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METHOD TO DETERMINE THE RELEASE DILUTION FACTOR FOR LIQUID RADIOACTIVE EFFLUENT

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ABSTRACT

The current Brazilian regulation foresees that the operational management concerning to liquid effluents release considers the characteristics of the discharged liquid, the knowledge of the dispersion conditions after discharge into the system receptor (sewage and environment), the characteristics description of the effluent receiving system environment and the estimate of the dose. This paper describes a fast and cheap methodology to estimate the dilution factor in the liberation of liquid radioactive effluents at Instituto de Pesquisas Energéticas e Nucleares (IPEN). The radioisotope tritium, generated in the routine operation of the Research Reactor IEA-R1, was used as radiotracer. The generated radioactive effluent was stored in a 300m^3 capacity tank, has been determined initially the concentration of present radioisotope tritium. The tank flow rate exit was estimated as 10.9 ± 0.9 $m^3.h^{-1}$ for liquid controlled dispenser. A planned release for stored effluent tank was carried-out. Simultaneously it was made sampling upstream of the storage tank discharge point, monitoring the tritium concentration in the mix sewerage system point E1 at IPEN. The initial concentration of the ^3H was determined as 56881±3255 Bq L^{-1}. The estimated dilution factor for the aqueous effluent, in the discharge point E1 was of 4.3 and 7.4 respectively relative to two consecutive days of planned release and diluted effluents sampling. The developed methodology was rapid and without additional environmental and monetary costs, as it used the radiotracer tritium already existent routinely in the effluent, doesn't increment radioisotope concentrations into sewage and environment. Integrate the current goal in the world, which is the development and improvement of clean technologies.

1. INTRODUCTION

The productive human activities generate residues and effluents. Liquid and airborne effluents that may contain radioactive or hazardous constituents are continually monitored when released to the environment. A radioactive or nuclear facility should accomplish the effective environmental safety regulation, obeying the established discharge limits, in Brazil under the statement of the Comissão Nacional de Energia Nuclear (CNEN), besides the established discharge limits for the stable chemical elements.

The Instituto de Pesquisas Energéticas e Nucleares, IPEN (Nuclear and Energy Research Institute) belonging to the Comissão Nacional de Energia Nuclear, CNEN, (National Nuclear Energy Commission), was built 50 years ago, in a remote site in the city of São Paulo, Brazil. Nowadays, with the growth of the city, the institute is surrounded by industrial and residential areas. The Institute has its facilities spread over an area of about 500,000 m^2, the buildings covering 85,000 m^2. IPEN comprises several nuclear and radioactive facilities including a research reactor (IEA-R1), two cyclotrons and the radioisotopes and radiopharmaceuticals production plant. The main objectives are to perform research and developments in the field

of the peaceful uses of nuclear energy and renovated sources of energy. Activities related with the nuclear fuel cycle are also developed at the Institute, as well as research in cited fields.

IEA-R1 is the largest research reactor in Brazil, with a maximum power rating of 5 MWth. IEA-R1 is a pool reactor, with light water as the coolant and moderator, and graphite and beryllium as reflectors. The reactor was commissioned on September 16, 1957, when it achieved its first criticality. Although designed to operate at 5 MW, the reactor operated only at 2 MW between the early 1960's and mid 1980's, on an operational cycle of 8 hours a day, 5 days a week. IEA-R1 is currently operating at 3.5 MWth with a 64-hour cycle per week. The reactor originally used 93% enriched U-Al fuel elements. Currently, it uses 20% enriched uranium (U_3O_8-Al and U_3Si_2-Al) fuel that is produced and fabricated at IPEN. The reactor is operated and maintained by the Centro do Reator de Pesquisa – CRPq (Research Reactor Center) at IPEN, which is also responsible for irradiation and other services [1].

The IEA-R1 reactor is located in a multidisciplinary facility which has been consistently used for research in nuclear and neutron related sciences and engineering. The reactor has also been used for training, radioisotope production for industrial and nuclear medicine applications, and for general irradiation services. Several departments of IPEN routinely use the reactor for their research and development work. Scientists and students from universities and other research institutions also use it for academic and technological research. The largest user of the reactor is the Research Reactor Center from IPEN, which is interested in basic and applied research in the areas of nuclear and neutron physics, nuclear metrology, and nuclear analytical techniques.

