



Revista de Gestão Costeira Integrada -
Journal of Integrated Coastal Zone
Management

E-ISSN: 1646-8872

rgci.editor@gmail.com

Associação Portuguesa dos Recursos
Hídricos

Malta, Margarida; Oliveira, João M.; Silva, Lúcia; Carvalho, Fernando P.
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Revista de Gestão Costeira Integrada - Journal of Integrated Coastal Zone Management,
vol. 13, núm. 4, 2013, pp. 399-408
Associação Portuguesa dos Recursos Hídricos
Lisboa, Portugal

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Radioactivity from Lisboa urban wastewater discharges in the Tejo River Estuary *

Radioactividade no estuário do Tejo proveniente das descargas de águas residuais de Lisboa

Margarida Malta ¹, João M. Oliveira ¹, Lídia Silva ¹, Fernando P. Carvalho ^{@,1}

ABSTRACT

Sediments, water, mussels and fish were collected monthly during a 19 month period at eight sampling stations along the North bank of the Tejo estuary (Lisboa, Portugal). Fresh samples were immediately analysed for short lived gamma emitting radionuclides. Amongst these, ¹³¹I and ^{99m}Tc were detected in most samples of mussels (*Mytilus galloprovincialis*) and mullets (*Liza ramada*). Spatial distribution of samples containing these radionuclides consistently indicated contamination at Alcântara and Praça do Comércio, and, on occasion, at Chelas, all sites inside the estuary. Round-the-year, generally there was no contamination at Algés, near the estuary mouth, and Beirolas, upstream by the Vasco da Gama Bridge. The estuarine areas found contaminated by radionuclides correspond to the areas receiving urban wastewater discharges from the city of Lisboa. The artificial radionuclides ¹³¹I and ^{99m}Tc are used in nuclear medicine practices and originate from hospital liquid effluent discharges into the city sewage system. Their presence near Praça do Comércio was related to the nearby discharges of a main sewer discharging untreated urban wastewater. Near Alcântara, the sewer outlet discharged treated urban wastewater from the Alcântara wastewater treatment plant and the presence of short-lived radionuclides indicated that sewage treatment and the time delay introduced before discharge of the treated effluent were not sufficient to allow for radioactive decay of those radionuclides. The highest concentration values of ¹³¹I and ^{99m}Tc determined in mussels and fish were 12±2 and 136±20 Bq kg⁻¹ wet weight (w.w.), respectively. These concentrations did not exceed the concentrations of naturally-occurring radionuclides, such as ⁴⁰K that averaged 66±13 Bq kg⁻¹ (w.w.) in mussels and 618±21 Bq kg⁻¹ (w.w.) in fish. Nevertheless, artificial radionuclides accumulated in biota increase the radiation dose received by organisms from natural radiation sources and may also increase the risk of radionuclide transfer to humans. Reduction of radioactivity in effluent discharges towards improved estuarine water and sediment quality is debated as a requirement for sustainable use of the estuary.

Keywords: Technetium-99m, Iodine-131, Cesium-137, Natural radionuclides, Radioactive contamination, Urban wastewater

RESUMO

Amostras de sedimento, água, mexilhão e peixe foram colhidos mensalmente durante um período de 19 meses em várias estações ao longo da margem norte do estuário do Tejo (Lisboa, Portugal). As amostras foram imediatamente analisadas para radionuclídeos emissores gama, em especial para os de origem artificial. Entre estes, o ¹³¹I e ^{99m}Tc foram detectados na maioria das amostras de mexilhões (*Mytilus galloprovincialis*) e de tainha (*Liza ramada*). A distribuição espacial das amostras contendo esses radionuclídeos indicou contaminação continuada em Alcântara e Praça do Comércio, e, nalgumas ocasiões, também em Chelas. Ao longo do ano, em geral não se detectou contaminação em Algés, na entrada do

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estuário, e em Beirolas na zona média do estuário cerca da Ponte Vasco da Gama, mas a contaminação radioactiva foi consistentemente detectada nas áreas que recebiam as descargas de águas residuais urbanas da cidade de Lisboa. Os radionuclídeos ^{131}I e $^{99\text{m}}\text{Tc}$ são utilizados em medicina nuclear e têm origem nos efluentes líquidos hospitalares descarregados no sistema de esgotos da cidade. A sua detecção junto à Praça do Comércio relacionou-se com as descargas do esgoto principal da Baixa de Lisboa que, nos anos da amostragem ainda descarregava naquela área águas residuais urbanas não tratadas. Perto de Alcântara, onde se localiza a descarga de águas residuais urbanas tratadas pela estação de tratamento (ETAR) de Alcântara, a detecção daqueles radionuclídeos indicou que o tratamento de esgotos e o atraso introduzido na descarga dos efluentes tratados não foram suficientes para permitir o decaimento radioactivo. Os valores mais elevados de concentração de ^{131}I e $^{99\text{m}}\text{Tc}$ foram 12 ± 2 e $136 \pm 20 \text{ Bq kg}^{-1}$ peso fresco (p.f.) em mexilhões e peixes, respectivamente. Essas concentrações não excederam as concentrações de radionuclídeos de origem natural, tais como o ^{40}K que em média foram de $66 \pm 13 \text{ Bq kg}^{-1}$ (p.f.) em mexilhão e $618 \pm 21 \text{ Bq kg}^{-1}$ (p.f.) no peixe. No entanto, a acumulação dos radionuclídeos artificiais nos organismos aquáticos aumenta a dose de radiação recebida por esses organismos e aumenta o risco de transferência dos radionuclídeos artificiais para os seres humanos. A redução da radioactividade nas descargas de efluentes urbanos para melhorar a qualidade radiológica da água e sedimento do estuário é debatida como condição necessária para um uso sustentável do estuário.

