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^{131}I and ^{99m}Tc in Effluents from a Nuclear Medicine Facility and Associated Sewage Treatment Unit

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Abstract The release of liquid effluents containing radionuclides from radioactive plants in urban areas has been the subject of discussions related to the environmental issue. This study presents the analysis of the activity concentrations (AC) of ^{131}I and ^{99m}Tc in sewage samples collected in a nuclear medicine facility (NMF), associated with an university hospital situated in Rio de Janeiro, Brazil. The analyzes and measurements by gamma spectrometry of the collected sewage samples were performed, whose the estimated values (EV) for AC obtained for ^{131}I in July (8.78×10^3 Bq m^{-3}) and September 2016 (9.80×10^3 Bq m^{-3}) and in February 2017 (1.14×10^4 Bq m^{-3}) were lower than the exemption level (EL = 1.90×10^4 Bq m^{-3}). For ^{99m}Tc, the EV for AC obtained from August (4.77×10^2 Bq m^{-3} and 2.96×10^4 Bq m^{-3}) to September 2017 (2.18×10^3 Bq m^{-3}) were lower than the EL (1.90×10^9 Bq m^{-3}). From an environmental point of view, the presence of ^{131}I in the sewage treatment unit (STU), even if below the legal release limit, points out the greater importance of this radionuclide, as it requires special precautions in its adequate storage for the radioactive decay in the NMF and the dilution effect to reduce its AC minimizing the environmental impact after the effluent release to the environment. The presence of ^{131}I in the STU was not expected pointing to the need and evaluation of the waste management system (WMS) of the NMF.

Keywords Radioactivity liquid wastes · Waste management · Environmental radiological protection · Occupational radiological protection

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1 Introduction

The interest in protecting the environment has been growing in recent years in relation to all aspects of human activity. Such interest is accompanied by the development and application of various means of

management and evaluation in terms of human impact on the environment (ICRP 2007). Growing awareness of potential impacts means that society has a better understanding of the approach related to the risks of radioactive exposure of biota (ARPANSA 2015; ICRP 2009; ICRP 2017).

Among these sources, medical exposures comprise the second largest contribution to radiation exposure in man with 20 % of the total, just being ranked after the background radiations (UNEP 2016).

Nuclear medicine is the medical modality in which radioactive isotopes are used in the form of unsealed sources, both for diagnostic and therapeutic purposes. Among radiopharmaceuticals, ^{99m}Tc (half-life of 6 hours and effective energy of 140 keV) is used in more than 80 % of total scintigraphy examinations. Its administration varies according to the type of examination and the patient's weight. For example, for renal scintigraphy exams, the activity varies between 37 and 555 MBq, and, for cerebral scintigraphy exams, the activity using ^{99m}Tc-ECD or ^{99m}Tc-HPMAO for adult patients with 70 kg of mass varies between 740 and 1110 MBq (Soares 2019). ^{131}I (half-life of 8 days and effective energy of 364.5 keV for gamma rays (81.5 % of total energy emission) and 606.3 keV for beta particles (89.9 % of total energy emission)) is applied both for diagnostic, whose dosage is up to 400 MBq, and for the treatment of patients with hyperthyroidism and differentiated carcinomas of the thyroid gland, whose activity is up to 10 GBq, with ablation in order of 7 GBq (Carolan et al. 2011).

The elimination of excreta from patients undergoing scintigraphy exams or with therapeutic doses of radiopharmaceuticals should possess sewage collection systems (CNEN 2013; Fischer et al. 2009; CNEN 2014; SSK 2004; Krawczyk et al. 2013). These wastes generated from nuclear medicine facility (NMF) can be dealt with by simply storing the wastes safety until the reduction of the activity concentration (AC) from the radioactive decay to a safe level and releasing the effluent with low AC into the sewage system, with the assumption that its radioactive level be diluted by many orders of magnitude within the primary sewage flow, which results to avoid significant exposure for the public in general (Jiménez et al. 2011), under permission from the regulatory authority and appropriate monitoring (Ravichandran et al. 2011). Some sewage treatment units (STU) receive these effluent discharges (Rose et al. 2012), where they are treated with several physical, chemical, and biological processes, being subsequently discharged to the environment (Jiménez et al. 2011). Release of effluents containing radionuclides to the aquatic environment provides a potential route by which the public can be exposed to radionuclides (Carolan et al. 2011).

The generation of liquid wastes containing radionuclides from nuclear medicine techniques plays a significant role due to the impact they cause on the environment (Mundschenk 1996; Barquero et al. 2008). Since its increasing medical employment in the beginning of the 1970s (Moss 1973; Birks 1975; Lee et al. 2018), ^{131}I has been caused greater concern than ^{99m}Tc due to the characteristics of both (half-life and effective energy) (Sundell-Bergman et al. 2008) so that the precautions for this radionuclide in the environment are essentials to protect human health (Lee et al. 2018). Regarding medical exposure, the concentrations of ^{131}I, ^{223}Ra, ^{177}Lu, and ^{90}Y, among others, generate exposures that require radioprotection for the medical staff and for the patients and their relatives (ICRP 2004). By the same characteristics, the occupational impact of ^{99m}Tc is lower than that ^{131}I.

Based on the above, this study intends to evaluate the activity concentration (AC) of ^{131}I and ^{99m}Tc from the collection and analysis of the sewage samples from a NMF in operation and sewage treatment unit to evaluate if the concentration levels of these radionuclides are in accordance with current recommendations.

