Manuscript Details

Manuscript number	JENVRAD_2017_904_R2
Title	PRESENCE OF ARTIFICIAL RADIONUCLIDES IN SAMPLES FROM POTABLE WATER AND WASTEWATER TREATMENT PLANTS
Article type	Research Paper

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 waste water 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, 131I, 60Co and 137Cs have been determined in the dewatered sludge samples from the PWTP, while 131I, 99mTc, 67Ga and 111In 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 131I 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.

Keywords	water treatment plant, artificial gamma radionuclides, sludge, nuclear power plant, nuclear medicine
Taxonomy	Inhalation Dose, External Radiation Exposure, Radioactivity in Urban Environment

Corresponding Author	Carme Aguilar
Order of Authors	Joana Martínez, Alejandra Penalver, Tatiana Baciu, Manuel Artigues, Mònica Danús, Carme Aguilar, Francesc Borrull

Suggested reviewers	Paula Rose

Submission Files Included in this PDF

File Name [File Type]

response to the referees_2.docx [Response to Reviewers (without Author Details)]

Manuscript file_J_revised_groc.docx [Revised Manuscript with Changes Marked (without Author Details)]

Highlights.docx [Highlights]

Abstract_v3_groc.docx [Abstract]

Title page_v2n.docx [Title Page (with Author Details)]

Manuscript file_J_revised.docx [Manuscript (without Author Details)]

Figure_1.jpg [Figure]

To view all the submission files, including those not included in the PDF, click on the manuscript title on your EVISE Homepage, then click 'Download zip file'.

Highlights

- ^{131}I, ^{99m}Tc, ^{67}Ga, ^{111}In, ^{60}Co, ^{137}Cs were determined in sludge from WWTP and PWTP.
- It was reported that ^{131}I was present in all the analysed sludge samples.
- The radioisotopes activity in water can be estimated through the values obtained for sludge.
- The exposure to sludge samples by the workers does not constitute any risk for them.

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 waste water 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.

Keywords: water treatment plant, artificial gamma radionuclides, sludge, nuclear power plant, nuclear medicine

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
[c]Departament de Medicina Nuclear
Hospital Universitari Sant Joan de Reus
Av. del Dr Josep Laporte, 2
43204 Reus

*Corresponding author: carme.aguilar@urv.cat

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 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.*, 2010b; Palomo *et al.*, 2007). 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

66 expected that some of the radionuclides present in the incoming waters of all these
67 plants are concentrated in the sludge generated. In addition, their presence can be
68 detected in the different compartments of the water treatment processes and handling of
69 the sludge generated may pose a potential radiological risk to workers. For this reason,
70 an exposure radiation assessment is also performed.

71 ## 2. Materials and methods

72 ### 2.1. Studied area and sample collection

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

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

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

93 The plant biologically treats activated sludge, performing anaerobic digestion followed
94 by dewatering with a press filter. It has an average daily flow of about 25,000 m^3/day
95 for an equivalent population of 195,833 inhabitants. In short, the water treatment
96 consists of a bar screen and grit chamber, a primary settling stage and a biological

97 treatment followed by a secondary settling stage. All the solids settled in the primary
98 settling are put into a gravity sludge thickener, while those from the secondary settling
99 are put into a flotation sludge thickener. Once extracted from the digester, the sludge
100 passes through a press filter to separate the water from the solids. As shown in **Figure**
101 **1b**, six different sampling points were considered. Point 1 corresponds to hospital
102 wastewater collected from the sewage outlet pipe of the medical centre. From this
103 sampling point, a total of six samples were taken on three different days, collecting two
104 grab samples per day. As one of our objectives is to study the radioactive profile of the
105 hospital discharges, sampling was performed at 08:00 and 16:00. In the case of the rest
106 of the points (sampling points 2, 3, 4, 5 and 6), these correspond to samples taken at
107 different points in the WWTP installations. Point 2 corresponds to a wastewater sample
108 from the influent water that has previously flowed through the bar screen and the grit
109 chamber to eliminate the materials with a higher gravity than the organic biodegradable
110 solids. From this sampling point, ten samples were taken, each one being a 24 h
111 composite sample. The other sampling points (3, 4, 5, and 6) correspond to the sludge
112 line. Point 3 and point 4 correspond to the thickened sludge samples. Five samples were
113 taken from primary and secondary thickeners as grab samples, respectively. Five grab
114 samples of digested sludge were also taken (point 5). The samples from sampling points
115 3, 4 and 5 had a dry matter content below 4% (discussed in more detail in Section 3.2)
116 so they were almost liquid. Lastly, at point 6, five dewatered sludge samples from the
117 press filter were also collected. From each sampling point at the WWTP, 2 L of sample
118 was collected in polyethylene bottles between May and August 2016.

119 The incoming water at the Tarragona WWTP includes wastewater from different towns
120 in the area, but is characterized by receiving some industrial effluents and influenced by
121 a medical centre with a nuclear medicine department. As in the case of the Reus
122 WWTP, it biologically treats activated sludge and performs anaerobic digestion. It has
123 an average daily flow of about 35,000 m^3/day for an equivalent population of 175,000
124 inhabitants. The water and the sludge treatment processes are similar to those followed
125 in the other WWTP (Reus). In this plant, five grab samples of dewatered sludge samples
126 were collected after the centrifuge process (point 1, **Figure 1c**). 2 L of sample was
127 collected in polyethylene bottles during August 2016.

