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Anthropogenic radiotracers assessment as tool to environmental compliance rules and management

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Abstract A framework, within models and studies can be applied, to develop coherent and logical environmental impact management methodologies for ionizing radiation, is essential. A number of components, which form the basis for such a system, included the dilution factor for radioactive liquid effluent releases. In this study we established a strategic, fast and cheap methodology to estimate the dilution factor for the release of liquid radioactive effluents at IPEN. The radioisotopes ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn, and ^{137}Cs, generated in the routine operation of the Research Reactor IEA-R1, were used as radiotracers. The generated liquid radioactive effluent was stored in a 300 m^3 capacity tank. The initial concentration of its present radioisotopes were determined. A planned release of effluent from the storage tank was carried-out. Simultaneously, the sampling was made upstream of the storage tank discharge point and the concentration ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs in the mix sewerage system discharge point E1 at IPEN were monitored. The estimated dilution factor of discharge point of the aqueous effluent for ^{3}H was 4.3 and 7.4, ^{54}Mn was12 and 16.1, ^{60}Co was 12.6 and 14.2, ^{65}Zn was 12 and 27.9 and ^{137}Cs was 6.2 and 13.9, respectively.

Keywords Liquid radioactive effluents · Estimated dilution factor · Liquid scintillation counting · Gamma-ray spectrometry

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Introduction

Under the existing Brazilian Regulatory requirements for liquid effluents [1–4], all liquid released from productive human activities as industries, nuclear power plants, radioactive facilities and others shall be discharged to the environment in a planned manner via systems and programs designed for that purpose. Then, all discharges from the nuclear plants, radioactive facilities or non-radioactive industries site are subject to operating controls to ensure that they comply with the regulations governing sampling and discharges.

Liquid and airborne effluents that may contain radioactive or hazardous constituents, shall be continually monitored. A radioactive or nuclear facility should accomplish the effective environmental safety regulation, following the established discharge limits under the statement of the Comissão Nacional de Energia Nuclear (CNEN), besides the established discharge limits for the stable chemical species [2–4].

The Instituto de Pesquisas Energéticas e Nucleares, IPEN (Nuclear and Energy Research Institute) belongs to the Comissão Nacional de Energia Nuclear, CNEN (National Nuclear Energy Commission). It 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 with the buildings cover 85,000 m^2. IPEN comprises several nuclear and radioactive facilities including two cyclotrons, radioisotopes and radiopharmaceuticals production plant, and a research reactor (IEA-R1). IPEN's 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

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also developed at the Institute, as well as research in cited fields.

Effluent monitoring data are evaluated to determine the effectiveness of regulatory compliance for each facility or the entire plant, as appropriate. If it allowed an evaluation of the adequacy of the radioactive liquid effluent emission and assessment of the environmental impact due to this practice [1, 5–8]. The evaluations are also useful in assessing the effectiveness of effluent treatment techniques, control systems and management practices.

The main objective of this study was establishment of a strategic methodology to estimate the dilution factor for liquid effluent, a parameter requested by Brazilian laws. It was carried-out for the liquid radioactive effluents released at IPEN. Had been considered the increased concentrations of studied radiotracers from storage tank TR01, to discharged samples with in discharge point E1.

Methodology

Liquid effluents released from retention effluent tank (TR01) had a potential to contain radioactive materials at prescribed threshold levels. They were measured for specific radionuclides, ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn, and ^{137}Cs, generated in the routine operation of the Research Reactor IEA-R1. In a planned effluent release samples were collected simultaneously at sewerage point E1 and determined for same radioisotopes. These radioisotopes were used as radiotracers.

Tritium was measured by liquid scintillation counting (LSC) system and ^{60}Co, ^{137}Cs, ^{54}Mn, ^{65}Zn were measured by gamma-ray spectrometry [9]. Presently, the main contributor to institutional source-term was the study area at IPEN, a reshaped actual sewerage system and effluents pipes with the discharger sanitary studied point E1 [10].

LSC is an analytical technique which is defined by the incorporation of the radionuclide into uniform distribution with an organic liquid chemical media capable of converting the kinetic energy of nuclear emissions into light energy photons. The radioactive liquid effluents samples were homogenized on a magnetic plate. An aliquot of 50 mL of mixed effluent was transferred with a volumetric pipette to a distillation apparatus, constituting by a round bottom flask coupled to a straight condenser. The distillation temperature was controlled to be between 96 and 97 °C. The distillation process was very slow. The first 10 mL of the distillate was discarded. An aliquot of 1.2 mL of the distillate was then transferred with a pipette into a 20 mL capacity scintillating vial. Scintillator solution Instagel-XF, volume of 18 mL, was added and gently mixed for approximately 1 min, until the total solution homogenized. After the homogenization the cocktail was refrigerated and kept in dark place for a minimum period of two hours before count in the liquid scintillation counting system (TRI-CARB 2100TR).