2 Methods

2.1 Collection of Sewage Samples in a Nuclear Medicine Facility

The collection of sewage samples was performed in a NMF located in the city of Rio de Janeiro, which has the following divisions:

- A therapeutic room with bathroom for hospitalization of the patient undergoing thyroid treatment with ^{131}I (licensed activity allowed up to $1.85 \cdot 10^{10}$ Bq per week), with direct connection to an external septic tank, which received excreta from the patient containing this radionuclide. This tank was connected to another external septic tank to transfer part of the stored sewage to dilute ^{131}I.

– A waiting room of patients for average weekly attendance of 18 patients for scintigraphy examinations with ^{99m}Tc (licensed activity allowed up to $1.48 \cdot 10^{11}$ Bq per week). This room has two bathrooms, one for each sex, with direct connection to a septic tank located outside.

The samples were collected in three stages: The first stage corresponded to the collection of samples containing ^{131}I before and after hospitalization between July and September 2016, aiming to test the methodology; the second stage corresponded to the collection of samples containing ^{131}I on the date of the patient's hospitalization in February 2017; and the third stage corresponded to the collection of samples containing ^{99m}Tc between August and September 2017. The sewage samples containing ^{131}I were stored in 500-mL polypropylene pots, while the sewage samples containing ^{99m}Tc were stored in 200-mL pots. These samples were sent to the laboratory for measurement and analysis, which allowed the measurement of different ACs for these radionuclides. These ACs, associated with the logistics of collecting sewage samples and the initial volume of septic tanks, allowed to manufacture the pots with different volumes for the storage of the collected samples, aiming at the sensitivity of the radiation detectors and the optimization of the samples' disposal with reduced volume generated.

2.2 Collection of Sewage Samples in a Sewage Treatment Unit

Samples were collected at the sewage treatment unit (STU) between October and November 2016. This station is located in the north zone of Rio de Janeiro and receives the sewage from some medical facilities and other non-radioactive facilities. The samples were collected in a tank that received the sewage from the primary decanter, where sedimentable suspended solids and floating solids were removed by physicochemical processes of coagulation, flocculation, and sedimentation. The storage of the collected sewage samples was also made using 500-mL polypropylene pots for measurement and analysis in the laboratory. Because it is an experimental station, the STU works by batch intermittently, that is, the mode of operation in which the system is operated discontinuously, in a transient or non-stationary state, thus not allowing to estimate parameters such as volume of deposits and dwell time.

2.3 Methodology for Measurement of Collected Sewage Samples

The samples were measured and analyzed by gamma spectrometry at the Environmental Analysis and Computational Simulation Laboratory (LAASC/PEN/COPPE/UFRJ).

For the analysis of the samples collected at the nuclear medicine facility, a 3″ × 3″ scintillator detector [NaI(Tl), Ortec, model 905-4] was used, with efficiency of 2 % for a calibrated source of ^{137}Cs of 1 μCi ($3.70 \cdot 10^4$ Bq). Software MAESTRO-32 MCA (Ortec) was used for energy calibration of NaI(Tl) and measurement of the radionuclides. The energy calibration process of NaI(Tl) was based on the use of properly calibrated and certified sealed sources in order to establish an energetic band, which corresponds the quantity of atoms that form a large quantity of energy levels close to each other, for detecting the radionuclides present in the samples. To measure the samples containing ^{131}I, sources of ^{137}Cs and ^{133}Ba were used, with a detector sensitivity of $2.06 \cdot 10^8$ Bq m^{-3}. For the samples with ^{99m}Tc, sources of ^{241}Am and ^{133}Ba, and a mixed source of ^{155}Eu/^{22}Na, were used, with a detector sensitivity of $5.42 \cdot 10^9$ Bq m^{-3}. The detector sensitivity is the ability to produce a usable signal for a given type of radiation and energy. The use of different sources for detecting radionuclides in the samples is due to the energy emission of gamma radiation within the energy band. The same methodology was used for a semiconductor detector (HPGe) employed to measure the samples collected at the STU, which its description is located below. The counting times used for the analysis of samples with ^{131}I and ^{99m}Tc were, respectively, 900 s and 1800 s. The time required for counting the samples was defined according to the laboratory's routine and the uncertainty of the activity of each radionuclide in order to obtain a reliable margin.

For the samples collected at the STU, a coaxial semiconductor detector (HPGe, Canberra Industries, model GC3020) was used, coupled to a low-noise preamplifier (Canberra Industries, model 2002 CSL), and software Genie 2000 (Canberra Industries) with multichannel system (8192 channels). With the aid of software Gamma Analysis from Genie 2000, the energy calibration of the HPGe detector was performed using sealed sources of ^{137}Cs, ^{60}Co, and a mixed source of ^{155}Eu/^{22}Na, in order to establish the linear association between channel and energy for identification of the

radionuclides present in the samples, with detector sensitivity of $6.58 \cdot 10^6$ Bq·m^{-3}, with the counting time of 28,800 s.

2.4 Acquisition of Activity Concentration Values for ^{131}I and ^{99m}Tc in the Collected Samples

Based on the data obtained by the detector systems, the AC values of ^{131}I and ^{99m}Tc in the sewage were calculated according to Eq. 1, being corrected in relation to the dates and times of the sample collections:

$$AC = \frac{N}{(P_\gamma * t * v * \varepsilon)} \quad (1)$$

where **N** is the net area under the efficiency curve, **ε** is the detection efficiency obtained by the Monte Carlo simulation (Code MCNPX), **Pγ** is the probability of gamma ray emission as a function of energy, **v** is the volume of collected sample given in m^3, and **t** is the sample measurement time given in seconds.

