental factors, which allows the escape of radon (Cook et al. 2003). The enrichment of radon in groundwater will depend on the contents of its progenitor, radium, which is widely distributed in minerals in aquifer rocks. Radium is present in particularly high concentrations in granitic rocks and is less abundant in many metamorphic and sedimentary rocks (UNSCEAR 2000). Radon has a medium solubility in cold water which decreases when the temperature becomes higher, and this solubility helps it to escape from the rock structure after the decay of radio atoms, through fissures or nanopores, to the surface waters (Bonotto 2004).

The mechanisms that can cause the ^{222}Rn emanation from the rock fragments and, therefore, contribute to the content in the waters are (Bonotto and Lima 1997):

- the rebound of ^{222}Rn after the alpha decay of the ^{226}Ra atoms near the surface of the rock grain, positioned at a distance of about $0.036\ \mu\text{m}$, corresponding to the rebound distance of the ^{222}Rn molecule;
- the diffusion of ^{222}Rn through the crystalline matrix, from locations distant from the surface of the rock;
- the diffusion of ^{222}Rn along crystalline defects, grain contours or micro-fractures that are distant from the surface of the rock.

^{222}Rn has been used to detect groundwater exfiltration in Bangkok channels (Chanyotha et al. 2014), to study the changes of its content in waters when seismic activities occurred in the Western Caucasus (Nevinsky et al. 2015), for water flow in underwater caves (Csondor et al. 2017), and for developing a better understanding of subsurface and surface water interactions in a coastal aquifer in France (Mayer et al. 2016). In Switzerland, radon helped to evaluate the recharge dynamics of a karst aquifer, and provided information on a time scale of infiltration into the aquifer (Savoy et al. 2011). In fact, radon has been used to study water dynamics, its processes, interactions and connections in surface and subsurface systems, in places such as USA (Dimova et al. 2013), Italy (Tallini et al. 2013), Australia (Harrington et al. 2014), New Zealand (Close et al. 2014) and even in the Arctic lakes (Dugan et al. 2012), among others.

The goal of this work is to present some results of a research project that used ^{222}Rn as a tracer in a small hydrographic basin, and to analyze the correlation between the natural gamma radiation and the presence of radon in the area of study. Fluorescent tracers were also essential tools used during the project to measure the natural flows of the stream.

Study area

The Juatuba basin (Fig. 1) is located in the headwaters of the São Francisco River, drains an area of $442\ \text{km}^2$ and is approximately 60 km from Belo Horizonte (the capital of the state). The main watercourses that form the Juatuba River are the Serra Azul and Mateus Leme streams, which have a drainage area of 265 and $155\ \text{km}^2$, respectively (Ferreira et al. 2015).

The area of the Juatuba Basin is adjacent to a region named “Iron Quadrangle”—the largest Brazilian iron ore producer. According to a geological survey, this area consists of rocks from the Archeozoic, Lower Proterozoic and Cenozoic ages (Simmons 1968). The region shows a predominance of gneiss and grey granitic rocks, where the dominant minerals are quartz, orthoclase, muscovite, biotite and epidote (Drumond 2004; Pinto et al. 2010).

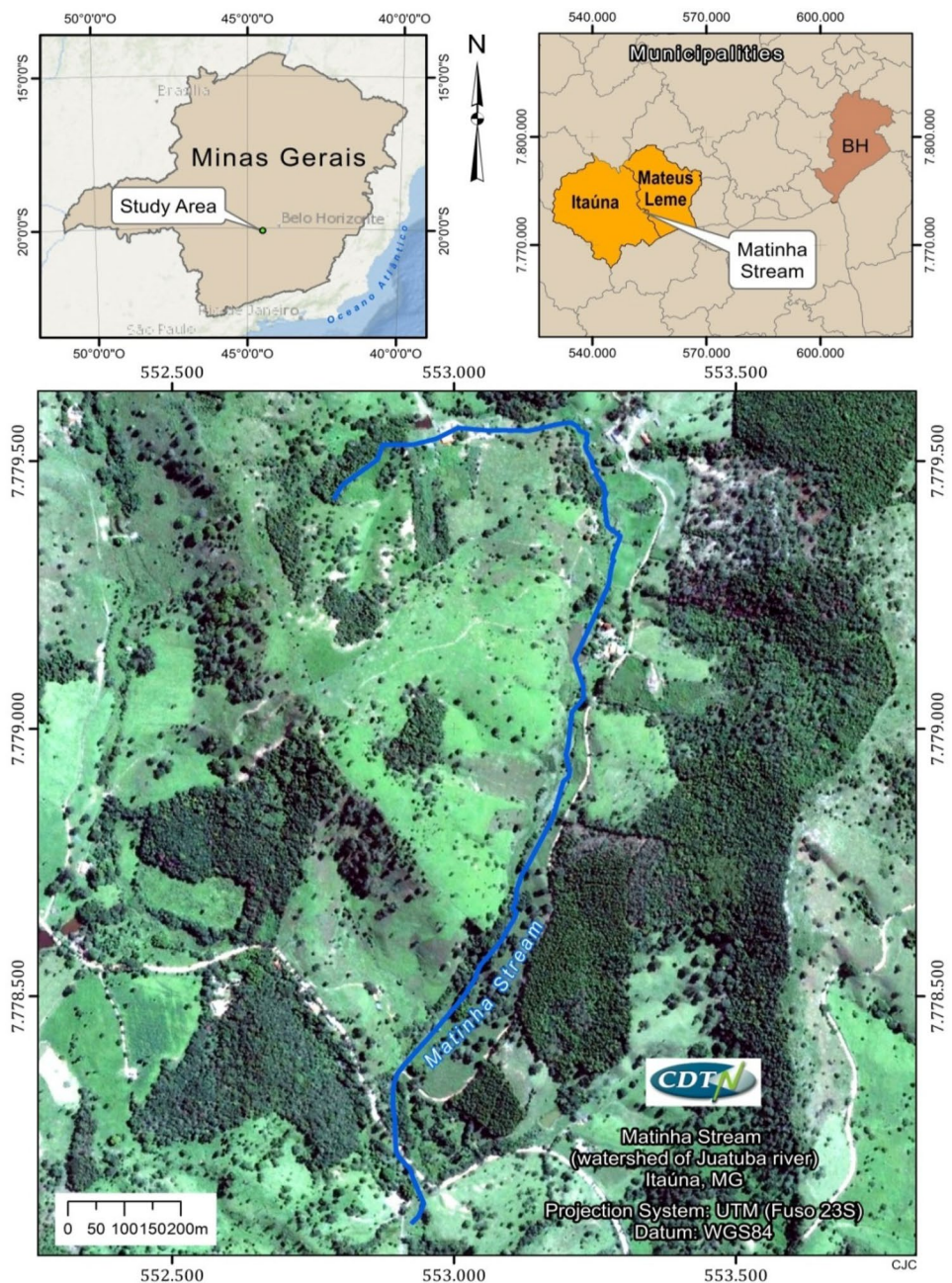
The Matinha stream belongs to the Juatuba basin and has an approximated area of $1.7\ \text{km}^2$. From the origin, until it meets the Mato Frio stream, it is approximately 2.2 km long (Chagas 2017).

