Journal of Environmental Radioactivity 192 (2018) 187–193

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

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

Presence of artificial radionuclides in samples from potable water and wastewater treatment plants

J. Martínez[a], A. Peñalver[a], T. Baciu[a], M. Artigues[b], M. Danús[c], C. Aguilar[a,*], F. Borrull[a]

[a] Departament de Química Analítica i Química Orgànica, Universitat Rovira i Virgili, Unitat de Radioquímica Ambiental i Sanitaria (URAIS), Consorci d'Aigües de Tarragona (CAT), Carretera Nacional 340, Km. 1094, 43895, L'Ampolla, Tarragona, Spain

[b] Department de Física Mèdica, Spain

[c] Departament de Medicina Nuclear, Hospital Universitari Sant Joan de Reus, Av. del Dr Josep Laporte, 243204, Reus, Spain

ARTICLE INFO

Keywords:
Wastewater treatment plant
Potable water treatment plant
Artificial gamma radionuclides
Sludge
Nuclear power plant
Nuclear medicine

ABSTRACT

Human activity, such as the operation of nuclear power plants (NPPs) and the use of radionuclides in nuclear medicine, results in the presence of artificial radionuclides in surface waters, which may even reach potable water treatment plants (PWTPs) and wastewater treatment plants (WWTPs).

In this study, water and sludge samples from a PWTP are radiologically monitored. The incoming water of the plant is influenced by the presence of an NPP upstream. Two WWTPs receiving wastewater from medical centres and other origins are also studied. As a result, ^{131}I, ^{60}Co and ^{137}Cs have been determined in the dewatered sludge samples from the PWTP, while ^{131}I, ^{99m}Tc, ^{67}Ga and ^{111}In were detected in the sludge samples from the WWTPs. The radionuclide activities in the influent water from the WWTPs studied were lower than the minimum detectable activity values. Therefore, on the basis of our results, the analysis of sludge samples is very useful as it enables the concentration of any radionuclides that may be present in the incoming water.

Lastly, as higher activity of ^{131}I was detected in the samples studied, the total effective dose was assessed for WWTP workers, as they handle dewatered sludge containing this radionuclide. It can be concluded that there is no risk in terms of total exposure.

1. Introduction

The population is exposed to ionizing radiations, of which radiation originating from medical applications is the main artificial source of exposure in the world. In a report by the United Nations Environment Programme (UNEP, 2016), the medical sources of radiation to which humans are exposed are quantified as 20% of the total.

Nuclear medicine involves the administration of radionuclides to patients in order to treat or diagnose diseases. The main and most common medical radionuclides are those shown in Table 1. Gamma-ray emitters with short half-lives are used in diagnostic applications, with ^{99m}Tc being the most widely used in gamma-ray imaging (International Atomic Energy Agency, 2010; Piñero García, 2013). Beta-emitting radionuclides, with a longer half-life than gamma-ray emitters, are used for therapeutic treatments. For example, ^{131}I, which is a betta emitter, is mainly administered as a therapeutic radiopharmaceutical to treat thyroid alterations (Veliscek Carolan et al., 2011).

Besides those of medical origin, radionuclides from nuclear applications, such as nuclear power plants (NPPs), also contribute to population exposure, but to a lesser extent (Sohrabi et al., 2013). One area of concern is the release of radionuclides from NPPs under normal operating conditions or in the event of an accident. The most common radionuclides originating from such waste streams are also presented in Table 1. In this case, they are mainly beta and gamma-ray emitters, usually with longer half-lives than those used in nuclear medicine. They are fission products and some are activation products.

As a result of all medical and NPP discharges, radioactive effluents can reach wastewater treatment plants (WWTPs) and potable water treatment plants (PWTPs). In the case of medical centres, the administered radiopharmaceuticals are incorporated into the patient's body via different routes depending on the examination: intravenously, orally or via inhalation. Depending on the administration route, the excretion pathway differs (Andersson, 2017). As one of the excretion pathways is urine, radionuclides generally reach the sewer system. This may be through direct patient discharges if a diagnostic dose is administered, or after radioactively decaying in hospital waste storage tanks if the patient remains in hospital after therapeutic treatment. During the normal operation of NPPs, very small quantities of certain radionuclides are released via liquid and aerosol discharges and can reach the environment. PWTPs may receive these liquid radioactive

* Corresponding author.
E-mail address: carme.aguilar@urv.cat (C. Aguilar).

https://doi.org/10.1016/j.jenvrad.2018.06.024
Received 19 December 2017; Received in revised form 25 June 2018; Accepted 27 June 2018

Table 1
Physical characteristics of common radionuclides from nuclear medicine and NPPs (Grupen, 2010; Vertés et al., 2011).

Radionuclide	Half-life	Mode of decay	E_γ keV (% intensity)	Origin
^{67}Ga	3.26 d	γ EC, no β^+	93.3 (39) 184.6 (21) 300.2 (16)	Diagnostic
^{99m}Tc	6 h	γ	141 (89)	Diagnostic
^{111}In	2.83 d	γ EC, no β^+	245.5 (94) 171.3 (90)	Diagnostic
^{131}I	8.02 d	γ, β^-	364.5 (81)	Therapeutic and diagnostic. Fission product
^{3}H	12.33 y	β^-		Fission product. Cosmogenic
^{54}Mn	312.19 d	EC, γ	834.8 (99)	Fission product
^{60}Co	5.27 y	β^-,γ	1173.24 (99.85) 1332.50 (99.99)	NPP reactor's steel structures
^{90}Sr	28.80 y	β^-		Fission product
^{137}Cs	30.05 y	β^-, γ	661.66 (85)	Fission product

effluents if the NPP is located upstream in its water catchment area.