128

2.2. Instrumentation

Gamma-ray emitters were measured with a high-resolution germanium detector (HPGe) (model 2020, Canberra Industries, Meriden, USA), equipped with a standard multichannel analyser. The operating conditions were: a voltage of 4,500 V, negative polarity and relative efficiency of 20%. Genie 2000 software (Canberra Industries, Meriden, USA) was used to acquire and analyse the information provided by the gamma-ray spectra. A monoenergetic gamma-ray radionuclide GC2 cocktail (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{54}Mn and ^{113}Sn), covering the energy range of 59.54 to 1,332.49 keV, supplied by CIEMAT (Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas) was used to prepare the different geometries to perform this study for the energy/efficiency calibration which were: a 100 mL polyethylene beaker, a 500 mL Marinelli beaker, a folded plastic film and a filter geometry. The counting efficiency of the detector was determined for each photoelectric peak and for each geometry used in this work in accordance with the specifications of the Spanish Standard UNE-EN ISO 10703 (AENOR, 2016). For example, the efficiency calibration of ^{137}Cs (661.7 KeV) is 0.0128, 0.0125, 0.0269 and 0.0338 for the 100 mL polyethylene beaker, 500 mL Marinelli beaker, folded plastic film and filter geometries, respectively.

2.3. Sample preparation and gamma-ray spectrometry measurement

Different methodologies for sample preparation were carried out depending on the type of sample. In the case of water from the River Ebre, samples were concentrated using an evaporation method (Gilfillan and Timmers, 2012; Korun, 2008; Krawczyk *et al.*, 2013; Rose *et al.*, 2013), consisting of pouring 2 L of sample on a tray previously covered with plastic film, before being dried at 70ºC in an oven. The film containing the dry residue was then folded and stored in a Petri box, which was measured for 144,000 s (72,000 s for each side of the Petri box).

To identify and quantify artificial radionuclides with a short half-life, the wastewater samples from the hospital sewage outlet pipe and the Reus WWTP influent were processed immediately after arrival at the laboratory. They were placed in a 500 mL Marinelli beaker and acidified with HCl 37% to pH 2 and then measured by gamma-ray spectrometry. ^{131}I is generally present in water samples at environmentally low activity levels (Jiménez *et al.*, 2011b), so a preconcentration step was applied to the samples.

This procedure consisted of vacuum filtering 1 L of each sample using glass fibre and cellulose filters. Once filtered, the samples were then acidified with HCl 37% to pH < 2. Next, the procedure used for ^{131}I precipitation described by Baeza *et al.* (Baeza *et al.*, 2004) was applied, which basically consists of two steps. The first involves the addition of 10 mL of KI at a concentration of 1.3 g/L, 0.5 g of Na_2SO_3, to reduce iodine to iodide, and 5 mL of H_2SO_4 conc. Then, 10 mL of $AgNO_3$ at a concentration of 3.2 g/L and 10 mL of H_2O_2 at a concentration of 0.3 g/L are added to obtain an Ag^+ excess in the media and eliminate $S_2O_3^{2-}$, respectively. This pretreatment is followed by the procedure established by ISO 10703:1997 (AENOR, 2016) in which 4M NaOH is used to increase the pH to 9. Once the ^{131}I is precipitated, it is collected onto eight glass-fibre filters and stored in a 5 cm i.d. Petri box and then measured by gamma-ray spectrometry. The counting time was 144,000 s (72,000 s for each side of the Petri box).

In the case of the samples from the sampling points 3, 4 and 5 (corresponding to primary, secondary and digested sludge samples from the Reus WWTP), gamma-ray spectrometry measurement was performed as soon as they arrived at the laboratory. Each sample was placed in a 500 mL Marinelli beaker, treated with formaldehyde (to prevent bacteriological growth), homogenized and then measured directly.

Lastly, dewatered sludge samples from the Reus WWTP filter press, Tarragona WWTP centrifuge and L'Ampolla PWTP centrifuge were dehydrated in accordance with the method for soil samples described by Camacho *et al.* (Camacho *et al.*, 2012). Wet samples were dried at 100ºC, then crushed in a ball mill and sifted using a 250 μm sieve for homogenization. The samples were measured by gamma-ray spectrometry using a 100 mL polyethylene beaker in the case of WWTP samples and a 500 mL Marinelli beaker in the case of PWTP sludge.

The sludge samples had different water content as well as organic matter. For this reason, the procedures described in Standard Methods (APHA, 2012) focusing on the determination of total solids (TS) and volatile solids (VS) were performed on each sludge sample. Briefly, the TS procedure consists of evaporating the sample to dryness at 105ºC for 1 h. In order to determine VS content, the dry residue obtained from TS procedure was then heated in a muffle furnace to 550ºC for 1 h. In both procedures, once the samples were at room temperature, they were weighed and then the corresponding calculation was performed.

2.4. Gamma-ray spectrometry measurement validation

In order to validate the measurement under these conditions, a tap water sample taken from Seibersdorf (Austria) from an intercomparison exercise conducted in 2017 by the International Atomic Energy Agency (IAEA) was spiked onto eight fibre-glass filters. This sample contains ^{133}Ba (the gamma-ray emission of which is 356.02 keV, similar to that of ^{131}I, 364.50 keV) and ^{152}Eu, with certified activities of 0.074 Bq/filter and 0.075 Bq/filter, respectively. The Z-score value obtained following our method was below one in both cases, with a relative bias of 2.2% and 2.3%, respectively.

^{99m}Tc was also determined in water samples from hospital sewage and the measurement was validated using a tap water sample from Seibersdorf, spiked with a mixture of fresh fission products (^{95}Zr, ^{99m}Tc, ^{99}Mo, ^{103}Ru, ^{132}I, ^{140}Ba, ^{141}Ce, ^{143}Ce, ^{144}Ce, ^{147}Nd, ^{239}Np) (IAEA, 2017). The ^{99m}Tc certified activity value was 53.8 ± 2.0 Bq/L. In this case, the Z-score value was around zero, with a relative bias of 5.4%.