Materials and methods

Radon detector (RAD 7)

In this equipment, there is an alpha solid-state detector (a semiconductor) that converts the alpha radiation into an electrical signal. The RAD 7 operates with a radon-in-water analysis kit known as RAD H_2O , that measures radon in water in a concentration range from less than $37 \times 10^{-5}\ \text{kBq L}^{-1}$ to more than $14.8\ \text{kBq L}^{-1}$. The RAD 7 is portable and powered by a battery. To obtain accurate radon reading in water, it should be after an approximate 30-min analysis with a sensitivity that corresponds or exceeds that present in liquid scintillation methods. For this study, all RAD 7 measurements were carried out using the protocol Wat 250, which means that glass flasks of 250 mL were filled with the water of the stream, and analyzed as soon as possible. In the first two sampling campaigns, a hypodermic syringe (with a capacity of 100 mL) was used to collect the water samples. However, this procedure took a long time. To optimize the process, a system composed of a peristaltic pump powered by a 12 V battery, and a pair of thin rubber hoses replaced the syringe and were used from the third to the last campaign. Also, a collector pipe was used, placed close to the bottom of the stream, and the presence of a metal screen with a small mesh avoided the collection of sediments through this device. All the water samples were collected along the stream in different campaigns.

Fig. 1 Location of the study area (coordinates UTM 23S—WGS84)



It is valid to observe that the RAD 7 has already been used in several projects that involve measurements of radon in water (Akawwi 2014; Ravikumar et al. 2014; Krishan et al. 2015; Le et al. 2015; Somashekar and Ravikumar 2010).

Gamma radiation monitor

The AT6101C spectrometer (Atomtex®), which is connected via Bluetooth to a handheld PC (HPC NAUTIZ X7®), is responsible for the immediate interpretation of the data received and its storage through the ATAS Scanner Mobile

software was used under the scope of this project. Its operation consists of calibrating the gamma detector with an accessory standard, passing through the study area and collecting the data. The equipment has an estimated weight of 7 kg, is inserted in a backpack, and has the ability to detect the presence of neutrons and/or gamma radiation. The device identifies the composition of the nuclide on finding a source of radiation (ATOMTEX 2017). Detection of gamma radiation is done using a sodium iodide inorganic scintillator activated through thallium, and the neutron detection uses two proportional counters in a polyethylene moderator (Aramburu and Bisbal 1996).

One of the advantages of the AT6101C Scanner is the storage capacity of the radiation dose data ($\mu\text{Sv h}^{-1}$) along with the geographical reference in UTM (WGS84), facilitating the transit of data in SIG type software and the visualization of these points in Google Earth.

Fluorimeter GGUN FL 30

This device has been already used in several experiments that include flow measurements and fluorescent tracers such as uranin or rhodamine (Meus et al. 2014; Schneider et al. 2014; Lemke et al. 2013). The device was placed underwater and the tracer cloud passage can be seen in real time on a notebook connected to the device. After this step, the FLUO software calculates the value of the natural flow of the watercourse under study.

The amount of tracer to be injected is quantified according to the reading ability of the fluorimeter (Gardner and Ely 1967), preventing saturation:

$$C(x, t) = \frac{A}{2a\sqrt{\pi kt}} \exp\left[-\frac{(x-ut)^2}{4kt}\right], \quad (1)$$

where x is the measurement point, t the time elapsed between the injection and the passage of the cloud, A the mass of the injected tracer, u the mean linear flow, a the cross-sectional area, k the dispersion coefficient, and $C(x, t)$ the concentration of the tracer. The distance between the points of injection and sampling was obtained applying the model proposed by Ward (1973):

$$D = 500KW^2h^{-1}, \quad (2)$$

where K is 0.08 for central injection, W is the mean length and h is the mean depth.

The calibration process of the GGUN is done twice. First, at the lab, standard solutions of deionised water, of tracers (10,000 ppb) and of turbidity (100 NTU), are used. Then in the field, the probe is inserted for 5 min in a bucket with 4950 mL of water of the stream under study, and 50 mL of tracer to be injected (100 ppb solution). It is valid to observe here that turbidity increases the baseline of the signal by directing excitation light into the detection optics. But this baseline can be measured by the GGUN in the stream before the plume and is subtracted. The turbidity can absorb a small amount of excitation light, reducing the signal. However, the procedure done earlier (bucket calibration) using the stream water has taken care of this effect (Schneeg et al. 2011).

Field experiments

In all, ten field experiments were done with several purposes. In seven campaigns, water samples were collected to determine the activity of the ^{222}Rn concentration using the

RAD 7. In five campaigns (where four of them were carried out simultaneously with water sampling), the natural flows of the Matinha stream were measured using the GGUN FL-30, and the measurements of the natural gamma radiation were made in a single campaign using the AT6101C spectrometer. The stream was monitored along its thalweg; however, in the sections where it was not possible to walk in the streambed, the monitoring was done on the banks of the stream, aiming to verify the existence of a correlation between the activity of ^{222}Rn in the waters of the stream and the natural gamma radiation. These measurements were only made at a limited number of locations near the stream source due to the presence of dense vegetation, fences and mires. The limited access near two artificial wetlands along the stream course also restricted the number of gamma radiation measurements that could be made. Otherwise, gamma radiation measurements could be made at regular intervals on-foot for a distance of 2.25 km along the stream.

All the radon analysis values in the water samples were corrected due to the half-life of ^{222}Rn and the elapsed time between sample collection and reading

$$A(t) = A_0 \times e^{-\lambda t}, \quad (3)$$

where $A(t)$ is the activity of ^{222}Rn obtained when the sample was analyzed, A_0 is the initial activity of the ^{222}Rn in the moment of the sampling and λ is the decay constant of the ^{222}Rn ($=\ln 2/T_{0.5}$, where $T_{0.5}$ is the half-life of ^{222}Rn).

The water sampling was carried out in a period of low rainfall in the region. The record of rainfall between 1977 and 2015 in a rainfall station about 2.5 km from the Matinha stream confirmed this. The accumulated rainfall between 2014 and 2015 is below the linear trend line (Fig. 2). There has been a downward trend in rainfall in the region over the last 20 years, except for the hydrological years 1996/1997, when precipitation exceeds 2000 mm.

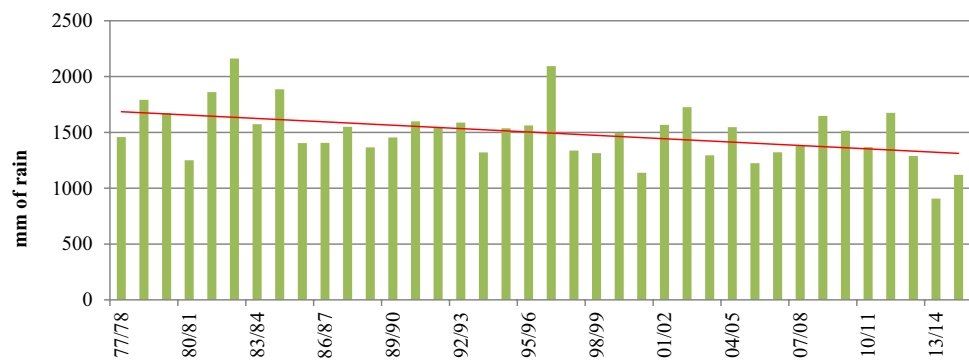
Results

Table 1 presents the activity of ^{222}Rn in the water samples collected throughout the project. It is possible to verify the presence of some sections where the values are more significant than others, which can indicate the presence of groundwater discharge zones.