In the early 1960's, IPEN produced ^{131}I, ^{32}P, ^{198}Au, ^{24}Na, ^{35}S, ^{51}Cr and labeled compounds for medical use. After 1980, it started producing ^{99m}Tc generator kits from the fission of ^{99}Mo imported from Canada. This production is continuously increasing, with the current rate of about 17000 Ci of ^{99m}Tc per year. The ^{99m}Tc generator kits, with activities varying from 250 mCi to 2,000 mCi, are distributed to more than 300 hospitals and clinics in Brazil. Several radiopharmaceutical products based on ^{131}I, ^{32}P, ^{51}Cr and ^{153}Sm are also produced at IPEN [1].

During the past few years, a concerted effort has been made in order to upgrade the reactor power to 5 MW. One of the reasons for this decision was to produce ^{99}Mo at IPEN, thus minimizing the cost and reliance on only one or two international suppliers. The reactor cycle will be gradually increased to 120 operating hours per week [1].

In order to estimate the release amount to the sanitary system, a liquid effluent monitoring program was established, in a continuous way. Then, in this IPEN program, any discharge of liquid radioactive effluent into the sewage is subject to prior measurement of volume and activity in the effluents storage tanks or bottles. Each batch of the effluents generated is representatively sampled and analyzed for radioactive isotopes prior to discharge

Effluent monitoring data are evaluated to determine the degree of regulatory compliance for each facility or the entire plant, as appropriate, it allowed an evaluation of the adequacy of the radioactive liquid effluent emission and assessment of the environmental impact due to this practice [2, 3, 4, 5, 6]. The evaluations are also useful in assessing the effectiveness of effluent treatment techniques, control systems and management practices

Tritium is produced naturally in the upper atmosphere when cosmic rays strike nitrogen molecules in the air. Tritium is also produced during nuclear weapons explosions, as a byproduct in reactors producing electricity, and in special production reactors, where the isotope lithium-6 is bombarded to produce tritium.

In research reactor IEA-R1 operation is also generated Tritium in the radioactive liquid effluent. It is a routinely monitored radioisotope. This measurement is performed after a representative sample is taken of the effluent to be discharged; it determines whether the effluent may be discharged into the receptor sewerage system and sets discharge conditions. The IEA-R1 is an integrating plant of the Centro do Reator de Pesquisa - CRPq (Research Reactor Center.) the more contributor to institutional source-term at the range time from 2004 to 2008 year [7, 8].

Radioactive wastewater generated at the IPEN-CNEN-SP is sent, via a sewer network, to the Barueri Treatment Plant (ETE-Barueri), which is owned and operated by Companhia de Saneamento Básico do Estado de São Paulo – SABESP [8]. The ETE-Barueri provides wastewater treatment for residential, commercial and industrial customers in the southwest place of the São Paulo Metropolitan Region. It is the main system under the IPEN–CNEN/SP radioactive effluent release influence.

The present paper presents an evaluation and estimative of the dilution factor, considering the recent facilities, the reshaping of the sewerage system and the Tritium concentration in the IEA-R1 retention effluent tank (TR-01) and comparative with the monitoring data downstream of discharge point. Dilution of effluents is one parameter that was used for estimating a more realistic radioactive dilution effluent factor.

Consideration of dilution factors liquid effluents released from nuclear and radioactive facility in Brazil is a recent rule [4, 5, 6]. Then, as the release is strongly dependent on the total amount of the effluent and on the dilution factor, special attention is needed in order to obtain the correct value of that last one [7, 8].

2. ESTIMATIVE OF DILUTION FACTOR USING TRITIUM RADIOTRACER

An important target of the Brazilian government is to increase the participation of nuclear energy in the national electric power production. This involves the continuous development of technology for the design, construction and operation of nuclear power plants and industrial facilities related to the nuclear fuel cycle. Its include the development of technologies resources for the establishment and continuity of monitoring and management of environment and health. Actually, one of the items of integrated management is the National Nuclear Power Policy, aiming at guiding research, development, production and safe use of all forms of nuclear energy, environmental planning and rules compliance.