The error associated with Eq. 1 can be estimated using the propagation of errors, as shown in Eq. 2:

$$\delta AC = \left\{ \frac{\delta N^2}{(v*\varepsilon*t*P_\gamma)^2} + N^2 \left[\frac{\delta \varepsilon^2}{(v*\varepsilon^2*t*P_\gamma)^2} + \frac{\delta v^2}{(v^2*\varepsilon*t*P_\gamma)^2} + \frac{\delta t^2}{(v*\varepsilon*t^2*P_\gamma)^2} \right] \right\}^{\frac{1}{2}} \quad (2)$$

where **δAC** is the uncertainty of the activity concentration, **δN** is the uncertainty of the net area under the efficiency curve, **δε** is the uncertainty of detection efficiency, **δv** is the uncertainty of the sample volume, **δt** is the uncertainty of the acquisition time of the gamma spectrum, and **δP$_\gamma$** is the uncertainty of the probability of gamma rays emission, but this probability was ignored, because the probability of decay was considered absolute.

2.5 Minimum Detection Limit

The minimum detectable activity (MDA), for a given radionuclide, at a 95 % confidence level, is based on the Currie derivation (L'Annunziata 2012) and is in accordance with the standards established by ISO Standards 10703 (2007) and 11929 (2010). The expression used for MDA is given by Eq. 3:

$$MDA = \frac{(2.71 + 4.66*\sigma)}{(P_\gamma * t * v * \varepsilon)} \quad (3)$$

where **σ** is the standard deviation of background radiation added to the sample holder, while the other parameters are the same as those referenced in Eq. 1.

2.6 Methodology for Radionuclide Dilution

In order for the sewage containing radionuclide in the septic tanks to be released as a liquid effluent, NMFs must comply with current normative recommendations (CNEN 2014). In order to estimate the activity concentrations of radionuclides in the effluents as shown in Eq. 4, a contour situation was delineated as follows:

1) The volume of the septic tank who receive part of the sewage for dilution $\mathbf{V_{st}}$ from the batch process given in m^3.
2) Consider that the release occurs in three batches in the month and each one with one-third of the volume of the septic tank and every 10 days:
   a. The first batch with the lowest AC value ($\mathbf{AC_{low}}$) given in Bq m^{-3}
   b. The second batch with the highest AC value ($\mathbf{AC_{high}}$) given in Bq m^{-3}
   c. The third batch with the intermediate value of AC ($\mathbf{AC_{int}}$) given in Bq m^{-3}
3) Consider the monthly volume of sanitary sewage generated in the nuclear medicine facility $\mathbf{V_m}$ given in m^3.
4) Consider the half-life $\mathbf{T_{1/2}}$ for radiological decay.

$$EV = \frac{\left\{ \left[\left(\frac{V_{st}*AC_{low}}{3} \right) + \left(\frac{V_{st}*AC_{high}}{3} \right) + \left(\frac{V_{st}*AC_{int}}{3} \right) \right] *e^{-\lambda*\Delta T} \right\}}{V_m} \quad (4)$$

where **EV** is the estimated value of activity concentration after dilution given in Bq m^{-3}, **λ** is the decay constant of radionuclide, and **ΔT** is the interval time for the radioactive decay.

2.7 Radioprotection Management of Collection, Transport, and Sample Analysis

The analyzed material was considered a radioactive material. The radioprotection protocol was based on the set of activities (sampling, transport, and analysis)

as a radioprotection operation (RO). Only the external exposure was considered, whose radioprotection procedures were elaborated aiming at protecting the team and not allowing the dispersion of the material collected. Individual protection equipments (IPEs) and careful handling of samples were used to avoid contamination. The technical team (TT) and the region surrounding the collection area were monitored to certify the absence of post-operation contamination. The time and the distance estimates were established in order to perform the same procedures with inert (non-radioactive) material. Thereby, according to Eq. 5, the external effective dose $\mathbf{E_{ext}}$ received by the technical team was estimated, based on Martínez et al. (2018):

$$E_{ext} = \frac{\left(\Gamma_i * A_{max(i)} * t\right)}{d^2} \quad (5)$$

where $\mathbf{\Gamma_i}$ is the specific constant for radionuclide **i** so that it was used the value of 52.20 μGy m^2 GBq^{-1} h^{-1} for ^{131}I and 14.10 μGy m^2 GBq^{-1} h^{-1} for ^{99m}Tc (Ninkovic and Androvic 2005), $\mathbf{A_{max(i)}}$ is the maximum activity found given in Becquerel (Bq), **t** is the conservative time of exposure given in minutes, and **d** is the distance between the sample and the members of TT given in meters.

The estimated external effective dose with the maximum value of radionuclide activity was used to estimate the maximum individual effective dose of the operation $\mathbf{E_{max}}$ (see Eq. 6), multiplied by the number of samples $\mathbf{N_a}$. Aiming the optimization process, the maximum individual effective dose of the operation was multiplied by the number of team members $\mathbf{N_e}$ to estimate the collective effective dose of the operation $\mathbf{E_{col}}$ (see Eq. 7).

$$E_{max} = E_{ext} * N_a \quad (6)$$

$$E_{col} = E_{max} * N_e \quad (7)$$

2.8 Radiological Management of Analyzed Samples

After the measurements, the samples were segregated and stored for a quarantine period to fit the exemption level of $1.00 \cdot 10^6$ Bq for ^{131}I and $1.00 \cdot 10^7$ Bq for ^{99m}Tc (CNEN 2011). The exemption level applies, among other criteria, the amount of radioactive material less than one ton, if it fits for the samples collected.