Figure 3 shows the values of the natural gamma radiation along the stream, and Fig. 4 presents the normal distribution of the values. The results show that 95% of the natural gamma radiation registered along the watercourse had a dose rate between 0.0426 and 0.1447 $\mu\text{Sv h}^{-1}$.

It is valid to observe that during the whole period of this study, a pluviograph previously installed in the study area (Ferreira et al. 2011) recorded a cumulative rainfall of 1120 mm for the hydrological year 2014/2015. The rainfall

Fig. 2 Data of a rain gauge located close to the studied area



registered by this pluviograph was below the average of the historical series registered in the rain gauge installed close to the study area, which was 1525 mm.

Table 2 shows the results of flow measurements done along the watercourse during the execution of the project with the help of the fluorimeter. It is possible to verify that the values become more significant from upstream to downstream what is also an indicative of the presence of discharge zones along the watercourse. For all measurements, the recovery rate of the tracer was obtained based on the same methodology presented by Metcalf and Eddy (2003).

Discussion

The discharge of groundwater was the primary source of water for the Matinha stream because there was no rainfall throughout the study period, making the concentrations of ^{222}Rn more evident and relatively higher in the observed interconnection sections. Excessive rainfall during this period could have caused the surface water in the stream to recharge the aquifer, reducing radon activity (Sophocleous 2002; Baskaran et al. 2009). Since the study area is a rural site, all the gamma radiation detected comes from radioactive isotopes that are naturally present in the soils, water, rocks and the environment. The average exposure of the radiation found was $0.094 \mu\text{Sv h}^{-1}$ which is equivalent to $0.82 \text{ mSv year}^{-1}$. This value, in the table of classification of the areas of radioactivity according to the effective dose of the natural terrestrial gamma radiation, is given as normal for mean doses smaller than $5.0 \text{ mSv year}^{-1}$.

Examples of areas with more natural radioactivity are China (regions of Dong-anling and Tongyou), India (Karunagappally), Iran (Ramsar and Mahallat), Brazil (municipality of Poços de Caldas) and Sudan (Kurrun Mountain and Ourro area). Studies done in the above areas presented values much higher than the ones found in the surroundings and along the Matinha stream (Saad et al. 2014; Secretariat of Health of Minas Gerais State 2009; Sohrbai 2013; Aliyu and Ramli 2015).

The radiometric map of the studied region presents an interpolation of 50 m for each side of the stream banks, as also in its thalweg. Figure 5 shows the natural gamma radiation spatial distribution registered during the field works. In all, 4168 points were measured in June 2016 with the ATC6101C, and all values are in the range $0.045\text{--}0.219 \mu\text{Sv h}^{-1}$.

Based on the results of the analyses of the water samples, it was possible to find four sections where there is a connection between subsurface and surface waters, represented by the higher values of ^{222}Rn (Fig. 6). The first discharge area is located between 350 and 500 m from the start of the transect, and the second one between 600 and 800 m. These two sets presented similar mean values of radon activity which are 32.1 and 28.3 Bq m^{-3} , respectively. The third discharge area, which is located between 1150 and 1350 m from the start of the transect, as well as the fourth one, located between 1800 and 2000 m from the start of the transect, also presented similar mean values of activity of 19.8 and 22.2 Bq m^{-3} , respectively.

The ^{222}Rn activities obtained along the stream are useful for understanding the extent of hydrological processes at the stream–aquifer interface. The results of the sampling of this environmental isotope indicated that the upper area of the stream is connected and actively recharged by “near stream–shallow alluvial aquifer” (Baskaran et al. 2009). These authors state that there is a hydraulic connection between groundwater and surface water in many landscapes.

However, it is valid to emphasize the complexity of the several phenomena that took place in the hyporheic zone (HZ). Bencala (2010) stated that the understanding of the hydrological processes that occur in the HZ is critical for the knowledge of the stream ecosystems. Boano et al. (2014) discussed 50 years of researches in this zone, debating issues such as fluvial ecosystems, water quality and restoration of the whole river environment, field observations and the theories proposed. The dynamics of some processes that happen in the HZ were modelled (Briggs et al. 2010; Cardenas 2009; Endreny et al. 2011; Krause et al. 2014) aiming to enhance