Currently, Brazil has some operating nuclear and radioactive plants. The Figure 1 presents the location of the main Brazilian nuclear and radioactive facilities and organizations [1] in the territorial country map. Besides, presents the study area at IPEN, a reshaped actual sewerage system and effluents pipes with the discharge sanitary studied point (E1) [7, 8].

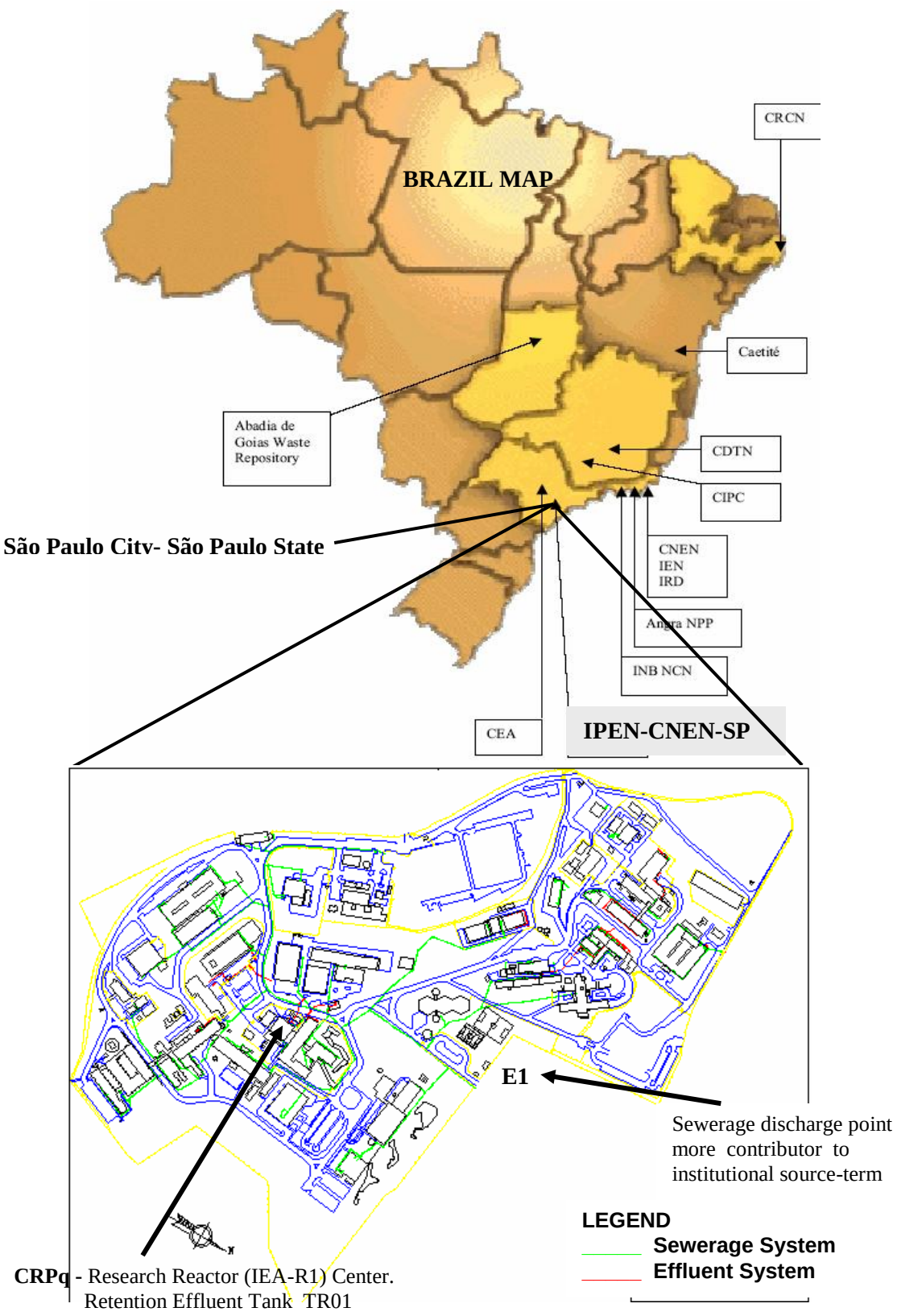


Figure 1. Study area at IPEN showing the retention effluent tank and discharge point for radiotracer Tritium dilution factor estimative [1, 7, 8]. IPEN location and Brazilian facilities [1]