3 Results and Discussions

3.1 Results of Sewage Samples Collected from a Hospital Facility

Forty-one sewage samples were collected at the NMF, 16 of which contained ^{131}I and 25 contained ^{99m}Tc. In these conditions, the sewage was not considered to be effluent.

In order to release the sewage as effluent, it was necessary to dilute the AC of ^{131}I according to Eq. 4 generating, thus, the estimated values (EV). Therefore, the AC values of samples collected from the septic tank connected to the therapeutic room between July 26 and 28, 2016, were used as inputs because they were continuous, as well as the samples collected between September 27 and 29, 2016. In July 2016, the lowest and the highest AC values obtained for the first and second batches ($\mathbf{AC_{low}}$ and $\mathbf{AC_{high}}$) were, respectively, $1.07 \cdot 10^8$ Bq m^{-3} and $8.96 \cdot 10^8$ Bq m^{-3}, with an intermediate value for the third batch ($\mathbf{AC_{int}}$) of $5.02 \cdot 10^8$ Bq m^{-3}. In September 2016, the lowest and the highest AC values obtained for the first and second batches ($\mathbf{AC_{low}}$ and $\mathbf{AC_{high}}$) were, respectively, $1.31 \cdot 10^3$ Bq m^{-3} and $1.12 \cdot 10^9$ Bq m^{-3}, with an intermediate value for the third batch ($\mathbf{AC_{int}}$) of $5.60 \cdot 10^8$ Bq m^{-3}. The reason for the different AC values for the samples collected in September 2016 was that, before the admission of the patient to the therapeutic room, there was low AC value for ^{131}I in the septic tank. After the start of the treatment, this radionuclide was admitted to the septic tank along with excreta released by the patient, thus increasing its concentration. The period ΔT for the decay of ^{131}I was considered as 8 days (1 half-life for this radionuclide). The volume of each batch of the septic tank ($\mathbf{V_{st}}$) was considered 0.70 m^3 (approximately 1/3 of total volume of 2.00 m^3) and $6.00 \cdot 10^4$ m^3 as the monthly volume of sewage generated at the NMF ($\mathbf{V_m}$) inputs for dilution, generating an EV of $8.78 \cdot 10^3$ Bq m^{-3} for July and $9.80 \cdot 10^3$ Bq m^{-3} for September. Table 1 presents the AC values obtained for the samples collected in the above periods.

In an analogous way, Table 2 presents the AC values of ^{131}I referring to the samples collected in February 2017. The data were obtained before, during, and after the patient's hospitalization, as indicated in the mentioned table. The lowest and the highest AC values of ^{131}I for the first and the second batches were, respectively, $9.18 \cdot 10^5$ Bq m^{-3} and $1.30 \cdot 10^9$ Bq m^{-3}, with an

intermediate value for the third batch of $6.50 \cdot 10^8$ Bq m^{-3}, generating, thus, an EV of $1.14 \cdot 10^4$ Bq m^{-3}. The reason for the different AC values for the samples collected was that:

1) Before the admission of the patient to the therapeutic room on February 15, there were high AC values for ^{131}I in the septic tank because another patient was hospitalized in the week prior to sample collection. After the admission, the AC values for ^{131}I started to rise more because of the excreta released by the patient.
2) After the release of the patient from the therapeutic room on February 16, there were recorded the stability levels the AC for ^{131}I in the septic tank until its decay began.

Analyzing the situations presented, it was observed that the estimated values of AC after dilution (EV) for ^{131}I were below the exemption level (EL). Other studies show the effective control of the AC of ^{131}I present in the septic tanks from the medical facilities: For Goddard (1999), the average AC values measured for ^{131}I present in the septic tanks of a university hospital in Al-Khod (Oman) ranged from $(4.18 \pm 0.01) \times 10^6$ Bq m^{-3} to $(6.59 \pm 0.01) \times 10^6$ Bq m^{-3}; for Krawczyk et al. (2013), the average values obtained for ^{131}I present in the effluents released in two medical facilities in Granada (Spain) ranged from $(1.21 \pm 0.58) \times 10^2$ Bq m^{-3} to $(5.93 \pm 0.27) \times 10^4$ Bq m^{-3}. The results presented above indicate that the waste containing ^{131}I of radiopharmaceutical therapy requires special precautions from storage suitable for radioactive decay and the effect of the dilution to reduce the activity concentration of this radionuclide in order to minimize the environmental impact after the effluent release (IAEA 2018a). The medical facilities should specify the ideal option related to the release of sewage, taking into consideration the economic, social, environmental, and safety characteristics, among others (IAEA 2018b). In addition, it shall determine the characteristics and activities at the sewage release points, the routes of exposure, and the radiological impacts on the environment in full with the characteristics of the radioprotection system, in accordance with the requirement of the national regulatory authority (IAEA 2014; IAEA 2018c). For the installation to be authorized, there is analysis in the WMS by the regulatory body where the facility demonstrates the efficiency of the dilution of all radioactive material consumed.

Table 3 presents the AC values of ^{99m}Tc obtained for the samples collected between August and September 2017. The activities were administrated according to the clinical situation of the patient and the type of scintigraphy examination. For infant patients, the activity varied from $3.70 \cdot 10^7$ Bq to $1.85 \cdot 10^8$ Bq and for adults from $7.40 \cdot 10^8$ Bq to $1.11 \cdot 10^9$ Bq. The results of the AC values with ^{99m}Tc were different from the AC values obtained for ^{131}I because, after excreted by injected patients and incorporated into conventional sewage, ^{99m}Tc acquires radiological characteristics to be released, according to radiological licensing.