Altogether, the results from the intercomparison of sample measurements indicate that our procedure is robust and satisfactory.

3. Results and discussion

3.1. Activity levels in PWTP samples

As mentioned above, the influent water from the L'Ampolla PWTP is influenced by the presence of an NPP upstream on the River Ebre, as well as from effluent from municipal WWTPs discharged along the river.

The Program of Environmental Radiological Monitoring (PVRA) conducted by the Nuclear Security Council in Spain (CSN) publishes annual figures on the effluents discharged into the environment by NPPs. The incoming water of the evaluated PWTP is supplied as drinking water after treatment. Therefore, the radiological quality of the River Ebre water should be monitored as it is influenced by radioactive effluents from the NPP. In previous studies focusing on the PWTP in L'Ampolla, such as the work by Palomo *et al.* (Palomo *et al.*, 2007), tritium was quantified in almost all the samples measured. The same authors obtained tritium activity values below the minimum detectable activity (MDA < 0.6 Bq/L) in water from rivers not affected by the presence

of an NPP. This radionuclide has a cosmogenic origin and it is also released from the NPP. This beta-emitting radionuclide has also been detected in two samples of our study, with an activity of 3.3 Bq/L and 43.4 Bq/L, suggesting the influence of the NPP on the influent water of the L'Ampolla PWTP. The tritium activity of the remaining samples was below the MDA value, which is 2 Bq/L. However, it was not possible to detect the presence of any artificial gamma-ray-emitting radionuclide, despite being preconcentrated (MDA showed in **Table 2**). This is probably due to the low activity levels at which these radionuclides are present in the evaluated samples, or larger sample volumes and longer counting time are required.

Artificial radionuclides were also determined in dewatered sludge from the same PWTP, which is influenced by the presence of a NPP upstream on the River Ebre, as it has been previously confirmed by some studies performed by our research group. In particular, different fission and activation products were found, such as ^{54}Mn, ^{58}Co, ^{60}Co, ^{110m}Ag and ^{137}Cs (Palomo *et al.*, 2010b), ^{90}Sr (Mola *et al.*, 2014) and ^{55}Fe and ^{63}Ni (Fonollosa *et al.*, 2016) in PWTP sludge. In the present study, ^{60}Co and ^{137}Cs could be quantified with activity levels ranging from 1.5 - 12.3 Bq/kg$_{dw}$ (5.5 Bq/kg$_{dw}$ average activity) and 0.4 - 1.1 Bq/kg$_{dw}$ (0.6 Bq/kg$_{dw}$ average activity), respectively. Moreover, in some of the analysed samples, ^{131}I was also found. The maximum activity level determined was 1.5 Bq/kg$_{dw}$. The possible origin of this radionuclide is not clear. As previously discussed, the River Ebre is influenced by discharges from the NPP located upstream. According to the CSN, the annual ^{131}I activity released from liquid and gaseous effluents of Spanish NPPs is around 1-10 MBq/y (Luque *et al.*, 2015). In view of these discharges, the presence of this radionuclide in dewatered sludge is expected on a monthly basis. However, the variations in the activity levels may suggest additional anthropogenic contributions, probably due to discharges from outpatients treated with ^{131}I living in the area (Connan *et al.*, 2008). Usually, a patient is administered a 296 MBq dose of ^{131}I for diagnostic purposes and the dose for treatment ranges from 555 MBq (outpatient) to 7,400 MBq. Annually, in Spain, the activity used for these treatments is 44.40 Gb/y for diagnostic purposes and ranges between 69.38 Gb/y (outpatient) and 740,000 Gb/y for therapeutic purposes (SEFM, 2002), which are higher than the annual activity discharged by a Spanish NPP under normal conditions.

3.2. Activity levels in sewage and WWTP samples

In the case of hospital wastewater (point 1, **Figure 1b**), the results are shown in **Table 3**. As shown in the table, two radionuclides commonly used in nuclear medicine were quantified at almost all the analysed samples: ^{99m}Tc, which is the most common radionuclide used for single photon emission computed tomography studies, and ^{67}Ga, which is less commonly used in the same diagnostic treatment (Qaim, 2017). In general, higher activity values were observed for ^{99m}Tc than for ^{67}Ga. The obtained values for ^{99m}Tc suggest a radioactive profile of the hospital discharges, as sampling was performed at different times on the same day. Based on the data provided by the medical staff (personal communication, March 30th, 2018), diagnostic treatments with ^{99m}Tc were mainly performed in the morning, although, on some occasions, patients were also treated in the afternoon. However, in the case of ^{67}Ga, the activity levels remain the same at the different times evaluated. On the sampling days, the hospital did not perform any diagnostic treatment with this radionuclide. However, some days before, a patient was treated with ^{67}Ga to diagnose an inflammatory lesion. Due to their disease, the patient remained in the hospital for several days. Therefore, the possible origin of ^{67}Ga in the analysed samples could be attributed to the excretion via that patient's urine. With respect to ^{131}I, it is administered at low doses to patients who require therapeutic treatments. They do not therefore remain in hospital, so the excretion of this radionuclide mainly occurs away from the hospital. This explains why ^{131}I was not detected in the hospital wastewater.