2.1 Tritium Radiotracer Use

Liquid effluents from TR01 with a potential to contain radioactive materials at prescribed threshold levels were measured for specific radionuclides. Quantities of the radionuclides ^{58}Co, ^{60}Co, ^{137}Cs, ^{54}Mn, ^{124}Sb and ^{65}Zn representing a total of 5.89 10^8 Bq and tritium with a total 1.08 10^{10} Bq, 2008 year source-term. However, most radionuclides in effluents at the tank TR01 are approaching levels indistinguishable from background concentrations. Tritium was measured by liquid scintillation counting (LSC) technique [7].

Tritium may be produced in nuclear reactors by the following five mechanisms: (a)fissioning of uranium, (b) neutron capture reactions with boron and lithium added to the reactor coolant, (c) neutron capture reactions with boron in control rods, (d) activation of deuterium (hydrogen-2) in water, and (e) high energy neutron capture reactions with structural materials.

The relative magnitude of tritium production by these sources is influenced by the reactor type, operating history, design characteristics, and materials of construction. The amount of tritium and manner in which it is released to the environment will also be affected by these parameters.

During the fission process the uranium nucleus usually splits into two more or less equal fragments plus several neutrons. About once in every 10,000 fissions, however, the nucleus is split into three portions (ternary fission) one of which may be a Tritium nucleus. This Tritium normally remains in the fuel unless it diffuses through the cladding material or a leak occurs.

A second source of fission-product Tritium in nuclear reactors is due to traces of uranium on the outer side of fuel elements which remain from the fuel fabrication process. This tramp uranium may be only a few micrograms per square inch of fuel surface, but because of the large surface area provided by the many fuel rods in a reactor it can produce detectable fission-product concentrations in the coolant.

The liquid scintillation counting is an analytical technique which is defined by the incorporation of the radionuclide into uniform distribution with a organic liquid chemical medium capable of converting the kinetic energy of nuclear emissions into light energy photons. The scintillation fluid is a mixture of 3 chemicals (solvent, emulsifier, and fluorine) which produces light flashes when it absorbs the energy of the radioactive decay particle.

The interaction of the energies beta for the liquid middle is processed by excitement, and they emit photons of ultraviolet radiation returning to first fundamental state. The photons happen in a tube photomultiplier, where the electrons are multiplied generating a proportional electric sign to the number of incident photons. The sample Tritium concentration was determined by the equation 1.

$$C = \left(\frac{Ca}{Ef_{a.V}} - \frac{Bg}{Ef_{bg.V}} \right). \quad (1)$$

Where:

***C*:** Tritium sample concentration (Bq L^{-1})
***C*$_a$:** sample counting (cps)
***Bg*:** background radiation (cps)

***Ef*$_a$:** sample counting efficiency (cps dps^{-1})
***Ef*$_{bg}$:** background counting efficiency ($cps.dps^{-1}$)
***V*:** sample volume (L)

Quench is a reduction in system efficiency as a result of energy loss in the liquid scintillation solution. Because of quench, the energy spectrum detected from the radionuclide appears to shift toward a lower energy. The three major types of quench encountered are photon, chemical, and optical quench. Chemical, sometimes called impurity, quenching causes energy losses in the transfer from solvent to solute. Photon quenching occurs with the incomplete transfer of beta particle energy to solvent molecules. Optical or color quenching causes the attenuation of photons produced in solute.

The Quench-indicating Parameter is the Spectral Index of the Sample (QIP) and the external source quench-indicating parameter is the transformed Spectral Index of the External Standard (tSIE). The counting efficiency was certain for the methodology "transformed Spectral Index f the standard External" (tSIE) using a source ^{133}Ba, wherein each radionuclide equation is near linear and is expressed in the form below.

$$y = mx + b \tag{2}$$

Where:

Where:
***y*:** QIP value;
***m*:** slope of plot of SIS versus tSIE;
***x*:** tSIE value; and
***b*:** intercept on SIS axis of a plot of SIS versus tSIE.