Table 1 Results of the measurements: ^{222}Rn activity

Distance from the source (m)	Time and date of the sampling	Radon activity (Bq m^{-3})	Distance from the source (m)	Time and date of the sampling	Radon activity (Bq m^{-3})
0	14/09/16—16:05	7.31 ± 2.37	392	15/07/15—15:25	20.45 ± 2.59
2	14/11/18—14:07	5.86 ± 1.03	397	15/07/15—15:17	46.70 ± 2.72
4	14/11/18—14:15	6.83 ± 1.68	401	15/02/11—11:45	27.90 ± 2.45
129	14/11/18—14:36	43.14 ± 2.58	402	15/07/15—15:07	17.01 ± 1.59
353	15/07/15—16:28	37.53 ± 2.34	407	15/07/15—15:00	17.04 ± 2.42
357	15/02/11—10:40	49.41 ± 3.21	412	15/07/15—14:48	12.30 ± 2.13
358	15/07/15—16:20	30.88 ± 1.33	412	15/02/11—11:55	26.93 ± 2.80
360	14/11/18—15:05	76.96 ± 6.36	418	15/07/15—14:40	17.56 ± 2.62
363	15/02/11—10:50	58.04 ± 0.91	422	15/02/11—12:10	35.71 ± 1.49
364	15/07/15—16:13	30.27 ± 4.49	423	15/07/15—14:30	21.02 ± 1.98
370	15/07/15—16:05	23.78 ± 4.47	427	14/11/18—15:20	48.18 ± 4.09
375	15/07/15—15:55	57.69 ± 1.97	432	15/02/11—12:20	28.65 ± 1.12
380	15/02/11—11:15	31.53 ± 4.23	442	15/02/11—12:30	35.98 ± 3.07
382	15/07/15—15:45	25.56 ± 2.32	452	14/09/16—16:25	1.03 ± 1.13
387	15/07/15—15:35	28.32 ± 2.83	456	15/02/11—12:43	22.06 ± 3.18
391	15/02/11—11:30	28.52 ± 3.18	466	15/02/11—12:50	19.21 ± 1.40
477	15/02/11—13:05	21.29 ± 0.81	1140	16/05/16—17:05	2.89 ± 0.80
523	14/11/18—15:40	14.42 ± 1.65	1155	16/05/16—16:55	25.45 ± 3.55
561	14/11/18—15:55	6.33 ± 1.38	1158	14/11/20—09:29	38.31 ± 2.94
562	14/09/16—16:55	35.09 ± 4.86	1165	14/11/20—09:25	26.44 ± 1.78
627	15/07/15—12:40	24.67 ± 1.65	1167	14/11/19—15:40	42.60 ± 1.68
631	15/07/15—12:35	17.10 ± 1.63	1170	16/05/16—16:43	23.69 ± 1.86
633	15/02/10—14:40	35.92 ± 3.67	1171	14/11/20—09:36	27.54 ± 1.91
633	15/07/15—13:30	10.09 ± 0.82	1185	16/05/16—16:25	17.80 ± 0.90
635	14/09/16—17:10	9.36 ± 2.68	1200	16/05/16—16:15	23.30 ± 1.60
637	15/07/15—12:20	14.34 ± 1.89	1218	16/05/16—16:00	25.18 ± 2.30
638	15/02/10—14:55	52.63 ± 4.92	1232	16/05/16—15:50	18.76 ± 0.89
642	15/07/15—12:00	42.19 ± 2.21	1246	16/05/16—15:40	19.20 ± 0.43
643	14/09/17—11:05	26.43 ± 4.32	1261	16/05/16—15:25	13.90 ± 1.13
643	15/02/10—15:06	71.14 ± 5.09	1276	16/05/16—15:05	4.99 ± 0.74
647	15/07/15—11:50	14.27 ± 2.14	1280	15/09/29—09:55	3.51 ± 1.30
648	15/02/10—15:30	15.80 ± 1.03	1292	16/05/16—14:55	24.23 ± 2.32
652	15/07/15—11:40	53.70 ± 1.86	1293	15/09/29—09:45	3.05 ± 1.23
654	15/02/10—15:45	20.24 ± 1.28	1297	14/11/19—16:00	13.11 ± 0.53
657	15/07/15—11:15	52.58 ± 4.16	1303	15/09/29—09:35	2.45 ± 1.40
661	15/02/10—15:55	28.81 ± 0.78	1305	16/05/16—14:40	22.47 ± 0.65
662	15/07/15—11:00	54.39 ± 2.50	1379	14/11/19—16:23	8.25 ± 0.78
667	15/07/15—10:45	12.92 ± 1.21	1550	14/09/17—11:45	7.13 ± 0.55
672	15/07/15—10:31	25.94 ± 2.04	1585	16/05/16—11:00	4.56 ± 1.51
677	15/07/15—10:20	10.50 ± 1.13	1662	14/11/19—17:00	0.52 ± 0.31
680	15/09/28—18:10	32.44 ± 2.92	1765	14/11/19—17:21	4.72 ± 0.53
694	14/11/19—09:38	52.26 ± 4.78	1834	15/09/29—15:05	26.76 ± 1.76
701	15/09/28—18:03	21.20 ± 1.50	1840	14/09/17—12:00	5.53 ± 0.67
715	15/09/28—17:50	9.89 ± 1.95	1846	15/09/29—14:55	38.07 ± 2.10
724	14/11/19—09:45	22.07 ± 1.93	1864	15/09/29—14:42	29.31 ± 2.80
735	15/09/29—11:20	5.08 ± 0.81	1880	15/09/29—14:26	7.28 ± 1.60
750	15/09/29—09:53	12.13 ± 1.17	1906	15/09/29—14:12	31.95 ± 2.78
760	15/09/28—17:07	1.63 ± 0.44	1913	14/11/20—09:55	13.77 ± 0.33
770	14/11/19—10:11	46.07 ± 3.59	1926	15/09/29—14:05	39.68 ± 4.25

Table 1 (continued)

Distance from the source (m)	Time and date of the sampling	Radon activity (Bq m ⁻³)	Distance from the source (m)	Time and date of the sampling	Radon activity (Bq m ⁻³)
773	15/09/28—16:50	4.77 ± 0.65	1938	15/09/29—13:52	28.89 ± 0.96
779	15/09/28—16:33	6.25 ± 0.52	1956	15/09/29—13:47	16.35 ± 1.55
795	15/09/28—16:20	2.93 ± 0.83	1975	15/09/29—13:22	14.95 ± 2.50
819	14/09/17—10:40	33.11 ± 4.84	1997	15/09/29—13:12	9.36 ± 0.93
858	14/11/19—10:37	16.20 ± 3.02	2003	14/11/20—10:05	5.83 ± 1.13
915	14/11/19—10:54	11.50 ± 1.27	2007	15/09/29—13:00	7.78 ± 1.42
1044	14/11/19—11:16	2.00 ± 0.32	2060	14/11/20—10:14	2.08 ± 0.74
1092	14/09/17—11:20	12.38 ± 1.08	2152	14/11/20—10:25	1.33 ± 0.30
1111	16/05/16—17:30	2.92 ± 0.98	2157	14/09/17—12:30	9.50 ± 1.38
1125	16/05/16—17:20	2.00 ± 0.63	–	–	–

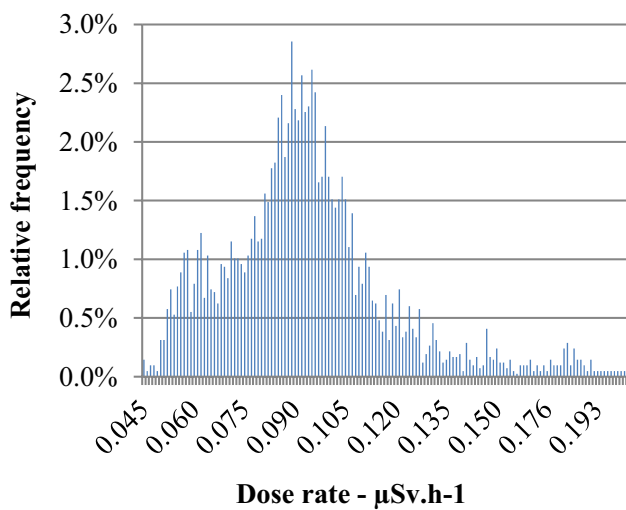


Fig. 3 Natural gamma radiation along the Matinha stream

the comprehension of the many variables that are involved in the interactions that happen in it.

Furthermore, Ciszewski and Aleksander-Kwaterczak (2016) proposed the HZ as a tracer to analyze the pollution of the waters. Schaper et al. (2018) verified that the HZ acts as an efficient bioreactor, capable of removing polar trace organic compounds along relatively short flow paths. A 3-year sampling of dissolved organic matter (DOM) in an Alpine stream found a coupling of DOM dynamics in the streamwater and the HZ (Fasching et al. 2015). Mendoza-Lera and Datry (2017) studied the restoration of the river ecosystem, attempting to increase the biogeochemical processes by increasing the hyporheic flow. Since their relationship is nonlinear, they presented a conceptual model where a Gaussian function, depending on hyporheic hydraulic conductivity, can correlate them.