The hospital sewage enters the public sewage system and then reaches the Reus WWTP (point 2, **Figure 1b**). For this reason, it was also necessary to determine the artificial gamma-ray radionuclides in the influent wastewater of this plant. The possible radioisotopes present in this sample may come from the hospital sewage but also from outpatients. Of the ten samples collected at this sampling point, ^{131}I was the only radionuclide detected in two of them, with activities of 0.07 ± 0.01 Bq/L and 0.39 ± 0.07 Bq/L. For the other samples, ^{131}I was not detected, with an MDA < 0.004 Bq/L. No other radioisotopes were detected. This can be explained by the dilution that the hospital sewage undergoes when mixed with domestic discharges before entering the WWTP, and also by the short half-life of the isotopes used for medical purposes.

The results obtained in this work are in line with studies by other authors who have analysed hospital effluents and WWTP influents (Chang *et al.*, 2011; Cosenza *et al.*, 2015; Jiménez *et al.*, 2011b; Krawczyk *et al.*, 2013). For example, Jiménez *et al.* (Jiménez *et al.*, 2011b) detected the presence of ^{131}I in influent WWTP samples using a precipitation method followed by liquid scintillation counting (LSC). The maximum ^{131}I activity found in the influents was 2.6 Bq/kg. In another study performed by Krawczyk *et al.* (Krawczyk *et al.*, 2013), ^{99m}Tc, ^{67}Ga and ^{131}I were detected using gamma-ray spectrometry in three hospital effluents from Spain (Andalucía). ^{99m}Tc activity levels ranged from 148.7 to 2,510 Bq/L and this radionuclide was found in all hospital effluents, while ^{131}I and ^{67}Ga were only found occasionally, with activity levels of 59.29 and 17.28 Bq/l, respectively.

In the case of sewage sludge samples, ^{131}I, ^{99m}Tc, ^{67}Ga and ^{111}In were quantified in both WWTPs evaluated in this study. The activity values are shown in **Table 4**. In the case of the Reus WWTP, different types of sludge samples resulting from the sewage treatment process were analysed. Higher activities of ^{131}I were obtained for the secondary sludge than for the primary sludge. This can be justified because, even though the residence time is longer, sludge is recirculated to the aerobic tank and is in continuous contact with influent water. Moreover, during the digestion process with anaerobic microorganisms, the organic matter present is partially eliminated as new sludge is continuously added to the digester tank with a residence time of 21 days. For this reason, the ^{131}I activity recorded in the digested sludge is lower than in the secondary sludge. With respect to the diagnostic radionuclides, as they were detected in hospital sewage samples, there were expected to be determined in the sludge samples. ^{99m}Tc could be found intermittently in primary, secondary and digested sludge samples at levels of 119.6 Bq/L, 2.6 Bq/L and 3.4 Bq/L, respectively. The heterogeneity of the measured values is probably due to the fact that the medical administration of this radionuclide depends on the frequency and the number of patients receiving diagnostic treatments. Moreover, decay takes place during WWTP processes and this clearly influences its presence. ^{67}Ga and ^{111}In were occasionally quantified at low activity levels and this is logical considering that, of all the medical radionuclides found, these are used to a lesser extent.

Dewatered sludge is the most widely studied sample from PWTPs and WWTPs in the literature (Cosenza *et al.*, 2015; Montaña *et al.*, 2013; Ortiz *et al.*, 2004) because its use

as fertilizer or its incineration may pose a risk to the population. Therefore, this kind of samples was analysed, as the evaluated WWTPs are both influenced by medical centres. In our samples, ^{131}I was detected at average values of 192.35 Bq/kg_{dw} in Reus and 13.13 Bq/kg_{dw} in Tarragona. The higher activity values at the Reus WWTP may be attributed to the fact that the Tarragona WWTP is mainly influenced by domestic and industrial wastewaters and, to a lesser extent, by a medical centre. It is suspected that ^{131}I therapeutic treatments are performed less frequently here. Occasionally, ^{67}Ga was detected in the dewatered sludge from Reus and Tarragona, while ^{111}In was only determined in Tarragona (**Table 4**).

^{131}I was found in all sludge samples from the WWTP compartments. In the case of the Reus WWTP, ^{131}I activity seems to follow the pattern: point 3 (Bq/L) < point 5 (Bq/L) < point 4 (Bq/L) (**Table 4**). This may be related to the content of the volatile solids that corresponds to the organic particles present in the sample, as stated by Cosenza *et al.* (Cosenza *et al.*, 2015), who also explained that the highest organic matter content corresponds to the highest ^{131}I activity because this is mainly associated with it. In our case, the VS values of all the sludge samples were as follows. 1 kg of digested sludge (point 5) was composed of 56 g of dry solids, 60% of which were VS. This sample displayed the lowest % VS because, during the digestion process, which takes place under anaerobic conditions, organic material is transformed into CO_2 and CH_4 (Appels *et al.*, 2008). The VS content of primary (point 3) and secondary (point 4) sludge was 70% and 81%, respectively, but the TS content was too low. For this reason, although the VS content was higher at points 3 and 4 than at points 5 and 6, these samples have the lowest ^{131}I activity. Digested sludge (point 5) displayed lower ^{131}I activity than secondary sludge (point 4). Digested sludge is held in a digester tank which is continuously fed with a mix of primary and secondary sludge. Consequently, digested sludge is continuously removed. The sludge remains in the digester for 21 days and decay takes place. Secondary sludge has a shorter retention time but decay is also taking place. This sample recirculates from the secondary settling tank to the aerobic tank, which can enrich the sludge in terms of ^{131}I.

High levels of ^{131}I were been detected in dewatered sludge samples. This sample was obtained after the dewatering process on digested sludge performed over a short period of time. For this reason, it could be said that the digested sludge and dewatered sludge are the same sample differentiated by their humidity content, which influences the

radionuclide activity concentration. Both samples were dry and analysed by gamma-ray spectrometry and the same ^{131}I activity was determined.