The dilution factor estimated for radioactive sample effluent in the sewerage point E1 was obtained by equation 3.

$$Fd_{E1,i} = \frac{C_{tr1,i}}{C_{E1,i}} \tag{3}$$

Where:

***Fd*$_{E1,i}$:** sewerage point E1 Dilution Factor for radioisotope i.
***C*$_{tr1,i}$:** radiotracer i initial concentration (Bq L^{-1}) for effluent inside tank TR1.
***C*$_{E1,i}$:** radiotracer i concentration (Bq L^{-1}) for sewerage point E1 effluent sampled (Bq L^{-1})

2.2 Sample Pre-treatment and Measurement

The radioactive liquid effluents samples were homogenized by magnetic plate and an aliquot of 50 mL was transferred with volumetric pipette for a distillation apparatus. Constituted by a round bottom flask coupled to a straight condenser, staying the controlled temperature from 96 to 97 °C with heating controller. The distillation process was very slow. The first 10 mL of the distilled was discarded, after was recoiled a distilled volume of about 10 mL.

An aliquot of 1,2 mL was transferred by using micro-pet inside a 20 mL capacity scintillating vial. Scintillator solution Instagel-XF, 18 mL, was increased and gentle mixed for

approximately 1 minute, until the total solution homogenization.After the homogenization the cocktail was refrigerated and kept at dark place by a minimum period of two hours before the liquid scintillation counting.

2.3 Calibrations and Methodology Verification

A Tri-Carb model 2100TR Liquid Scintillation Counter composed by two photomultipliers tube coupled to a discriminator signal pulses was calibrated with the adopted cocktail. The time counting was of 120 minutes. For background radiation detection system determination same scintillating vial kind with deionized water was used, in the same amount and experimental conditions.

In this study, the efficiency counting was determined by the relative quenching of the sample, by using an external source quench-indicating parameter (QIP) such as the Spectral Index of the External Standard (SIE). This method involves counting the sample with a gamma radiation source (^{133}Ba) external the vial and subsequently in the absence of the gamma radiation source such that the contribution of only the gamma radiation source is studied. The gamma radiation source generates compton electrons in the sample solution which behave in a similar manner to decaying nuclear particles.

If quenching is present the pulse-height energy distribution spectra from the gamma radiation generated events will be compressed towards a lower apparent energy. External standard quench-indicating parameters (QIP) include the external standard ratio (ESR), Spectral Index of the External Standard (SIE); H Number, ESP and the transformed Spectral Index of External Standard (tSIE).

For the curve quenching construction was used 6 quench cocktail standard with different quenching agents with activity of 327 ± 18 Bq and an standard without quench with activity 4630 ± 68 Bq. The efficiency results and the propagated uncertainties are presented in the Table 1.

Table 1: Calculated counting efficiency by tSIE method for Tritium liquid scintillation counting

Activity (Bq)	QIP	Efficiency (cps dps^{-1})
4630	976	0.621 ± 0.023
327	614	0.500 ± 0.033
327	499	0.447 ± 0.029
327	396	0.382 ± 0.025
327	320	0.328 ± 0.022
327	238	0.250 ± 0.016
327	114	0.095 ± 0.006

The Figure 2 presents the curve with quenching parameters for ^{3}H calculated for the present study, the ones which were adjusted by shown degree 4 polynomial equation.