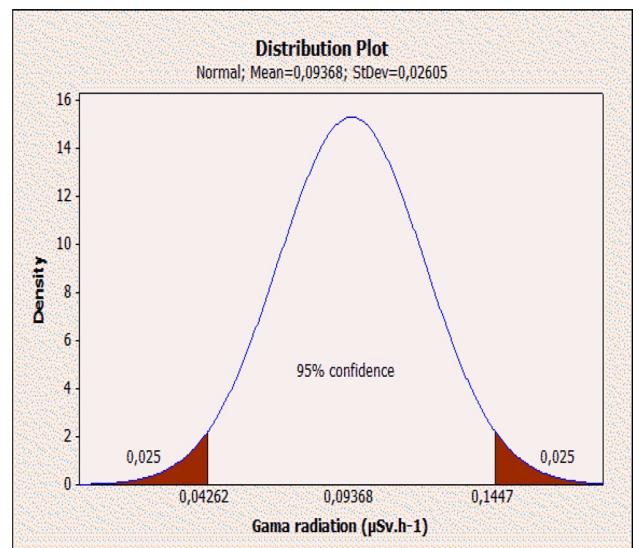
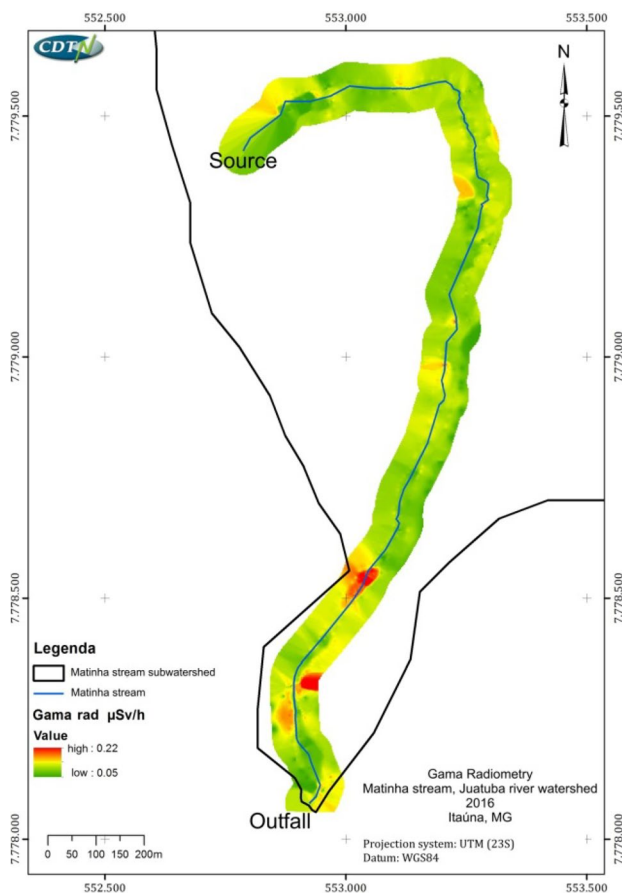


Fig. 4 Normal distribution of the gamma radiation registered along the Matinha stream

The response of the HZ to fluctuations and extreme events showed changes in the denitrification rates, biogeochemical cycling and natural attenuation, among other factors (Trauth and Fleckestein 2016; Malzone et al. 2015; Fuller and Bargar 2014; Zimmer and Lautz 2013; Sawyer et al. 2014). According to these examples, it is possible to see that this is a very important and complex ecosystem (Jones and Stanley 2016; Liu et al. 2017; Peralta-Maraver et al. 2018), and to understand it, “a technological expertise is required but also the knowledge about the natural processes that structures the interface” (UK Environment Agency 2009). The monitoring of some parameters is important, but not enough to find the desired answers. Even the results of the tests done with environmental tracers in the HZ should be analyzed carefully (Wondzell 2006).

Table 2 Results of the flow measurements along the Matinha stream ($L s^{-1}$)

Date	Distance from the source (m)	Flow ($L s^{-1}$)	Transit time of the tracer (s)	Recovery rate of the tracer (%)	Coordinates UTM 23S	Mass injected (mg)
15/07/15	350	1.75 ± 0.05	2424	76	553,090–7,779,558	25
15/07/15	500	2.10 ± 0.06	2196	78	553,234–7,779,552	40
15/12/16	350	1.19 ± 0.05	2064	83	553,090–7,779,558	30
15/12/16	700	2.10 ± 0.09	1620	86	553,283–7,779,369	30
15/12/16	2150	3.54 ± 0.14	372	95	552,931–7,778,079	30
16/05/10	700	2.59 ± 0.11	4422	78	553,283–7,779,369	50
16/05/10	1600	3.06 ± 0.12	1640	94	553,047–7,778,558	100
16/05/24	350	1.83 ± 0.07	3315	92	553,090–7,779,558	100
16/05/24	2150	4.76 ± 0.19	900	95	552,931–7,778,079	100

**Fig. 5** Gamma radiometry in the surroundings and in the thalweg of the stream

When the correlation between ^{222}Rn and the natural gamma radiation is observed in the Matinha stream (Fig. 7), it is possible to see a similarity in the studied sections. The main difference is close to the waterfall, where there is a

peak of gamma radiation but the activity of ^{222}Rn decreases. This is probably caused by the dispersion of the radon in the air due to the turbulence in that area, which facilitates the escape of ^{222}Rn . So, since along the stream the areas where the natural gamma radiation and ^{222}Rn activity in levels higher than the background are coincidental, a previous scan of a watercourse can be a useful tool to help in finding the introductory sections where ^{222}Rn can be found in experiments with goals similar to this one.