For the dilution of the AC of ^{99m}Tc contained in the sewage before its release as effluent, it was necessary to input the AC values for the samples collected on August 8 and 23 and September 12 generating, thus, the estimated values (EV) of dilution, according to Eq. 4. For the EV on August 8, the lowest and the highest AC values obtained were, respectively, $2.62 \cdot 10^8$ Bq m^{-3} and $2.79 \cdot 10^9$ Bq m^{-3}, with an intermediate value of $1.53 \cdot 10^9$ Bq m^{-3}. For the EV on August 23, the lowest and the highest AC values obtained were, respectively, $1.24 \cdot 10^9$ Bq m^{-3} and $1.88 \cdot 10^{11}$ Bq m^{-3}, with an intermediate value of $9.46 \cdot 10^{10}$ Bq m^{-3}. For the EV on September 12, the lowest and the highest AC values were, respectively, $1.11 \cdot 10^9$ Bq m^{-3} and $1.28 \cdot 10^{10}$ Bq m^{-3}, with an intermediate value of $6.96 \cdot 10^9$ Bq m^{-3}. Considering the period ΔT as 24 hours (4 times the half-life of ^{99m}Tc), the volume of each batch of the septic tank as 0.10 m^3, and the monthly volume of the nuclear medicine facility inputs for dilution as $6.00 \cdot 10^4$ m^3, the EV generated for August 8 was $4.78 \cdot 10^2$ Bq m^{-3}, 2.96 10^4 Bq m^{-3} for August 23, and $2.18 \cdot 10^3$ Bq m^{-3} for September 12. The estimated values for the activity concentration after dilution for ^{99m}Tc were lower than the exemption level. In relation to the study made by Krawczyk et al. (2013), the average AC values for ^{99m}Tc present in the effluents released in three medical facilities in Granada (Spain) ranged from $(1.49 \pm 0.10) \times 10^5$ Bq m^{-3} to $(2.51 \pm 0.16) \times 10^6$ Bq m^{-3}. In relation with ^{131}I, the results obtained for ^{99m}Tc indicate a faster reduction in AC and a lower environmental impact.

Table 1 Activity concentration values of ^{131}I in the sewage samples collected in July and September 2016

Samples	HD	Activity (Bq)	PH	CD	AC (Bq m^{-3})	EV (Bq m^{-3})	EL (Bq m^{-3})
S01	July 27	$6.29 \cdot 10^{9}$	Before	July 26	$(1.07 \pm 0.01) \cdot 10^{8}$	$8.78 \cdot 10^{3}$	$1.90 \cdot 10^{4}$
S02			After	July 28	$(8.96 \pm 0.02) \cdot 10^{8}$		
S03	Sept 28	$5.71 \cdot 10^{9}$	Before	Sept 27	$(1.31 \pm 0.00) \cdot 10^{3}$	$9.80 \cdot 10^{3}$	
S04			After	Sept 29	$(1.12 \pm 0.01) \cdot 10^{9}$		

HD hospitalization day, *PH* patient hospitalization, *CD* collection day, *AC* activity concentration, *EV* estimated value of activity concentration after dilution, *EL* exemption level (CNEN 2014)

3.2 Results of Sewage Samples Collected at the Sewage Treatment Unit

The HPGe gamma spectrometric measurement system detected the presence of ^{131}I energy peaks above the MDA in all 5 samples collected at STU between October and November 2016. Table 4 presents the data referring the AC values of ^{131}I, with sample S42 having the highest AC value, being in the same order of magnitude of the EL.

Due to the regulation of radioactive waste current in Brazil (CNEN 2014), AC values of ^{131}I in the EL magnitude were not expected, pointing out the need to reevaluate the WMS of the NMF evaluated in this study. Other studies have pointed out the presence of this radionuclide in the STU: Carolan et al. (2011) evaluated the average AC value for ^{131}I for the samples collected at a STU near the city of Sydney (Australia) in $(1.60 \pm 1.50) \times 10^{3}$ Bq m^{-3}; Lee et al. (2018) detected the presence of ^{131}I in water samples collected at a STU in Daejeon (South Korea), near the Korean Atomic Energy Research Institute (KAERI), whose results of the AC for ^{131}I ranged from 0.34×10^{3} Bq m^{-3} to 3.21×10^{3} Bq m^{-3}. However, in this study, ^{99m}Tc was not detected in the samples analyzed by the system due to the dilution of this radionuclide in the sewage system. For ^{99m}Tc, the WMS is in compliance.

3.3 Effective doses associated with collection and analysis

For the samples collected at the hospital facility, the external effective dose $\mathbf{E_{ext}}$ for ^{131}I was $1.22 \cdot 10^{-4}$ mSv, while the external effective dose for ^{99m}Tc was $1.44 \cdot 10^{-3}$ mSv. Conservatively, the number of samples $\mathbf{N_a}$ was equal to 20 for ^{131}I and 30 for ^{99m}Tc in Eq. 6 generating, respectively, the maximum effective doses $\mathbf{E_{max}}$ of $2.45 \cdot 10^{-3}$ mSv and $4.32 \cdot 10^{-2}$ mSv and that, by aggregation, the maximum individual effective dose in TT was $4.56 \cdot 10^{-2}$ mSv. The number of members $\mathbf{N_e}$ of the TT

Table 2 Activity concentration values of ^{131}I in the sewage samples collected in February 2017