It has been shown that sludge acts as a concentrator of some of the radionuclides present in the incoming water of the PWTP and WWTP (Camacho *et al.*, 2012; Fonollosa *et al.*, 2016, 2014; Montaña *et al.*, 2013) and this was observed in the present work. Sludge samples can be used as a source to estimate the theoretical activity concentration in water. To illustrate this, a simple calculation can be performed to determine the activity levels in the water samples. 1 L of influent water of the Reus WWTP generates 1 g of dewatered sludge and 1 L of influent water from the L'Ampolla PWTP generates 0.014 g of dewatered sludge. The data are provided by the technical staff from those facilities (personal communication, November 30th, 2017). Based on this technical information, the ^{131}I activity in the water samples can be estimated. In the case of the Reus WWTP and L'Ampolla PWTP, the estimated activity values are 561.8 mBq/L and 0.02 mBq/L, respectively. These values relate to the activity level in the sludge samples from the Reus WWTP and L'Ampolla PWTP (213.9 Bq/kg_{dw} and 1.5 Bq/kg_{dw}, respectively). Therefore, using obtained activity values from sludge samples to estimate the activity in water is a useful strategy because it avoids working and handling high volume of samples, which is time-consuming. In this respect, Connan *et al.* (Connan *et al.*, 2008) quantified ^{131}I in river water in the range of 0.4 to 11.0 Bq/L. However, in order to be able to obtain these activity values, 100 L of river water was used.

3.3. Exposure radiation assessment

The presence of artificial radionuclides in PWTP and WWTP sludge samples is a concern because workers are exposed to them when handling this kind of samples. For this reason, it is important to assess the potential risk of radiological impact due to long term exposure for the workers. Although different radionuclides have been found in the sludge samples, our focus was the exposure due to the presence of ^{131}I in dewatered sludge from the WWTP because the obtained activity levels for this isotope were noticeably higher than the others and it was also present in all the analysed samples. Two possible exposure pathways for workers are inhalation due to resuspended sludge dust and external radiation due to proximity to the sample.

Using the ICRP 60 1990 formula, the effective dose from inhalation of ^{131}I was calculated:

$$E_{131I,\,inh} = h_{131I,\,inh} * J_{131I,\,inh} \quad (1)$$

Where $J_{131I,inh}$ is the inhalation intake activity in Bq, using a ^{131}I activity equal to 213.9 $Bq/kg_{dw,}$ which is the highest concentration of this radioisotope determined in dewatered sewage sludge in this study. $h_{131I,inh}$ is the committed effective dose per unit intake for inhaled ^{131}I equal to $1.10 \cdot 10^{-8}$ Sv/Bq (value published in Directive 96/29/Euratom (Council of the European Union, 1996)). According to Jiménez *et al.* (Jiménez *et al.*, 2011a), a conservative value for the yearly amount of dust that a worker inhaled was estimated to be 9,000 mg/y. The effective dose from inhalation is $2.12 \cdot 10^{-5}$ mSv/y, which is lower than 1 mSv/y (the exposure limit for unexposed workers).

The total effective dose from external radiation as a consequence of the ^{131}I sludge content is calculated using equation (2) from the work mentioned above.

$$E_{ext} = \frac{\Gamma_{131I} \cdot A \cdot f}{d^2} \quad (2)$$

Where Γ_{131I} = 52.20 μGy·m²/GBq·h is the ^{131}I specific gamma-ray constant (Ninkovic and Adrovic, 2005). A = 1,46 MBq is ^{131}I activity of the daily dry sewage sludge produced, with the average daily production being 25,000 kg. It is calculated considering a maximum concentration of 213.9 Bq/kg_{dw} of ^{131}I and taking into account its humidity content. f = 2,000 h per year is the conservative annual exposure time (equivalent to the total number of working hours per year) and d = 1 m is the distance in metres between the radioactive source and the worker. The total effective dose from external radiation is $1.52 \cdot 10^{-2}$ mSv/y, which is below the dose limit for the general population (1 mSv/y).

The total dose to which a WWTP worker is exposed is obtained by adding up the previously calculated doses (effective dose from inhalation plus total effective dose from external radiation), giving a total of $1.53 \cdot 10^{-2}$ mSv/y. This calculation estimates the total dose exposure in the worst-case scenario, assuming that a WWTP worker is dealing 2,000 h per year the highest ^{131}I activity determined in dewatered sludge. It can therefore be concluded that there is no risk to WWTP workers.

4. Conclusions

The detection of artificial radionuclides in water samples is difficult due to their low concentration, as waste effluents are diluted. However, evaporating and performing a measurement on the dry residue significantly reduced the MDA compared to measurements performed using a 500 mL Marinelli beaker. Artificial radionuclides

were detected in water samples collected at the sampling points at which the sample was not mixed with sewage. Further studies should conduct longer collection schemes in order to perform this radioisotope detection procedure on discrete hospital discharges. The detection of some radionuclides in sludge samples is feasible because of the accumulation capacity of the matrix. In particular, ^{131}I was detected in all sludge samples, with dewatered sludge displaying the highest activity. Moreover, activity levels in water could theoretically be estimated through the analysis of the corresponding sludge sample, thereby simplifying the sample pretreatment procedures. Although ^{131}I activity levels were found in sludge and water samples, the dewatered sludge may be used for agricultural purposes and the WWTP effluent is suitable for irrigating fields due to the short half-life of this radioisotope. According to the exposure radiation assessment performed, it can be concluded that, from a radiological protection point of view, there is no risk to WWTP workers in terms of total exposure and that the main contribution to the dose to which workers are exposed is via the external exposure pathway.

