Journal of Environmental Radioactivity xxx (2016) 1–6

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

The effectiveness of wastewater treatment in nuclear medicine: Performance data and radioecological considerations

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ARTICLE INFO

Article history:
Received 29 October 2015
Received in revised form
10 March 2016
Accepted 3 April 2016
Available online xxx

Keywords:
Radioiodine therapy
Liquid radioactive waste
Discharge control
Wastewater radioactivity
Radioecology

ABSTRACT

For planned and ongoing storage of liquid radioactive waste in a designated plant for a nuclear medicine therapy ward (decontamination system/decay system), detailed knowledge of basic parameters such as the amount of radioactivity and the necessary decay time in the plant is required. The design of the plant at the Department of Nuclear Medicine of the University of Cologne, built in 2001, was based on assumptions about the individual discharge of activity from patients, which we can now retrospectively validate. The decontamination factor of the plant is at present in the order of 10^{-9} for ^{131}I. The annual discharges have been continuously reduced over the period of operation and are now in the region of a few kilobecquerels. This work emphasizes the high efficacy of the decontamination plant to reduce the amount of radioactivity released from the nuclear medicine ward into the environment to almost negligible levels.

1. Introduction

Much attention has been paid to the accumulation of radioactivity in wastewater since the Chernobyl accident in 1986. But from a more general radioecological point-of-view, investigations into this topic have continued [Imhoff et al., 1988, Martin and Fenner 1997, Nedveckaite et al., 2000]. Public interest was focused again on measurements of radioactivity in water after the Fukushima accident [Kosaka et al., 2012, Manolopoulou et al., 2011]. The possibility of transferring radioactive effluents into potable water was and still is a crucial aspect of radioecology.

Apart from the radiological accidents discussed, the contribution of radioactivity in wastewater from nuclear medicine departments needs to be considered. In nuclear medicine departments radioactive substances are administered for diagnostic and therapeutic purposes. Despite the considerable benefits for patients, the problem of a small but not negligible radiation exposure remains.

a) for the patient,
b) for the personnel in the nuclear medicine department and
c) for members of the public.

A minor radiation exposure to members of the public will inevitably occur after nuclear medicine procedures. Nevertheless, it is still mandatory to minimize the possible sources of radiation exposure to an acceptable level. It is furthermore imperative that the exposure of members of the public to radiation does not exceed legal limits (1 mSv). Exposure of the patient as a result of the application of radiopharmaceuticals [Stabin et al., 1999] and of personnel has been exhaustively studied [Sudbrock et al., 2008, Sudbrock et al. 2009] but little is known about the excretion and exhalation of radioactivity by the patient [Schomäcker et al., 2011, Wellner and Schicha 1993].

A simple and enormously effective but time-consuming way to reduce this transfer of radioactivity into the sewage system can be achieved by the natural decay of radionuclides and hence a delayed discharge so that no further removal of radionuclides is required [Goddard 1999, Wellner and Schicha 1993]. Though criticized as expensive, the advantages of decay systems for short-lived nuclides ($T_{1/2}$ < 100 d) are obvious and it is still the standard approach of widespread use in many countries [Andrés et al., 2011, Barquero et al., 2008, Driver and Packer 2001, Goddard 1999, Leung and Nikolic 1998, Ravichandran et al., 2011, Sundell-Bergman et al., 2008]. Alternative approaches have also been developed [Rodríguez, 2012].

With a total of about 50 000 radioiodine therapies with roughly

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http://dx.doi.org/10.1016/j.jenvrad.2016.04.002
0265-931X/

50 TBq radioiodine administered per year in Germany alone, this could account for a significant elevation of the respective radionuclide concentrations in the environment. In addition, two million diagnostic procedures with a variety of nuclides are carried out [per year] (Table 1). In Germany it is therefore mandatory to release radioactive fluids to the public sewage system when the respective activity concentrations fall below the exemption level (maximum permissible activity, MPAC).

Diagnostic procedures typically make use of short-lived radionuclides (Table 1) which will not pose serious difficulties due to their short decay time, but for therapeutic procedures nuclides with longer half-lives are relevant (Table 1). In particular the so-called radioiodine therapy which makes use of the isotope ^{131}I plays a predominant role with respect to wastewater management in nuclear medicine [Fischer et al., 2009]. From the radioecological point-of-view, it should be mentioned that the most important nuclide, ^{131}I, is well-known in radioecology and that most of the nuclides used for radiotherapy can be measured easily and with low detection limits by means of gamma-ray spectrometry (Table 1).