Samples	HD	Activity (Bq)	PH	CD	AC (Bq m^{-3})	EV (Bq m^{-3})	EL (Bq m^{-3})
S05	Feb 15	$3.90 \cdot 10^{9}$	Before	Feb 15	$(9.18 \pm 0.00) \cdot 10^{5}$	$1.14 \cdot 10^{4}$	$1.90 \cdot 10^{4}$
S06			Before		$(9.17 \pm 0.00) \cdot 10^{5}$		
S07			During		$(7.74 \pm 0.00) \cdot 10^{6}$		
S08			During		$(5.39 \pm 0.00) \cdot 10^{8}$		
S09			During		$(5.79 \pm 0.00) \cdot 10^{8}$		
S10			During		$(1.30 \pm 0.00) \cdot 10^{9}$		
S11			After	Feb 16	$(1.00 \pm 0.00) \cdot 10^{9}$		
S12			After		$(1.02 \pm 0.00) \cdot 10^{9}$		
S13			After		$(1.00 \pm 0.00) \cdot 10^{9}$		
S14			After		$(1.04 \pm 0.00) \cdot 10^{9}$		
S15			After	Feb 17	$(7.26 \pm 0.00) \cdot 10^{8}$		
S16			After	Feb 20	$(3.85 \pm 0.00) \cdot 10^{8}$		

HD hospitalization day, *PH* patient hospitalization, *CD* collection day, *AC* activity concentration, *EV* estimated value of activity concentration after dilution, *EL* exemption level (CNEN 2014)

Table 3 Activity concentration values of ^{99m}Tc in the sewage samples collected between August and September 2017

Samples	CD	Collection time (h)	AC (Bq m^{-3})	EV (Bq m^{-3})	EL (Bq m^{-3})
S17	Aug 08	10:08 a.m.	$(1.43 \pm 0.00)\cdot10^{9}$	$4.77\cdot10^{2}$	$1.90\cdot10^{9}$
S18		10:30 a.m.	$(2.62 \pm 0.01)\cdot10^{8}$		
S19		10:50 a.m.	$(1.26 \pm 0.00)\cdot10^{9}$		
S20		02:29 p.m.	$(2.79 \pm 0.00)\cdot10^{9}$		
S21		02:44 p.m.	$(8.28 \pm 0.02)\cdot10^{8}$		
S22		02:57 p.m.	$(3.47 \pm 0.01)\cdot10^{8}$		
S23	Aug 23	09:34 a.m.	$(6.59 \pm 0.02)\cdot10^{9}$	$2.96\cdot10^{4}$	
S24		09:44 a.m.	$(1.48 \pm 0.02)\cdot10^{9}$		
S25		09:52 a.m.	$(1.24 \pm 0.02)\cdot10^{9}$		
S26		10:00 a.m.	$(1.88 \pm 0.00)\cdot10^{11}$		
S27		10:04 a.m.	$(2.16 \pm 0.00)\cdot10^{10}$		
S28		10:11 a.m.	$(5.50 \pm 0.00)\cdot10^{10}$		
S29		10:16 a.m.	$(2.95 \pm 0.00)\cdot10^{10}$		
S30		10:23 a.m.	$(4.00 \pm 0.00)\cdot10^{10}$		
S31	Sept 12	09:31 a.m.	$(1.67 \pm 0.00)\cdot10^{9}$	$2.18\cdot10^{3}$	
S32		09:35 a.m.	$(1.11 \pm 0.00)\cdot10^{9}$		
S33		09:42 a.m.	$(1.72 \pm 0.00)\cdot10^{9}$		
S34		09:49 a.m.	$(1.17 \pm 0.00)\cdot10^{10}$		
S35		09:54 a.m.	$(6.12 \pm 0.00)\cdot10^{9}$		
S36		10:00 a.m.	$(3.89 \pm 0.00)\cdot10^{9}$		
S37		10:06 a.m.	$(1.28 \pm 0.00)\cdot10^{10}$		
S38		10:12 a.m.	$(5.25 \pm 0.00)\cdot10^{9}$		
S39		10:18 a.m.	$(4.34 \pm 0.00)\cdot10^{9}$		
S40		10:24 a.m.	$(2.19 \pm 0.00)\cdot10^{9}$		
S41		10:30 a.m.	$(1.60 \pm 0.00)\cdot10^{9}$		

CD collection day, *AC* activity concentration, *EV* estimated value of activity concentration after dilution, *EL* exemption level (CNEN 2014)

was composed by 3 members generating, in Eq. 7, the collective effective dose $\mathbf{E_{col}}$ of $7.34\cdot10^{-3}$ mSv for ^{131}I and $1.30\cdot10^{-1}$ mSv for ^{99m}Tc. The maximum external dose in the RO was 0.14 mSv in a time interval of, approximately, 17 hours. Pointing to a total collective effective dose of the RO that is compatible with the effective dose rate of the public (0.30 mSv year^{-1}), there is no need to perform a radiation protection optimization process as recommended (ICRP 2007; IAEA 2014). Table 5 summarizes these results.

3.4 Release of Samples from the Laboratory

The time to reach the unconditional exemption level of the samples, by exponential decay, stored after their measurement from LAASC/PEN/COPEE/UFRJ containing ^{131}I, was 30 days; for the samples containing ^{99m}Tc, the exemption time was 5 days, thus meeting the current recommendations (CNEN 2011). After this period of quarantine, the samples were counting and confirmed the AC below the exception limit; these samples were released into the sewage system.