Activity concentrations of ^{60}Co, ^{137}Cs, ^{54}Mn and ^{65}Zn were measured by gamma-spectrometry with hyper-pure germanium detector, EGPC 25 coaxial, 25 % relative efficiency and 2.10 keV at 1,332 keV resolution (INTERTECHNIQUE) coupled to an amplifier and multichannel analyser. The detector was calibrated using radionuclides source certified by Amersham. Each sample was packed in a polyethylene flask of 0.85 L in volume (F850) and counted for 10,000 or 50,000 s. Spectra were analyzed by a ORTEC WinnerGamma software. The ^{60}Co content was determined from mean activity of two photopeaks of the 1,173 and 1,332 keV gamma-ray. The ^{137}Cs, ^{54}Mn and ^{65}Zn were determined from the 661, 834 and 1,115 keV gamma-ray peak, respectively.

The dilution factor estimated for radioactive sample effluent in the sewerage point E1 was obtained by Eq. 1.

$$\mathrm{Fd}_{\mathrm{E1},i} = \frac{\mathrm{C}_{\mathrm{tr1},i}}{\mathrm{C}_{\mathrm{E1},i}} \quad (1)$$

where: $\mathrm{Fd}_{\mathrm{E1}}$ sewerage point E1 Dilution Factor for radioisotope i, $\mathrm{C}_{\mathrm{tr1,i}}$Radiotracer I Initial Concentration (Bq L^{-1}) For Effluent Inside Tank TR1, $\mathrm{C}_{\mathrm{E1,i}}$'radiotracer i concentration (Bq L^{-1}) for sewerage point E1 effluent sampled

Results and discussion

The procedure for effluent dilution factor determination in the sewerage discharge point E1 was the following. Three samples of 1.00 ± 0.10 L in volume were collected from the retention tank TR1 before planned and controlled effluent release. The ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs, generated as operational unfavorable by product of IEA-R1 research reactor were used as a able radiotracer.

No environmental or financial costs were needed by this operational and *in loco* radiotracer assay. The concentration of ^{3}H was analyzed by LSC and ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs, were analyzed by gamma-spectrometry, three spectra were evaluated and calculated for the concentration average and their respective standard deviations. Tritium concentration before planned effluent TR1 discharge was 56,881 ± 3,255 Bq L^{-1} and those for ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs, were 41.5 ± 2.9, 1,332 ± 11, 401 ± 8 and 291 ± 7 Bq L^{-1},respectively.

The effluent discharge flow rate in retention tank TR1 was estimated by measuring the inside tank liquid level. The estimated value was 10.9 ± 0.9 m^{3} h^{-1}.

After the initial sampling of effluent TR1 tank, continuous samplings were carried out at the discharge point E1, simultaneously to the effluent tank TR1 controlled release.

Table 1 Concentrations and uncertainties of the ^{3}H radiotracer in sewerage E1 IPEN point effluents samples and estimated dilution factor

Concentrations (Bq L^{-1})						
Days	Time	^{3}H	^{54}Mn	^{60}Co	^{65}Zn	^{137}Cs
1	9 h37	10,361 ± 518	2.1 ± 0.8	60.4 ± 2.4	18.4 ± 2.2	72.2 ± 3.0
1	10 h35	14,881 ± 748	3.0 ± 1.2	84.3 ± 3.2	39.6 ± 3.1	43.4 ± 2.1
1	11 h31	21,963 ± 1,098	5.6 ± 1.2	130 ± 5	48.6 ± 4.3	65.9 ± 2.9
1	12 h37	14,629 ± 732	4.9 ± 1.0	146 ± 5	46.2 ± 3.5	55.8 ± 2.6
1	13 h33	11,247 ± 563	3.6 ± 1.2	142 ± 5	35.5 ± 3.1	39.4 ± 2.1
1	14 h30	11,913 ± 596	3.8 ± 1.2	139 ± 5	33.9 ± 3.3	30.8 ± 1.8
2	9 h35	9,727 ± 486	2.7 ± 1.0	110 ± 4	15.8 ± 2.5	34.8 ± 1.9
2	10 h30	9,849 ± 493	3.7 ± 1.1	113 ± 4	15.8 ± 2.2	24.7 ± 1.7
2	11 h20	5,367 ± 269	1.8 ± 1.0	69.6 ± 2.7	12.2 ± 2.0	13.5 ± 1.3

The ^{3}H concentration was obtained by LSC and ^{54}Mn, ^{60}Co, ^{65}Zn, and ^{137}Cs were analyzed by gamma-spectrometry. For each point E1 effluent, the spectra were evaluated for the ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn, and ^{137}Cs concentration average and their respective standard deviations.

The ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn, and ^{137}Cs concentrations obtained for effluent TR1 tank and the effluent point E1 were compared and the pertinent dilution factor was estimated by Eq. 1. Table 1 presents the results of the ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn, and ^{137}Cs radiotracers concentrations and their uncertainties (Bq L^{-1}), in collected effluents samples for each time at point E1, during the retention tank TR1 controlled effluent discharge, for two consecutives study days.

The ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn, and ^{137}Cs concentrations obtained for effluent TR1 tank and effluent point E1 were compared and the pertinent dilution factor was estimated by equation 1. Table 2 presents the estimated dilution factor for the aqueous effluent, in the discharge point E1.

During planned effluent release from Tank T01 and collection of diluted radioactive effluents samples at point E1, as shown in the Table 2, the estimated dilution factor for the aqueous effluent, in the discharge point E1 averaged 4.3 and 7.4 for ^{3}H, 12 and 16.1 for ^{54}Mn, 12.6 and 14.2 for ^{60}Co, 12 and 27.9 ^{65}Zn, and 6.2 and 13.9 for ^{137}Cs, for day 1 and 2, consecutively. The ratio of the dilution factors calculated for ^{3}H and ^{137}Cs are coherent with the ratio verified in the 12 hydrometers distributed in the campus of IPEN in the days of the samplings, the ratio of the hydrometers was of the approximate 1.9.

It was observed from data in Table 2, that the value of the dilution factor varied for radiotracer, in sequence of ^{3}H, ^{137}Cs, ^{54}Mn, ^{60}Co, and ^{65}Zn.

This study did identify that the potential exists for planned and monitored application uses of containing non-radioactive elements and radioactive isotopes releases as tracers or radiotracers respectively. Dilution factor estimative and uses for optimization and improvement of new procedures and environmental management approaches for all kind of liquid effluent released from productive human activities are possible, as chemical industries, nuclear power plants, radioactive facilities and others. These practices permit no increment of new chemical elements or radioisotopes to environment, no costs, no contamination of waters by new substances, fast and clean methodology.

Table 2 Estimated dilution factor for the aqueous effluent

Days	Time	^{3}H	^{54}Mn	^{60}Co	^{65}Zn	^{137}Cs
1	9 h37	5.5	19.8	21.9	21.7	4.0
1	10 h35	3.8	13.8	15.7	10.1	6.7
1	11 h31	2.6	7.6	10.2	8.3	4.4
1	12 h37	3.9	8.5	9.1	8.7	5.2
1	13 h33	5.1	1.4	9.3	11.3	7.4
1	14 h30	4.8	10.9	9.5	11.8	9.4
13/05/08	Average	4.3	12.0	12.6	12.0	6.2
2	9 h35	5.8	15.4	12.0	25.5	8.4
2	10 h30	5.8	11.0	11.7	25.4	11.8
2	11 h20	10.6	21.9	19.0	32.9	21.5
14/05/08	Average	7.4	16.1	14.2	27.9	13.9

Another aspect in this connection is the study of synergistic or antagonistic effects between different pollutants. The large injections of radionuclides from nuclear weapons testing, reprocessing and the nuclear accident, added to chemical composition increment to environment by industries has made it possible. Similar protocols could be used to radioecology and chemoecology studies.

Conclusion

An in situ dilution factors calculated by this study appears to be a reasonable assumption for management and decisions concerning on radioprotection practices and regulatory adjustment, contribute to adjusting of the current

practices to the recent rules and also to optimization of several procedures. It must be determined case by case, in addition, a table of expected dilution volumes may be prepared by continuous monitoring. It is apparent that the dilution factor within the sewage network during daytime will not be the same as that of the nighttime.

The estimated dilution factor for the aqueous effluent, in the discharge point E1 at IPEN averaged 4.3 and 7.4 for ^{3}H, 12 and 16.1 for ^{54}Mn, 12.6 and 14.2 for ^{60}Co, 12 and 27.9 ^{65}Zn and 6.2 and 13.9 for ^{137}Cs, for day 1 and 2 consecutive for planned release TR1 effluent, respectively.

In the operational level, it was verified that the dilution factor is differentiated by chemical group. The estimation of the factors of dilution of ^{3}H and the alkaline metal ^{137}Cs, had presented consistent values between them and with the ratio of the twelve hydrometers installed at IPEN. Concerning on metals, physical and chemical characteristics, ion complex behavior, low mobility for some chemical species in waters, the possible transformations and reactions inside tank TR1 and pipes, feature the tank TR1, the dynamics of homogenization and the release, co-precipitation, sorption, desorption, could explain the values for estimated dilution factors for the radiotracers ^{54}Mn, ^{60}Co, and ^{65}Zn.

The liquid scintillation counting used for measuring of Tritium concentration and gamma-spectrometry used for measuring of ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs, both methods showed compliance by participating in intercomparison run.

The developed methodology was rapid and without additional environmental and monetary costs, as it used the radiotracer already exist in the effluent, doesn't increase radioisotope concentrations into sewage and environment. It integrate, the current goal in the world, which is the development and improvement of cleaner production technologies.

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