Recent studies on this topic conclude that, once radionuclides have entered PWTPs or WWTPs, some of them may associate and concentrate on organic particles settled during the water treatment process (Montaña et al., 2013; Palomo et al., 2007, 2010). In this way, the radiological analysis of sludge samples is extremely useful because it can provide valuable information about the presence of radionuclides in the incoming water samples treated at these plants without needing to analyse them directly. Therefore, sludge samples can be used as a sensitive indicator of the radiological content of the incoming water samples (Sundell-Bergman et al., 2008). With respect to the environmental fate of sludges, they are reused for agricultural purposes, gardening, manufacturing fertilizers and fuel, or they are disposed of in landfill sites. In Spain, the management and agricultural use of the generated sludge is regulated by Royal Decree 1310/1990 (Royal Decree 1310/1990U - Use of sludge from purificiation processes, 1990). This legislation establishes the parametric values of heavy metals and other parameters in the sludge generated in WWTPs. There is also national legislation to protect workers, specifically due to their activity, as well as members of the public (Royal Decree 783/2001, of 6th July, which approves the Regulations on sanitary protection against ionizing radiation, 2001), specifying the limits of radiation doses coming from an artificial or natural source.

Previous studies have radiologically characterized liquid effluents and solid samples from WWTPs (Camacho et al., 2012; Krawczyk et al., 2013; Rose et al., 2012), but there is still limited information related to the fate and behaviour of certain radionuclides in these plants, such as the case of ^{131}I (Cosenza et al., 2015). In view of the above and in order to contribute towards increasing the data in the field, the main objective of this study is to monitor the occurrence of gamma-ray-emitting artificial radionuclides in samples from a PWTP located in L'Ampolla and from two different WWTPs, one located in Reus and the other in Tarragona. All these facilities are influenced by the possible discharge of artificial radionuclides. In the case of the PWTP, which collects water directly from the River Ebre, the main source of the potentially encountered radionuclides is a NPP located upstream from the water treatment plant. For the two WWTPs, the possible radiological influence is the result of the presence of hospitals, the discharges from which may reach these plants. Consequently, it is expected that some of the radionuclides present in the incoming waters of all these plants are concentrated in the sludge generated. In addition, their presence can be detected in the different compartments of the water treatment processes and handling of the sludge generated may pose a potential radiological risk to workers. For this reason, an exposure radiation assessment is also performed.

2. Materials and methods

2.1. Studied area and sample collection

The presence of artificial gamma-ray emitters was evaluated in water, wastewater and sludge samples obtained from a PWTP located in L'Ampolla and from two WWTPs in Reus and Tarragona.

The L'Ampolla PWTP, in the south of Catalonia (Spain), collects its influent water directly from the River Ebre in Campredó, 70 km downstream from an NPP. The water treatment performed in this facility comprises the following steps: (a) the pre-ozonation of the influent water, (b) coagulant treatment with $FeCl_3$, (c) flocculation of the particles in suspension adding PoliDADMAC as the flocculant, (d) flocculation and lamellar decantation, (e) filtration with sand bed, (f) post-ozonation, (g) filtration with granular active carbon and (h) post-chlorination. The average daily water production is 2 m^3/s. For this study, twelve single influent water samples (2 L) were collected from this PWTP, with one sample taken every month throughout 2016 (point 1, Fig. 1a). At the same plant, twelve dewatered sludge samples were collected from the centrifuge (point 2, Fig. 1a). Each sludge sample was a mixture of one single sample collected once a week over the course of a month, throughout 2016.

The Reus WWTP receives wastewater from domestic effluents, as well as the liquid effluents of a leading hospital in the field of nuclear medicine. Most of the treatments performed in the hospital are diagnostic and so low therapeutic doses are administered. Consequently, it is common practice to discharge radioactive effluents directly into the sewage system.

The plant biologically treats activated sludge, performing anaerobic digestion followed by dewatering with a press filter. It has an average daily flow of about 25,000 m^3/day for an equivalent population of 195,833 inhabitants. In short, the water treatment consists of a bar screen and grit chamber, a primary settling stage and a biological treatment followed by a secondary settling stage. All the solids settled in the primary settling are put into a gravity sludge thickener, while those from the secondary settling are put into a flotation sludge thickener. Once extracted from the digester, the sludge passes through a press filter to separate the water from the solids. As shown in Fig. 1b, six different sampling points were considered. Point 1 corresponds to hospital wastewater collected from the sewage outlet pipe of the medical centre. From this sampling point, a total of six samples were taken on three different days, collecting two grab samples per day. As one of our objectives is to study the radioactive profile of the hospital discharges, sampling was performed at 08:00 and 16:00. In the case of the rest of the points (sampling points 2, 3, 4, 5 and 6), these