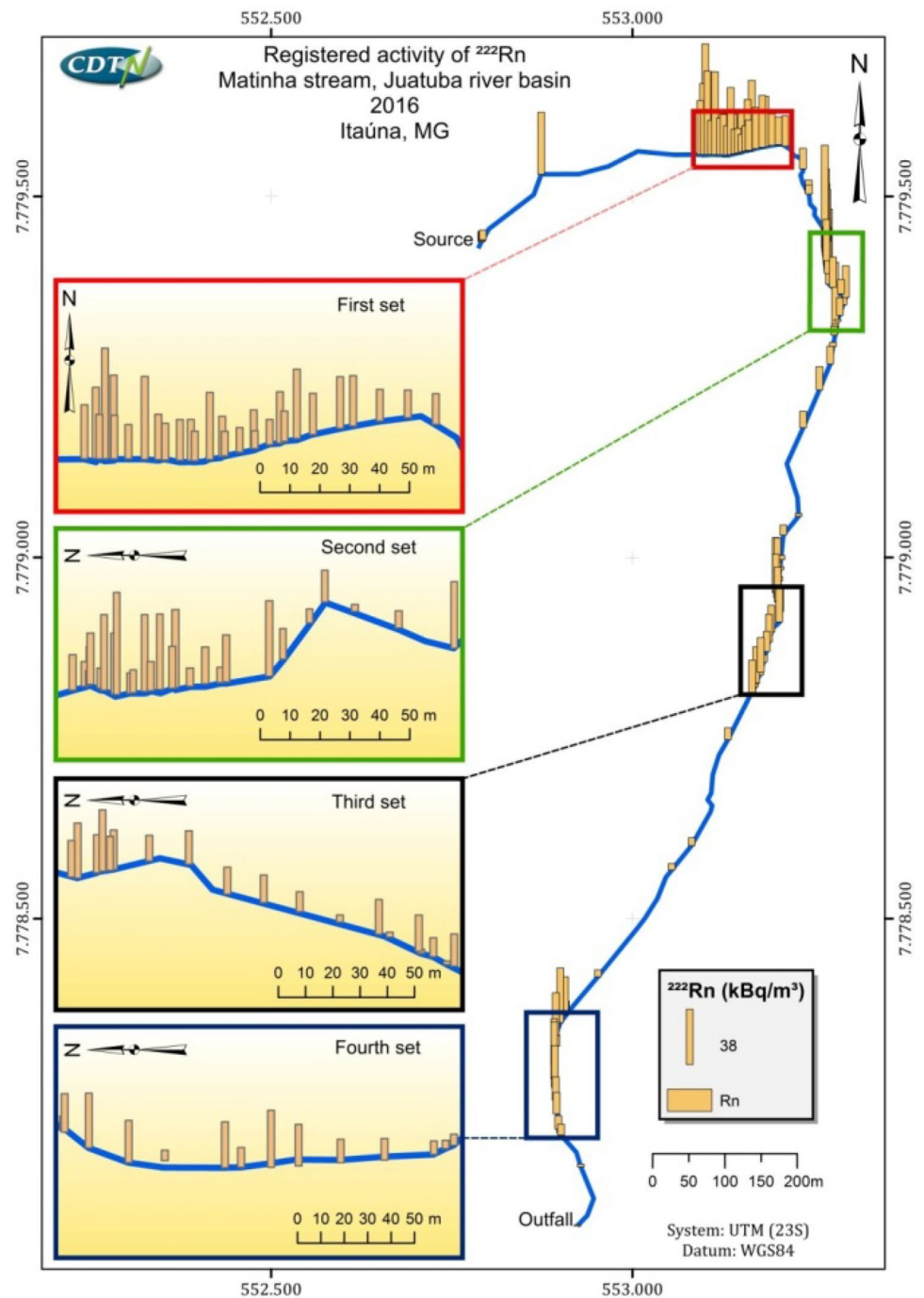
Considering that during this work, the water samplings were done along the whole stream but in different campaigns, it was not possible to obtain a mathematical equation that could correlate the growth of the flows and the activity of ^{222}Rn in a continuous way. However, as an example of some assumptions that could be made, Fig. 8 shows the values of the experiments done in May 2016, in a section of the stream. It is possible to see the growth of the natural flow, as the values of radon activity, where some points present values much higher than others, what indicates the existence of the discharge into the Matinha stream.

Since the discharge sections were found in the stream, in a future project, several measurements of ^{222}Rn could be done every 5 m, and the activity above the background values could be used to obtain an equation that could also include the growth of the natural flow, with measurements taken upstream and downstream of the discharge sections.

Conclusions

A natural gamma radiation scanning can be a useful tool for locating groundwater discharge areas in streams or small rivers and can help identify areas where detailed sampling should be carried out. The amount of fluorescent tracers used was very small, and the results of the plot combining

Fig. 6 Activity of ^{222}Rn along the stream course



the ^{222}Rn activity along the stream in conjunction with the flows measured were an effective way of analyzing the relationship between stream flow rate and the activity of ^{222}Rn .

Finally, it was observed that in post-monsoon and low-rainfall periods, ^{222}Rn isotopes are useful tracers for the identification sections of the stream where groundwater takes place. The ability to identify discharge areas can

help with the rehabilitation of streams that are affected by groundwater contamination. However, the HZ is a complex study area and many variables must be studied for a better understanding of the processes that occur in it. It has a high biodiversity and a strong chemical reactivity, the environment is heterogeneous, dynamic, and difficult to study.

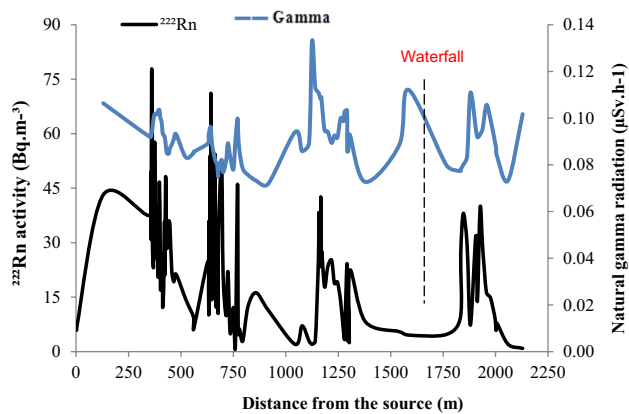


Fig. 7 Correlation between ^{222}Rn and gamma radiation along the stream

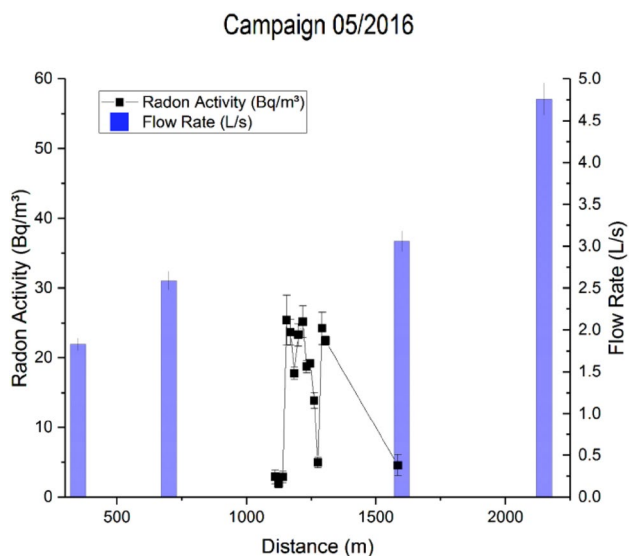


Fig. 8 Results of the radon activity and the flow rate in a section studied in May 2016

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References

- Akawwi E (2014) Radon-222 concentrations in groundwater along Eastern Jordan Rift. *J Appl Sci* 14(4):309–316. <https://doi.org/10.3923/jas.2014.309.316>
- Aliyu AS, Ramli AT (2015) The world's high background natural radiation areas (HBNRAs) revisited: a broad overview of the