Acknowledgements

The authors are grateful for the support of the Consorci d'Aigües de Tarragona, Aigües de Reus and EMATSA for providing the different samples from the L'Ampolla PWTP, Reus WWTP and Tarragona WWTP.

References

AENOR, 2016. Water quality. Determination of the activity concentration of radionuclides. Method by high resolution gamma-ray spectrometry. (ISO 10703:2007). Madrid.

Andersson, M., 2017. Radiation dose to patients in diagnostic nuclear medicine. Implementation of improved anatomical and biokinetic models for assessment of organ absorbed dose and effective dose. Lund University, Faculty of Medicine.

APHA, 2012. Standard Methods For the Examinaton of Water and Wastewater, 22nd Ed. APHA, AWWA, WEF, Washington, DC.

Appels, L., Baeyens, J., Degrève, J., Dewil, R., 2008. Principles and potential of the anaerobic digestion of waste-activated sludge. Prog. Energy Combust. Sci. 34, 755–781. doi:10.1016/j.pecs.2008.06.002

Baeza, A., Miró, C., Soleto, C., 2004. Spectrometric determination of low activities of gamma emitters in water samples. Appl. Radiat. Isot. Isot. 61, 203–6. doi:10.1016/j.apradiso.2004.03.046

Camacho, A., Montaña, M., Vallés, I., Devesa, R., Céspedes-Sánchez, R., Serrano, I., 2012. Temporal evolution of radionuclides in sludge from wastewater treatment plants. J. Radioanal. Nucl. Chem. 297–306. doi:10.1007/s10967-012-1977-6

Chang, B.U., Choi, S.W., Song, M.H., Lee, J.S., Kim, Y., 2011. Medically used radionuclides (^{131}I, ^{99m}Tc) in the urban sewage system: The case of the Daejeon metropolitan city, Korea. Radiat. Prot. Dosimetry 146, 318–321. doi:10.1093/rpd/ncr108

Connan, O., Tessier, E., Maro, D., Amouroux, D., Hébert, D., Rozet, M., Voiseux, C., Solier, L., 2008. Water to atmosphere fluxes of ^{131}I in relation with alkyl-iodide compounds from the Seine Estuary (France). J. Environ. Radioact. 99, 1102–1110. doi:10.1016/j.jenvrad.2008.01.001

Cosenza, A., Rizzo, S., Sansone Santamaria, A., Viviani, G., 2015. Radionuclides in wastewater treatment plants: monitoring of Sicilian plants. Water Sci. Technol. 71, 252–258. doi:10.2166/wst.2014.501

Council of the European Union, 1996. Council Directive 96/29/Euratom of 13th May 1996 laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation. Official Journal of the European Communities. Brussels, 13th May 1996.

Fonollosa, E., Nieto, A., Peñalver, A., Aguilar, C., Borrull, F., 2014. Presence of radionuclides in sludge from conventional drinking water treatment plants. A review. J. Environ. Radioact. 141C, 24–31. doi:10.1016/j.jenvrad.2014.11.017

Fonollosa, E., Nieto, A., Peñalver, A., Borrull, F., Aguilar, C., 2016. Determination of artificial beta-emitters in sludge samples. J. Radioanal. Nucl. Chem. 309, 1077–1085. doi:10.1007/s10967-016-4705-9

Gilfillan, N.R., Timmers, H., 2012. Detection and tracing of the medical radioisotope ^{131}I in the Canberra environment. EPJ Web Conf. 35, 4002. doi:10.1051/epjconf/20123504002

Grupen, C., 2010. Introduction to Radiation Protection, CEUR Workshop Proceedings. Springer Berlin Heidelberg. doi:10.1007/978-3-642-02586-0

International Atomic Energy Agency, 2010. Technetium-99m Radiopharmaceuticals: Status and Trends, IAEA Radioisotopes and Radiopharmaceuticals Series No. 1. Vienna.

Jiménez, F., Debán, L., Pardo, R., López, R., García-Talavera, M., 2011a. Levels of ^{131}I and six natural radionuclides in sludge from the sewage treatment plant of Valladolid, Spain. Water. Air. Soil Pollut. 217, 515–521. doi:10.1007/s11270-010-0605-8

Jiménez, F., López, R., Pardo, R., Debán, L., García-Talavera, M., 2011b. The determination and monitoring of ^{131}I activity in sewage treatment plants based on A2/O processes. Radiat. Meas. 46, 104–108. doi:10.1016/j.radmeas.2010.07.030

Korun, M., 2008. Optimization of sampling and counting times for gamma-ray spectrometric measurements of short-lived gamma-ray emitters in aqueous samples. Appl. Radiat. Isot. 66, 841–844. doi:10.1016/j.apradiso.2008.02.024

Krawczyk, E., Piñero-García, F., Ferro-García, M.A., 2013. Discharges of nuclear medicine radioisotopes in Spanish hospitals. J. Environ. Radioact. 116, 93–8. doi:10.1016/j.jenvrad.2012.08.011

Luque, S., Marugán, I., Rey, R., Ramos, L., 2015. Programas de vigilancia radiológica ambiental, Resultados 2014, in: CSN (Ed.), Colección Informes Técnicos 47.2015. Madrid, pp. 3–172.