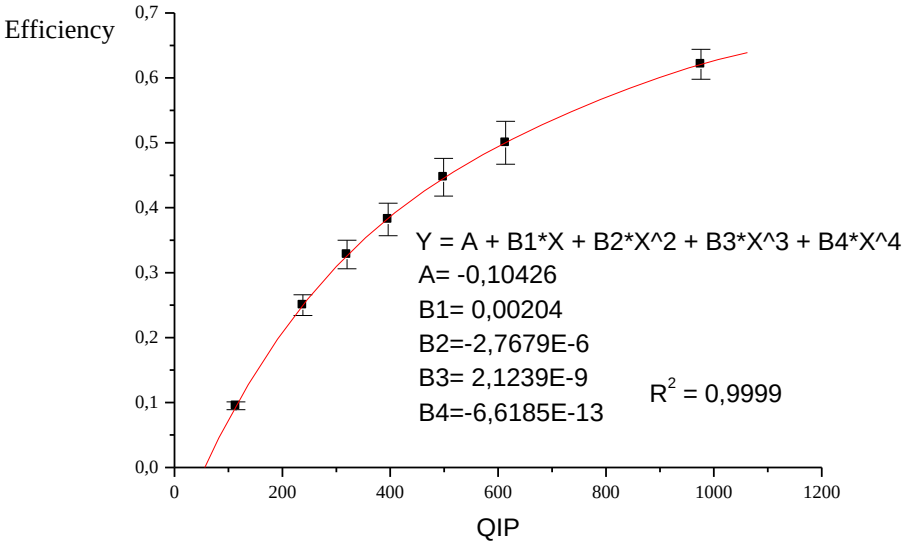


Figure 2. Quenching curve obtained for Tritium radioisotope by liquid phase scintillation counting

The ^{3}H calibration results are shown in Table 1 and Figure 2. Note the extremes of values for the quenching parameter (tSIE). A maximum efficiency of approximately 62% was achieved with a quench parameter (QIP) of 976. The medium efficiency of about 38% was obtained with a quench QIP of 396. Thus a quench QIP of 114 or below would result in essentially very close background counts (efficiency < 10%). The Figure 3 shows the distillation apparatus, liquid scintillation counter system and quenching standard solutions kit used.

Figure 3. Distillation apparatus, liquid scintillation counter and quenching standard solutions

The reliability efficiency and methodology verification was accomplished by participation in the Brazilian Intercomparison Analytical Results Run, called Programa Nacional de Intercomparação (PNI), following the same procedure. For the runs of December 2007 and August 2008 the analytical results obtained for Tritium quantification presented an excellent agreement with the real value.

After calibration and method verification the counter was then used to measure tritium levels in effluents samples from tank TR01 and discharge sewerage point E1.

2.4 Effluent Dilution Factor Estimative by Tritium Effluent Radiotracer Use

The procedure for effluent dilution factor determination in the sewerage discharge point E1 was the following. Three samples of 1.00 ± 0.10L were collected of the retention tank TR1 before of the planned and controlled effluent release. The ^{3}H, generated as operational unfavorable IEA-R1 research reactor radioisotope, was used as a able radiotracer.

No environmental or financial costs were added by this operational and *in loco* radiotracer assay. The concentration of ^{3}H was analyzed by LSC, three spectra evaluated and calculated the concentration average and their respective standard deviations. The found value for Tritium concentration was 56881 ± 3255 Bq L^{-1} before planned effluent TR1 discharge.

The flow rate effluent discharge retention tank TR1 was estimated by measuring the inside tank liquid high. The estimated value of 10.9 ± 0.9 $m^{3}.h^{-1}$ for liquid controlled dispenser.

After the initial sampling effluent TR1 tank, continuous samplings were carried out in the discharge point E1, simultaneously to the effluent tank TR1 controlled release. The ^{3}H concentration was obtained by LSC, for each point E1 effluent and spectra evaluated for calculation of the ^{3}H concentration average and their respective standard deviations.

The ^{3}H concentration obtained for effluent TR1 tank and for the effluent point E1 were compared and the pertinent dilution factor was estimated by equation 3. The Table 2 presents the results of the ^{3}H radiotracer concentration and uncertainties (Bq L^{-1}), for each point E1 collected effluents samples, during the retention tank TR1 controlled effluent discharge, for two consecutives study days, may 2008 period. The Table 2 shows also the estimative and average effluent dilution factor, for sewerage effluent point E1. The Figure 4 presents the retention tank TR1 and station monitoring E1 sewerage point with sampling effluent.

Table 2: Concentrations and uncertainties of the ^{3}H radiotracer in sewerage E1 IPEN point effluents samples and estimated dilution factor.