Table 4 Measurement data of the activity concentration of ^{131}I for the samples at the STU between October and November 2016

Samples	Collection day	AC (Bq m^{-3})	EV	EL (Bq m^{-3})
S42	Oct 31	$(1.12 \pm 0.06)\cdot10^{4}$	NA	$1.90\cdot10^{4}$
S43	Nov 01	$(9.43 \pm 0.51)\cdot10^{3}$		
S44	Nov 03	$(8.19 \pm 0.45)\cdot10^{3}$		
S45	Nov 07	$(6.29 \pm 0.34)\cdot10^{3}$		
S46	Nov 09	$(5.52 \pm 0.30)\cdot10^{3}$		

AC activity concentration, *EV* estimated value, *NA* not applicable because it is not a radioactive facility, *EL* exemption level (CNEN 2014)

4 Conclusions

At a maximum effective dose of $4.56 \cdot 10^{-2}$ mSv and a total collective effective dose of 0.14 mSv, the dose is compatible with the individual of the public, and, to this magnitude, the RO optimization is not justified (see Table 5 (optimization if collective dose > 0.30 mSv y^{-1})).

The samples analyzed in this study indicated different situations for ^{131}I and ^{99m}Tc. With high AC values, associated with a longer half-life and higher effective energy, ^{131}I represents a greater environmental concern when compared to ^{99m}Tc. The ^{131}I values found in the STU point to a revaluation of the radioactive WMS of the NMF.

As a prospect of improvement, one possible technical solution for this situation is to employ an active control of the release of the effluents, i.e., only to perform the effluent release after confirmation based on gamma spectrometry that they are below the limits, such as that performed at LAASC/PEN/COPPE/UFRJ to release samples. The release, based on the installation design, which predicts the dilution of the sewage, did not prove efficient for ^{131}I with currently WMS. The revaluation of the WMS must be carried out so that the concept of dilution of the effluent is proven so that the monitoring of the discharge is not required. Otherwise, the design and implementation of an Environmental Radiological Monitoring Program (ERMP) can assess the environmental impact with more precision (CNEN 2005), pointing out new paths based on data evaluations obtained in this proposed ERMP.

Table 5 Results related to the effective doses as a function of the collection and analysis of samples containing ^{131}I and ^{99m}Tc

Data	^{131}I	^{99m}Tc
Maximum activity A_{max} (Bq)	$6.33 \cdot 10^{5}$	$2.76 \cdot 10^{7}$
Time *t* (hour)	0.33	0.33
Specific constant Γ (μGy m^2 GBq^{-1} h^{-1})	52.20	14.10
Distance d (meter)	0.30	0.30
External effective dose E_{ext} (mSv)	$1.22 \cdot 10^{-4}$	$1.44 \cdot 10^{-3}$
Number of samples (dimensionless)	20	30
Maximum effective dose E_{max} (mSv)	$2.45 \cdot 10^{-3}$	$4.32 \cdot 10^{-2}$
Number of members of the TT	3	3
Collective effective dose E_{col} (mSv)	$7.34 \cdot 10^{-3}$	$1.30 \cdot 10^{-1}$
Total collective effective dose (mSv)	0.14	
Dose limit for individuals of the public (mSv $year^{-1}$)	1	
Optimization (mSv $year^{-1}$)	> 0.3	

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Data Availability Not applicable.

Declarations

Conflict of interest Not applicable.

References

ARPANSA (Australian Radiation Protection and Nuclear Safety Agency). (2015). *Radiation Protection of the environment – Guide G1*. Victoria, Australia.

Barquero, R., Agulla, M. M., & Ruiz, A. (2008). Liquid discharges from the use of radionuclides in medicine (diagnosis). *Journal of Environmental Radioactivy, 99*(*10*), 1535–1538. https://doi.org/10.1016/j.jenvrad.2007.12.009.

Birks, J. L. (1975). Medical radionuclides in marine environment. *Nature, 255*, 621–622.

Carolan, J. V., Hughes, C. E., & Hoffmann, E. L. (2011). Dose assessment for marine biota and humans from discharge of ^{131}I to the marine environment and uptake by algae in Sydney, Australia. *Journal of Environmental Radioactivy, 102*, 953–963. https://doi.org/10.1016/j.jenvrad.2009.10.002.

CNEN (Comissão Nacional de Energia Nuclear). (2005). *Posição Regulatória-3.01/007:2005 – Níveis de Intervenção e de Ação para Exposição Crônica*. Rio de Janeiro, RJ, Brasil (in Portuguese).

CNEN (Comissão Nacional de Energia Nuclear). (2011). *Posição Regulatória-3.01/001:2011 – Critérios de Exclusão, Isenção e Dispensa de Requisitos de Proteção Radiológica*. Rio de Janeiro, RJ, Brasil (in Portuguese).

CNEN (Comissão Nacional de Energia Nuclear). (2013). *Norma CNEN-NN-3.05: Requisitos de Segurança e Proteção Radiológica para Serviços de Medicina Nuclear. Resolução*

CNEN 159/13, 2013. In *Rio de Janeiro*. Brasil: RJ (in Portuguese).

CNEN (Comissão Nacional de Energia Nuclear). (2014). *Norma CNEN-NN-8.01: Gerência de Rejeitos Radioativos de Baixo e Médio Níveis de Radiação – Resolução CNEN 167/2014*. Rio de Janeiro,. RJ, Brasil (in Portuguese).

Fischer, H. W., Ulbrich, S., Pittauerová, D., & Hettwig, B. (2009). Medical radioisotopes in the environment – following the pathway from patient to river sediment. *Journal of Environmental Radioactivy, 100*, 1079–1085. https://doi.org/10.1016/j.jenvrad.2009.05.002.

Goddard, C. (1999). The use of delay tanks in the management of radioactive waste from thyroid therapy. *Nuclear Medicine Communitations, 20(1)*, 85–94.