Mola, M., Avivar, J., Nieto, A., Peñalver, A., Aguilar, C., Ferrer, L., Cerdà, V., Borrull, F., 2014. Determination of ^{90}Sr and ^{210}Pb in sludge samples using a LOV-MSFIA system and liquid scintillation counting. Appl. Radiat. Isot. 86, 28–35. doi:10.1016/j.apradiso.2013.11.123

Montaña, M., Camacho, A., Devesa, R., Vallés, I., Céspedes, R., Serrano, I., Blàzquez,

S., Barjola, V., 2013. The presence of radionuclides in wastewater treatment plants in Spain and their effect on human health. J. Clean. Prod. 60, 77–82. doi:10.1016/j.jclepro.2011.07.007

Ninkovic, M.M., Adrovic, F., 2005. Chapter 1. Air Kerma Rate Constants for Nuclides Important to Gamma Ray Dosimetry and Practical Application, in: Gamma Radiation. pp. 3–16. doi:10.5772/39170

Ortiz, J., Ballesteros, L., Zarza, I., Serradell, V., 2004. Radioactivity study in a Sewage Treatment Plant (STP). Radiological Impact Evaluation. IRPA Congress 5G (1).

Palomo, M., Peñalver, A., Aguilar, C., Borrull, F., 2007. Tritium activity levels in environmental water samples from different origins. Appl. Radiat. Isot. 65, 1048–1056. doi:10.1016/j.apradiso.2007.03.013

Palomo, M., Peñalver, A., Aguilar, C., Borrull, F., 2010. Impact of industries in the accumulation of radionuclides in the lower part of Ebro river (Catalonia, Spain). Radioprotection 45, 459–475. doi:10.1051/radiopro/2010032

Piñero García, F., 2013. Estudio radiológico ambiental en el sur-este de la Península Ibérica. Universidad de Granada.

Qaim, S.M., 2017. Nuclear data for production and medical application of radionuclides: Present status and future needs. Nucl. Med. Biol. 44, 31–49. doi:10.1016/j.nucmedbio.2016.08.016

Rose, P.S., Smith, J.P., Cochran, J.K., Aller, R.C., Swanson, R.L., 2013. Behavior of medically-derived 131I in the tidal Potomac River. Sci. Total Environ. 452–453, 87–97. doi:10.1016/j.scitotenv.2013.01.055

Rose, P.S., Swanson, R.L., Cochran, J.K., 2012. Medically-derived ^{131}I in municipal sewage effluent. Water Res. 46, 5663–71. doi:10.1016/j.watres.2012.07.045

Royal Decree 1310/1990U - Use of sludge from purificiation processes, 1990, Official State Gazette 262. Madrid, 29th October 1990.

Royal Decree 783/2001, of 6th July, which approves the Regulations on sanitary protection against ionizing radiation, 2001, Official State Gazette 178. Madrid, 6th July 2001.

SEFM, 2002. Grupo de efluentes del foro de protección radiológica en el medio hospitalario. Informe de actividades [WWW Document]. URL www.sefm.es/docs/actsefm/informefinalgrupoefluentes.pdf

Sohrabi, M., Parsouzi, Z., Amrollahi, R., Khamooshy, C., Ghasemi, M., 2013. Public exposure from environmental release of radioactive material under normal operation of unit-1 Bushehr nuclear power plant. Ann. Nucl. Energy 55, 351–358. doi:10.1016/j.anucene.2012.12.002

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. J. Environ. Radioact. 99, 1572–1577. doi:10.1016/j.jenvrad.2007.12.005

UNEP, 2016. Radiation Effects and Sources: What is Radiation? What does Radiation do to us? Where does Radiation come from? New York. doi:http://dx.doi.org/10.18356/b1749f17-en

Veliscek Carolan, J., 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. J. Environ. Radioact. 102, 953–963. doi:10.1016/j.jenvrad.2009.10.002

Vertés, A., Sándor, N., Klencsár, Z., Lovas, R., Rösch, F. (Eds.), 2011. Handbook of Nuclear Chemistry, Second Ed. Springer.

562 **Table 1** Physical characteristics of common radionuclides from nuclear medicine and NPPs
563 (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

564

565

566 **Table 2** MDA from all the gamma-ray geometries used and their respective counting time

Radionuclide	Gamma-ray calibration geometries			
	100 mL beaker [a] [Bq/kg]	500 mL beaker [a] [Bq/L]	Folded plastic film [b] [Bq/L]	Filter [b] [Bq/L]
^{60}Co	1.200	0.250	0.010	0.005
^{67}Ga	1.500	0.510	0.100	0.005
^{99m}Tc	1.800	0.200	0.040	0.002
^{111}In	0.800	0.150	0.040	0.003
^{131}I	1.100	0.200	0.060	0.004
^{137}Cs	1.200	0.250	0.080	0.004

567 [a] Counting time: 72,000 s

568 [b] Counting time: 144,000 s

569

570 **Table 3** ^{99m}Tc and ^{67}Ga activities in daily integrated hospital sewage effluent samples (Bq/L)

Day	Sampling collection time	^{99m}Tc	^{67}Ga
1	08:00	11.7 ± 1.1	n. d.[a]
	16:00	0.9 ± 0.6	n. d.[a]
2	08:00	1268.5 ± 42.0	10.0 ± 0.6
	16:00	70.3 ± 2.4	16.9 ± 0.6
3	08:00	1738.9 ± 55.7	2.1 ± 0.6
	16:00	34.8 ± 2.7	16.4 ± 0.3

571 n.d. not detected

572 [a] ^{67}Ga MDA <0.51 Bq/L

Table 4 Gamma-ray emitting radionuclide activity in sludge from Reus and Tarragona WWTPs.