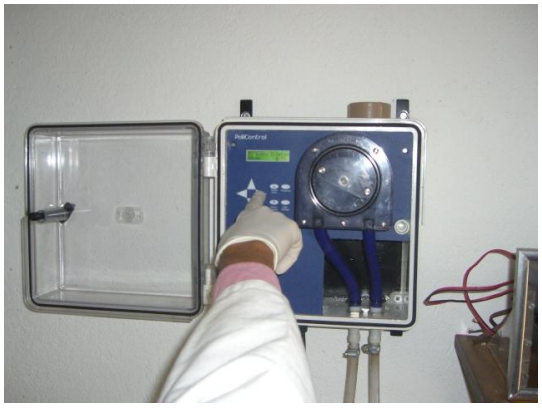
Day	Time	^{3}H (Bq L^{-1})	Dilution Factor
1	9h37	10361 ± 518	5.5
1	10h35	14881 ± 748	3.8
1	11h31	21963 ± 1098	2.6
1	12h37	14629 ± 732	3.9
1	13h33	11247 ± 563	5.1
1	14h30	11913 ± 596	4.8
average			**4.3**
2	9h35	9727 ± 486	5.8
2	10h30	9849 ± 493	5.8
2	11h20	5367 ± 269	10.6
average			**7.4**

As shown in the Table 2, the estimated dilution factor for the aqueous effluent, in the discharge point E1 was of 4.3 and 7.4 respectively to day 1 and 2, consecutive for planned release and diluted effluents sampling.

Effluent retention tank TR1

Internal access to effluent retention tank TR1

Sewerage discharge point E1 Station



Adjustment of the sampling peristaltic pump

E1 point sewer pipe entrance

Cooled E1 sewerage discharge point E1 sample

Figure 4 Retention tank TR1 and station monitoring E1 sewerage point sampling effluent.

Tritium is a hydrogen atom that has two neutrons in the nucleus, in addition to its single proton, giving it an atomic weight near three. Although tritium can be a gas, its most common form is in water, because, like non-radioactive hydrogen, radioactive tritium reacts with oxygen to form water. Tritium replaces one of the stable hydrogens in the water molecule, H_2O, tritiated water. Tritium readily forms water when exposed to oxygen. Like H_2O, tritiated water is colorless and odorless. Tritium has a half-life of 12.3 years and emits a very weak beta particle and transforms to stable, nonradioactive helium.

Tritium atoms can exchange with any hydrogen atoms. If the hydrogen atom is part of an organic molecule, the tritium becomes organically bound and is transported with the molecule rather than moving freely like water. Thus, the equation 4 and 5 present the mechanisms for Tritium dispersion and dilution in aqueous medium.

$$2\,H_2O \underset{k_w}{\rightleftharpoons} H_3O^+ + OH^- \tag{4}$$

In a simplified way it can be written as the equation 5

$$H_2O \underset{k_w}{\rightleftharpoons} H^+ + OH^- \tag{5}$$

Where:
k_w: self-ionization water equilibrium constant

The ion H^+ present could be formed starting from the atoms of the Hydrogen -^{1}H or Tritium -^{3}H, which possess same properties and behavior physical, chemical and energy, besides all the characteristics of the element Hydrogen (Z=1). The dilution factor estimated for Tritium radioisotope corresponds to the water dilution factor. Emphasizing that water is the solvent or macro constituent of the effluents release at TR1 tank IPEN. The Figure 5 shows a procedure protocol and practical dilution factor as environmental management instrument.