- dosimetric, epidemiological and radiobiological issues. *Radiat Meas* 73:51–59. <https://doi.org/10.1016/j.radmeas.2015.01.007>
- Aramburu XO, Bisbal JJ (1996) Radiaciones ionizantes: su utilización y riesgos, 1st edn. Ed. Univ. Politèc. de Catalunya, Barcelona
- ATOMTEX (2017) Instruments and technologies for nuclear measurements and radiation monitoring. <http://www.atomtex.com/en/products/radionuclide-identification-devices-rids/at6101c-spectrometer/>. Accessed 25 Jan 2017
- Baskaran S, Ransley T, Brodie RS, Baker P (2009) Investigating groundwater–river interactions using environmental tracers. *Aust J Earth Sci* 56(1):13–19. <https://doi.org/10.1080/08120090802541887>
- Bencala KE (2010) Hyporheic zone hydrological processes. *Hydrol Process* 14:2797–2798. [https://doi.org/10.1002/1099-1085\(20001030\)14:15%3C2797::AID-HYP402%3E3.0.CO;2-6](https://doi.org/10.1002/1099-1085(20001030)14:15%3C2797::AID-HYP402%3E3.0.CO;2-6)
- Boano F, Harvey JW, Marion A, Packman AI, Revelli R, Ridolfi L, Wörman A (2014) Hyporheic flow and transport processes: mechanisms, models, and biogeochemical implications. *Rev Geophys* 52:603–679. <https://doi.org/10.1002/2012RG000417>
- Bonotto DM (2004) Radioactivity in the waters: from England to Guarani [in Portuguese]. Editora UNESP, São Paulo
- Bonotto DM, Lima JLN (1997) Transfer of radon 222 from the rocks of the Poços de Caldas Plateau for the waters [in Portuguese]. *Braz J Geosci* 27(2):189–198
- Briggs MA, Gooseff MN, Peterson BJ, Morkeski K, Wollheim WM, Hopkinson CS (2010) Surface and hyporheic transient storage dynamics throughout a coastal stream network. *Water Resour Res*. <https://doi.org/10.1029/2009WR008222>
- Burnett WC, Peterson R, Moore WS, Oliveira J (2008) Radon and radium isotopes as tracer of submarine groundwater discharge—results from the Ubatuba, Brazil SGD assessment intercomparison. *Estuar Coast Shelf Sci* 76(3):501–511. <https://doi.org/10.1016/j.ecss.2007.07.027>
- Cardenas MB (2009) A model for lateral hyporheic flow based on valley slope and channel sinuosity. *Water Resour Res*. <https://doi.org/10.1029/2008WR007442>
- Chagas CJ (2017) The use of radon as a tracer for the identification of discharge of aquifers along the Matinha stream, MG [in Portuguese]. Dissertation. Federal University of Minas Gerais—Brazil
- Chanyotha S, Chutima K, Burnett WC, Lane-Smith D, Simko J (2014) Prospecting for groundwater discharge in the canals of Bangkok via natural radon and thoron. *J Hydrol* 519(B):1485–1492. <https://doi.org/10.1016/j.jhydrol.2014.09.014>
- Chen J, Liu X, Sun X, Su Z, Yong B (2014) The origin of groundwater in Zhangye Basin, northwestern China, using isotopic signature. *Hydrogeol J* 22:411–424. <https://doi.org/10.1007/s10040-013-1051-7>
- Ciszewski D, Aleksander-Kwaterczak U (2016) Contrasting sediment and water chemistry indicates the extent of the hyporheic zone in a polluted river system. *Geol Geophys Environ* 42(2):151–159. <https://doi.org/10.7494/geol.2016.42.2.151>
- Close M, Matthews M, Burbery L, Abraham P, Scott D (2014) Use of radon to characterise surface water recharge to groundwater. *J Hydrol* 53(2):113–127
- Cook PG, Favreau G, Dighton JC, Tickell S (2003) Determining natural groundwater influx to a tropical river using radon, chlorofluorocarbons and ionic environmental tracers. *J Hydrol* 277:74–88. [https://doi.org/10.1016/S0022-1694\(03\)00087-8](https://doi.org/10.1016/S0022-1694(03)00087-8)
- Csondor K, Eröss A, Horváth A, Szieberth D (2017) Radon as a natural tracer for underwater cave exploration. *J Environ Radioact* 173:51–57. <https://doi.org/10.1016/j.jenvrad.2016.10.020>
- Davis SN, Thompson GM, Bentley HW, Stiles G (1980) Groundwater tracers—a short review. *Ground Water* 18(1):14–23
- Dimova NT, Burnett WC, Chanton JP, Corbetta JE (2013) Application of radon-222 to investigate groundwater discharge into small