IAEA (International Atomic Energy Agency). (2014). *IAEA safety standards – radiation protection and safety of radiation sources: international basic safety standards*, *General Safety Requirements Part 3, No. GSR Part 3. Planned Exposure Situations. 29*. Austria: Printed by the IAEA in Vienna.

IAEA (International Atomic Energy Agency). (2018a). *Radiation protection and safety in medical uses of ionizing radiation – specific safety guide no*, *SSG-46*. Vienna: Austria.

IAEA (International Atomic Energy Agency). (2018b). *Regulatory control of radioactive discharges to the environment – IAEA general safety guide no*, *GSG-9*. Vienna: Austria.

IAEA (International Atomic Energy Agency). (2018c). *Prospective radiological environmental impact for facilities and activities – IAEA general safety guide no*, *GSG-10*. Vienna: Austria.

ICRP (International Commission on Radiological Protection). (2004). *Release of patients after therapy with unsealed radionuclides. ICRP Publication 94*. Annals of the ICRP 34 (2). Elsevier.

ICRP (International Commission on Radiological Protection). (2007). *The 2007 recommendations of the international commission on radiological protection. ICRP Publication 103*. Annals of the ICRP 37 (2-4), Elsevier.

ICRP (International Commission on Radiological Protection). (2009). *Environmental protection: transfer parameters for reference animals and plants. ICRP Publication 114*. Annals of the ICRP 39(6). Elsevier.

ICRP (International Commission on Radiological Protection). (2017). *Dose coefficients for non-human biota environmentally exposed to radiation. ICRP Publication 136*. Annals of the ICRP 46 (2). SAGE.

ISO (International Organization for Standardization). (2007). *ISO 10703:2007 – Water quality – determination of the activity concentration of radionuclides – method by high resolution gamma-ray spectrometry*.

ISO (International Organization for Standardization). (2010). *ISO 11929:2010 – Determination of characteristic limits (decision threshold, detection limit and limits of the confidence interval) for measurements of ionizing radiation – fundamentals and applications.*

Jiménez, F., Debán, L., Pardo, L., López, R., & García-Talavera, M. (2011). Levels of ^{131}I and Six Natural Radionuclides in Sludge from the Sewage Treatment Plant of Valladolid, Spain. *Water, Air, and Soil Pollution, 217*, 515–521. https://doi.org/10.1007/s11270-010-0605-8.

Krawczyk, E., Piñero-Garcia, F., & Ferro-Garcia, M. A. (2013). Discharges of nuclear medicine radioisotopes in Spanish hospitals. *Journal of Environmental Radioactivy, 116*, 93–98. https://doi.org/10.1016/j.jenvrad.2012.08.011.

L'Annunziata, M. F. (2012). *Handbook of Radioactive Analysis* (3rd ed.). Academic Press.

Lee, U., Kim, M. J., & Kim, H. R. (2018). Radioactive iodine analysis in environmental samples around nuclear facilities and sewage treatment plants. *Nuclear Engineering and Technology, 50*, 1355–1363. https://doi.org/10.1016/j.net.2018.07.017.

Martínez, J., Peñalver, A., Baciu, T., Artigues, M., Danús, M., et al. (2018). Presence of artificial radionuclides in samples from potable water and wastewater treatment plants. *Journal of Environmental Radioactivy, 192*, 187–193. https://doi.org/10.1016/j.jenvrad.2018.06.024.

Moss, C. E. (1973). Control of of radioisotope releases to the environment from diagnostic isotope procedures. *Health Physics, 71*, 197–198.

Mundschenk, H. (1996). Occurence and behaviour of radionuclides in the Moselle River – part I: entry of natural and artificial radionuclides. *Journal of Environmental Radioactivy, 30(3)*, 199–213. https://doi.org/10.1016/0265-931X(95)00009-Y.

Ninkovic, M. M., & Androvic, F. (2005). *Chapter 1* (pp. 3–19). Gamma Radiation: Air kerma rate constants for nuclides important to gamma ray dosimetry and practical application. In. https://doi.org/10.5772/39170.

Ravichandran, R., Binukumar, J. P., Sreeram, R., & Arunkumar, L. S. (2011). An overview of radioactive waste disposal procedures of a nuclear medicine department. *Journal of Medical Physics, 36(2)*, 95–99. https://doi.org/10.4103/0971-6203.79692.

Rose, P. S., Swanson, R. L., & Cochran, J. K. (2012). Medically-derived 131I in municipal sewage effluent. *Water Research, 46*, 5663–5671. https://doi.org/10.1016/j.watres.2012.07.045.

Soares, J. A. (2019). *Protocolos Cintilográficos do Sistema Nervoso (title in Portuguese)*, *BSc Monography in Radiological Technology. Instituto Federal de Educação, Ciência e Tecnologia de Santa Catarina*. SC, Brazil: Florianópolis.

SSK (Strahlenschutzkommission). (2004). *Ermittlung der VorbelastungdurchRadionuklid-Ausscheidungen von Patient der Nuklearmedizin*. Bonn: Empfehlung der Strahlenschutzkommission (in German).

Sundell-Bergman, S., de la Cruz, I., Avila, R., & Hasselblad, S. (2008). A new approach to assessment and management of the impact from medical liquid radioactive waste. *Journal of Environmental Radioactivy, 99(10)*, 1572–1577. https://doi.org/10.1016/j.jenvrad.2007.12.005.

UNEP (United Nations Environment Programme). (2016). Radiation effects and sources: what is radiation? What does radiation do to us? Where does Radiation Come From? New York, 10.18356/b1749f17-en.