This work presents, as an example, the design and operation details of the wastewater plant of the University Hospital of Cologne.

2. Materials and methods

2.1. Wastewater storage and the decontamination plant

The liquid waste storage and decontamination plant ("cooling plant") was built in 2001 (EnviroDTS, Friedberg, Germany). It consists of a closed tank-stack system, as described by [Westmeier 2012], where the liquid waste is treated and measured automatically while decaying. The system was planned and designed for a nuclear medicine department with a diagnostic, a therapeutic and a radiochemical section. A stack of 19 collector (or "decay") tanks was installed for the whole department. These tanks are separated into five subdivisions (A and B: inflow from the therapy ward, C: inflow from diagnostic procedures, D and E: inflow from laboratories) each separating an inflow representing a "typical" nuclide composition. Inflow from the controlled area is completely discharged into the decontamination plant. Table 2 shows an outline of the storage and decay system with a focus on the assignment of tanks and their respective capacity.

The radioactive inventory of each cooling tank is determined via γ-ray spectrometry. For this purpose, a 2"x2" NaI(Tl) scintillation counter is inserted into each collector tank via an immersion tube. All measurements are performed and analysed automatically (program "Sodigam", Dr. Westmeier GmbH, Ebsdorfergrund, Germany), proceeding in a cyclical manner from one decay tank to the next. The activity concentration is calculated from the spectra when the activity concentration is low enough to avoid the necessity of correcting count-rate losses due to dead-time of the counter. The measuring time is adjustable - currently 10 800 s (3 h) - therefore one measurement is obtained every 1 - 1.5 days for each decay tank and the radioactive inventory of all tanks and of the whole decontamination plant is thus closely monitored. Half-life times can be determined from the decay curves and together with information from the spectra, radionuclides can be identified and quantified unambiguously.

In addition to the fully automated analyses, wastewater samples can be drawn from the decay tanks. These samples can be measured in a beaker by means of high-resolution γ-ray spectrometry in a separate room. A high-purity germanium detector (HPGe) with 40% relative efficiency and lead shielding in a separate measurement space is used for this purpose. The energy resolution of this detector is 0.95 keV @ 122 keV (Co-57) and 1.9 keV @ 1332 keV (Co-60) [Herpers 1986]. It allows measurements of complex γ-ray spectra containing several radionuclides at very low activity concentrations in the region of 0.1 Bq/L ^{131}I within a short measuring time of about 5 h.

Table 1
Radioecologically relevant nuclides in the decontamination plant due to nuclear medicine procedures and their properties.

Nuclide	Application	Half-life	E_γ [keV]	$E_{\beta(max)}$ (E_α)/E_{IC} [keV]	Possible contaminants, half-life, percentage
Tc-99m	D[a]	6.015 h	140.5	-/120	Mo-99, 66 h, <0.04%
F-18	D[a]	109 min	511[c]	638/-	–
Ga-68	D[b]	67.6 min	511[c]	1899	Ge-68, 270 d, <0.05%
			1077	2921	
I-123	D	13 h	159	1074	–
In-111	D	2.804 d	170	448	–
			245		
I-131	T[a]	8.021 d	364	606	–
			637	333	
			722…		
Lu-177	T[b]	6.734 d	113	498	Lu-177 m, 160 d, <0.05%
			208	385	
Sm-153	T	46.284 h	103	705	Eu-152, 13,5 a <0.01%
				635	Eu-154, 8,6 a, <0.01%
					Eu-155, 4,8 a, <0.01%
					Eu-156, 15,2 d, <0.01%
Re-188	T	17.001 h	155	2120	–
				1965	
P-32	T	14.262 d	pure beta	1711	–
Y-90	T	64.00 h	nearly pure beta	2280	–
Ra-223	T	11.435 d	269.45	5716.23 (α)	–
			154.21	5606.73 (α)	
			323.87	5539.80 (α)	
			144.23	5747.00 (α)	

D: Diagnostic procedures, T: Therapeutic procedures, IC: Internal Conversion.
[a] Frequently used nuclides.
[b] Nuclides with increasing use in the recent years.
[c] Annihilation radiation.

Please cite this article in press as: Sudbrock, F., et al., The effectiveness of wastewater treatment in nuclear medicine: Performance data and radioecological considerations, Journal of Environmental Radioactivity (2016), http://dx.doi.org/10.1016/j.jenvrad.2016.04.002