Isotopes	**Reus WWTP**				**Tarragona WWTP**
	Primary sludge [Bq/L]	**Secondary sludge [Bq/L]**	**Digested sludge [Bq/L]**	**Dewatered sludge [Bq/kg_{dw}]**	**Dewatered sludge [Bq/kg_{dw}]**
^{131}I	0.5 - 4.2 (1.9)	4.2 - 28.7 (18.9)	2.1 – 7.0 (5.3)	176.5 – 213.9 (192.4)	12.9 – 13.4 (13.1)
^{99m}Tc	119.6 ± 3.7*	2.6 ± 0.3*	3.4 ± 0.2*		
^{67}Ga	2.9 ± 0.4*	0.8 ± 0.2*	0.8 ± 0.3*	20.4 ± 2.0*	11.4 ± 0.5*
^{111}In			0.20 ± 0.03*		3.3 ± 1.1*
^{60}Co					
^{137}Cs					
Sludge residence time	6 - 24 h	6 - 24 h	21 d	21 d	21 d
TS (%)	4	3	1	17	
VS (%)	70	81	60	65	

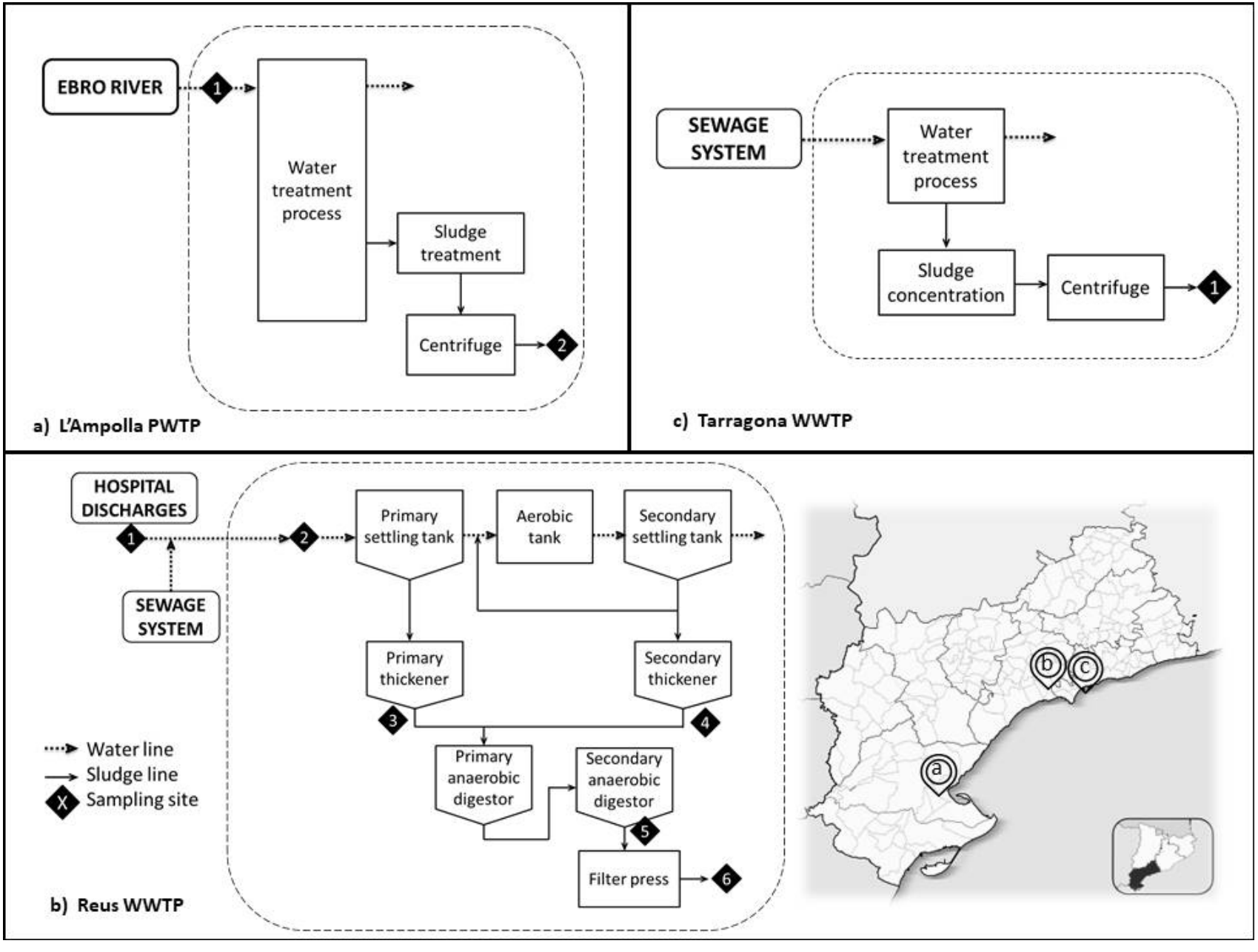
Activity expressed as minimum – maximum. Average indicated in parentheses

*Activity found in single samples. The figures refer to expanded uncertainty (k=2)

573 **FIGURE CAPTION**

574 **Fig. 1** Schematic representation of the different sampling collection points in a) the L'Ampolla
575 PTWP; b) the Reus WWTP; c) the Tarragona WWTP.

576


EBRO RIVER
1
Water treatment process
Sludge treatment
Centrifuge
2
a) L'Ampolla PWTP
SEWAGE SYSTEM
Water treatment process
Sludge concentration
Centrifuge
1
c) Tarragona WWTP
HOSPITAL DISCHARGES
1
2
SEWAGE SYSTEM
Primary settling tank
Aerobic tank
Secondary settling tank
Primary thickener
Secondary thickener
3
4
Primary anaerobic digestor
Secondary anaerobic digestor
5
Filter press
6
Water line
Sludge line
Sampling site
b) Reus WWTP
a
b
c