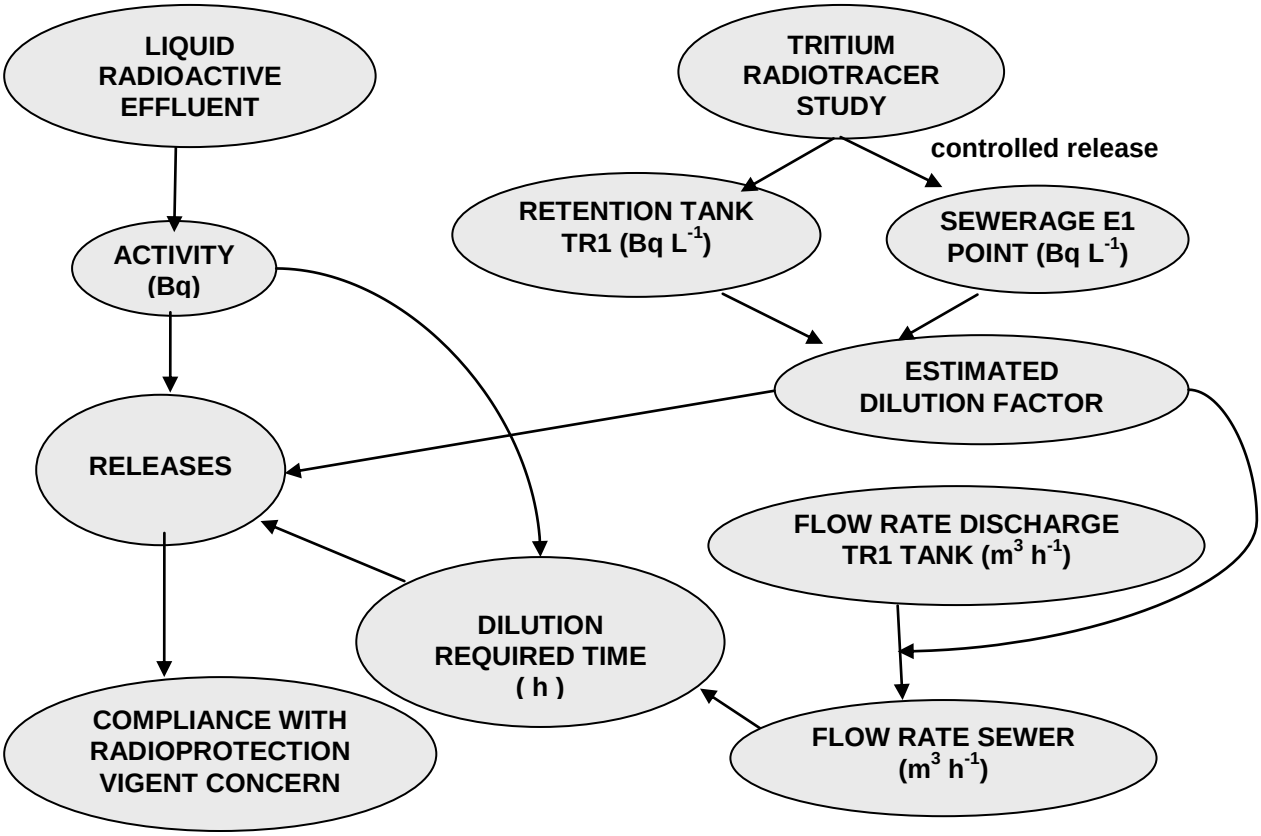


Figure 5 Practical uses of dilution factor to environmental management.

3 CONCLUSIONS

An inside-field dilution factor calculated by this study appears to be a reasonable assumption to management and decisions concerning on radioprotection practices and regulatory adjustment. Previously assumed dilution factors were for shoreline points, outside the plant. It is the first study contributing to internal dilution factor statement.

The dilution volume (or flow) must be determined specifically for each plant. In addition, a table of expected dilution volumes may be prepared by continuous monitoring.

Known dilution factors within the sewage system will be useful to the individual user, but it is apparent that the daytime dilution factor within the sewage network will not be the same as nighttime dilution factors. The estimated dilution factor for the aqueous effluent, in the discharge point E1 at IPEN was of 4.3 and 7.4 respectively to day 1 and 2, daytime consecutive for planned release TR1 effluent.

The liquid scintillation counting shows to be able to measure Tritium concentration and shows method compliance by intercomparison run participating.

Known dilution factors within the sewage system of the liquid radioactive effluents, besides contribute to adjusting the current practices to the recent rules must contribute to optimization of several procedures.

The ^{3}H, generated as operational unfavorable IEA-R1 research reactor radioisotope, was used as a able radiotracer.No environmental or financial costs were added by this operational and *in loco* radiotracer assay.

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