- shallow lakes. *J Hydrol* 486:112–122. <https://doi.org/10.1016/j.jhydrol.2013.01.043>
- Drumond MM (2004) The tracer technique and its potential to increase the hydrological knowledge on the Brazilian basins: a study applied to the Juatuba Representative Basin—MG [in Portuguese]. Thesis. Federal University of Minas Gerais—Brazil
- Dugan HA, Gleeson T, Lamoureux SF, Novakowski K (2012) Tracing groundwater discharge in a high Arctic lake using radon-222. *Environ Earth Sci* 66(5):1385–1392. <https://doi.org/10.1007/s12665-011-1348-6>
- Endreny T, Lautz L, Siegel DI (2011) Hyporheic flow path response to hydraulic jumps at river steps: flume and hydrodynamic models. *Water Resour Res*. <https://doi.org/10.1029/2009WR008631>
- Fasching C, Ulseth AJ, Schelker J, Steniczka G, Battin TJ (2015) Hydrology controls dissolved organic matter export and composition in an alpine stream and its hyporheic zone. *Limnol Oceanogr* 61(2):558–571. <https://doi.org/10.1002/lno.10232>
- Ferreira VVM, Camargos CC, Chagas CJ, Aleixo BL, Ulhoa BMA (2011) Hydrological studies in the Juatuba basin. In: INAC-international nuclear Atlantic conference, Belo Horizonte, Brazil
- Ferreira VVM, Oliveira AL, Fonseca RLM, Oliveira NMG, Albuquerque RO, Auler LMLA, Carvalho Filho CA (2015) Assessment of Fluvial Sediments at Serra Azul Stream, Minas Gerais—Brazil. *J Geogr Environ Earth Sci Int* 2:92–109. <https://doi.org/10.9734/JGEESI/2015/16299>
- Fuller CC, Bargar JR (2014) Processes of zinc attenuation by biogenic manganese oxides forming in the hyporheic zone of Pinal Creek, Arizona. *Environ Sci Technol* 48(4):2165–2172. <https://doi.org/10.1021/es402576f>
- Gardner RP, Ely RL (1967) Radioisotope measurement applications in engineering. Van Nostrand Reinhold Inc., New York
- Harrington G, Gardner WP, Munday TJ (2014) Tracking groundwater discharge to a large river using tracers and geophysics. *Groundwater* 52(6):837–852. <https://doi.org/10.1111/gwat.1212>
- Jones JB, Stanley EH (2016) Stream ecosystems in a changing environment. Academic Press, Amsterdam
- Krause S, Boano F, Cuthbert MO, Fleckenstein JH, Lewandowski J (2014) Understanding process dynamics at aquifer–surface water interfaces: an introduction to the special section on new modeling approaches and novel experimental technologies. *Water Resour Res* 50:1847–1855. <https://doi.org/10.1002/2013WR014755>
- Krishan G, Rao MS, Kumar CP, Semwal P (2015) Radon concentration in groundwater of east coast of West Bengal, India. *J Radioanal Nucl Chem* 303(3):2221–2225. <https://doi.org/10.1007/s10967-014-3808-4>
- Le CH, Huynh NPT, Nguyen VT, Le QB (2015) Radon and radium concentrations in drinkable water supplies of the Thu Duc region in Ho Chi Minh City, Vietnam. *Appl Radiat Isot* 105:219–224. <https://doi.org/10.1016/j.apradiso.2015.08.033>
- Leibundgut C, Maloszewski P, Külls C (2009) Tracers in hydrology. Wiley-Blackwell, Chichester
- Lemke D, Liao Z, Wöhling T, Osenbrück K (2013) Concurrent conservative and reactive tracer tests in a stream undergoing hyporheic exchange. *Water Resour Res* 49(5):3024–3037. <https://doi.org/10.1002/wrcr.20277>
- Liu Y, Liu C, Nelson WC, Shi L, Xu F, Liu Y, Yan A, Zhong L, Thompson C, Fredrickson JK, Zachara JM (2017) Effect of water chemistry and hydrodynamics on nitrogen transformation activity and microbial community functional potential in hyporheic zone sediment columns. *Environ Sci Technol* 51(9):4877–4886. <https://doi.org/10.1021/acs.est.6b05018>
- Malzone JM, Anseeuw SK, Lowry CS, Allen-King R (2015) Temporal hyporheic zone response to water table fluctuations. *Groundwater* 54(2):274–285. <https://doi.org/10.1111/gwat.12352>
- Marques JM, Matos C, Carreira PM, Espinha Marques J, Teixeira J, Chaminé HI (2014) Assessment of mixing between shallow and thermal waters using geochemical and environmental isotope tracers (N Portugal): a review and reinterpretation. *Environ Earth Sci* 72(7):2557–2567. <https://doi.org/10.1007/s12665-014-3162-4>
- Mayer A, Nguyen BT, Banton O (2016) Using radon-222 to study coastal groundwater/surface–water interaction in the Crau coastal aquifer (southeastern France). *Hydrogeol J* 24(7):1775–1789. <https://doi.org/10.1007/s10040-016-1424-9>
- Mendoza-Lera C, Detry T (2017) Relating hydraulic conductivity and hyporheic zone biogeochemical processing to conserve and restore river ecosystem services. *Sci Total Environ* 579:1815–1821. <https://doi.org/10.1016/j.scitotenv.2016.11.166>
- Metcalfe E, Eddy HI (2003) Wastewater engineering: treatment and reuse. McGraw-Hill, New York
- Meus P, Moureaux P, Gailliez S, Flament J, Delloye F, Nix P (2014) In situ monitoring of karst springs in Wallonia (southern Belgium). *Environ Earth Sci* 71(2):533–541. <https://doi.org/10.1007/s12665-013-2760-x>
- Nevinsky I, Tsvetkova N, Nevinskaya E (2015) Measurement of radon in ground waters of the Western Caucasus for seismological application. *J Environ Radioact* 149:19–35. <https://doi.org/10.1016/j.jenvrad.2015.07.005>
- Peralta-Maraver I, Reiss J, Robertson AL (2018) Interplay of hydrology, community ecology and pollutant attenuation in the hyporheic zone. *Sci Total Environ* 610–611:267–275. <https://doi.org/10.1016/j.scitotenv.2017.08.036>
- Pinto EJA, Lima JES, Davis EG, Silva AJ, Dantas CEO, Candido MO, Palmier LR, Monte-Mor RCA (2010) Estimation of the natural recharge of the free aquifer of a sub basin of the representative Juatuba basin (MG) applying the method of variation of water levels [in Portuguese]. In: XVI Brazilian congress of underground waters, São Luís, MA, Brazil
- Ravikumar P, Davis D, Matthew S, Somashekar RK, Prakash KL (2014) Spatio-temporal variation in radon concentration in groundwater with respect to rock types: a case study from Chitradurga district, Karnataka. *J Geol Soc India* 83(2):156–164. <https://doi.org/10.1007/s12594-014-0027-0>
- Saad MH, Tamboul J, Yousef M (2014) Evaluation of natural radioactivity in different regions in Sudan. *J Am Sci* 10(2):14–18
- Savoy L, Surbeck H, Hunkeler D (2011) Radon and CO₂ as natural tracers to investigate the recharge dynamics of karst aquifers. *Hydrol J* 406(3–4):148–157. <https://doi.org/10.1016/j.jhydrol.2011.05.031>
- Sawyer AH, Kaplan LA, Lazareva O, Michael HA (2014) Hydrologic dynamics and geochemical responses within a floodplain aquifer and hyporheic zone during Hurricane Sandy. *Water Resour Res* 50:4877–4892. <https://doi.org/10.1002/2013WR015101>
- Schaper JL, Seher W, Nützmann G, Putschew A, Jekel M, Lewandowski J (2018) The fate of polar trace organic compounds in the hyporheic zone. *Water Res* 140:158–166. <https://doi.org/10.1016/j.watres.2018.04.040>
- Schnegg PA, Perret C, Hauet A, Parrel D, Saysset G, Vignon P (2011) Stream gauging by dilution of fluorescent tracers and state of the art of the EDF hydroclimatological observation network. In: 9th conference on limestone hydrogeology, Besançon, France, pp 435–438
- Schneider P, Pool S, Strouhal L, Seibert J (2014) True colors—experimental identification of hydrological processes at a hillslope prone to slide. *Hydrol Earth Syst Sci* 18:875–892. <https://doi.org/10.5194/hess-18-875-2014>
- Secretariat of Health of Minas Gerais State (2009) Project Poços de Caldas Plateau—Research: cancer and natural radiation: Minas Gerais-Brasil—2004 to 2009 [in Portuguese]. SES/MG, Belo Horizonte

- Simmons GC (1968) Geology and iron deposits of the western Serra do Curral, Minas Gerais, Brazil. Geological survey professional paper, Washington 341-G
- Sohrbai J (2013) World high background natural radiation areas: need to protect public from radiation exposure. *Radiat Meas* 50:166–171. <https://doi.org/10.1016/j.radmeas.2012.03.011>
- Somashekar RK, Ravikumar P (2010) Radon concentration in groundwater of Varahi and Markandeya river basins, Karnataka State, India. *J Radioanal Nucl Chem* 285(2):343–351. <https://doi.org/10.1007/s10967-010-0573-x>
- Sophocleous M (2002) Interactions between groundwater and surface water: the state of science. *Hydrogeol J* 10(1):52–67. <https://doi.org/10.1007/s10040-001-0170-8>
- Tallini M, Parisse B, Petitta M, Spizzico M (2013) Long-term spatio-temporal hydrochemical and ^{222}Rn tracing to investigate groundwater flow and water–rock interaction in the Gran Sasso (central Italy) carbonate aquifer. *Hydrogeol J* 21:1447–1467. <https://doi.org/10.1007/s10040-013-1023-y>
- Trauth N, Fleckestein JH (2016) Single discharge events increase reactive efficiency of the hyporheic zone. *Water Resour Res* 53:779–798. <https://doi.org/10.1002/2016WR019488>
- UK Environment Agency (2009) The hyporheic handbook: a handbook on the groundwater–surface water interface and hyporheic zone for environment managers. UK Environmental Agency, Bristol
- UNSCEAR-United Nations Scientific Committee on the Effects of Atomic Radiation (2000) Sources and effects of ionizing radiation, 1st edn. United Nations Publication, New York
- Ward PRB (1973) Prediction of mixing lengths for river flow gaging. *J Hydraul Div* 99(7):1069–1081
- Wondzell SM (2006) Effect of morphology and discharge on hyporheic exchange flows in two small streams in the Cascade Mountains of Oregon, USA. *Hydrol Process* 20(2):267–287. <https://doi.org/10.1002/hyp.5902>
- Zimmer MA, Lautz LK (2013) Temporal and spatial response of hyporheic zone geochemistry to a storm event. *Hydrol Process* 28:2324–2337. <https://doi.org/10.1002/hyp.9778>