Palavras-chave: Tecnécio-99m, Iodo-131, Césio-137, Radionuclídeos naturais, Contaminação radioactiva, Águas urbanas residuais.

1. INTRODUCTION

In nuclear medicine, radioisotopes are used both for diagnostic and therapeutic purposes. Currently, about 20 radioisotopes are produced for use in nuclear medicine, such as iodine (^{131}I , physical half-life $T_{1/2} = 8.0 \text{ d}$), technetium ($^{99\text{m}}\text{Tc}$, $T_{1/2} = 6.01 \text{ h}$), chromium (^{51}Cr , $T_{1/2} = 27.7 \text{ d}$), gallium (^{68}Ga , $T_{1/2} = 3.26 \text{ d}$) and cobalt (^{58}Co , $T_{1/2} = 70.96 \text{ d}$) (Parrington 1996). In Lisboa, several hospitals and other medical facilities daily use some of these radioisotopes imported from main international radiopharmaceutical suppliers. From this use, hospital solid waste and liquid effluents containing radioactivity are produced. Radioprotection measures are implemented in the medical facilities according to international standards, to prevent or reduce the irradiation and contamination of the staff and facilities. Those measures include procedures for solid waste segregation and safe disposal, and procedures for liquid waste management including special bathrooms for patients under treatment with radiopharmaceuticals.

The radioactive liquid effluents produced at the medical facilities (from patient bathrooms and laboratory sinks) may contain relatively high levels of radioactivity depending on the clinical specialities in the facilities, amount of radioisotopes applied, and number of patients treated. The discharge of radioactive liquid effluents from medical facilities to the environment has been a matter of some concern and investigation in several large European cities leading to enforcement of regulations and inspections to control better radioactive discharges (Mundschenk, 1996; Barquero *et al.*, 2008; Bergman *et al.*, 2008; Fischer *et al.*, 2009; Krawczyk *et al.*, 2013). In the case of coastal discharges through marine outfalls the environmental radioactivity enhancement has also been assessed in some areas with a view to protection of fisheries and human consumers against contamination and to the protection of non-human biota against radiation (Carolan *et al.*, 2011). Often, environmental radioactivity surveys of such areas are carried out to check compliance with radiation dose limits to members of the public (EU, 1996).

In Lisboa, some hospitals are equipped with tanks for temporary storage of radioactive liquid effluents in order to allow for radioactive decay before release into the city sewage

system. Nonetheless, not every medical facility performs radioactivity analysis of the effluents and keeps records of radionuclide composition and amounts of radioactivity released into the Lisboa sewage system. Besides, patients can go home and use the ordinary sewage collection and treatment works from there. After collection in the sewers system, urban wastewaters are mostly treated. Treated wastewaters plus still untreated urban wastewater from some city areas were discharged through several sewer outlets into the Tejo estuary in the years of the sampling (Figueira and Monteiro 2001; Carvalho *et al.* 2002).

To assess the radioactivity levels in the Tejo estuary resulting from the sewage discharges from Lisboa, an assessment experiment was established in collaboration with the Lisboa municipal administration. Sampling took place in 2004-2005, and was based upon the use of estuarine aquatic compartments (biotic and abiotic) as indicators of contamination along the north bank.

2. MATERIALS AND METHODS

Sampling

Monthly sampling was carried out during a 19-month period, from June 2004 to December 2005, with the help of the Lisboa Fire Brigade. Sampling stations were selected along the shore, from Algés near the maritime traffic control tower at the sea entrance of the estuary, to the mid estuary near Sacavém in front of the Nuclear and Technological Institute (ITN). Most stations were located between the 25th April and the Vasco da Gama bridges. Locations were selected near the main urban wastewater discharges (sewer outlets) and in between them in places with walls or piers with intertidal mussel beds (Figure 1).

Water, bottom sediment, mussels (*Mytilus galloprovincialis*), and mullets (*Liza ramada*) were sampled at low tide. Both species were selected for monitoring because they are common biota of the Tejo estuary. Mussels are sessile bivalves eating upon suspended particulate matter that they filter from water, thus integrating and averaging contamination levels at the same place, while mullets are pelagic fish swimming up and down the estuary and are opportunistic feeders. Surface water samples were collected at about 3-5 m from the shoreline, where water depth

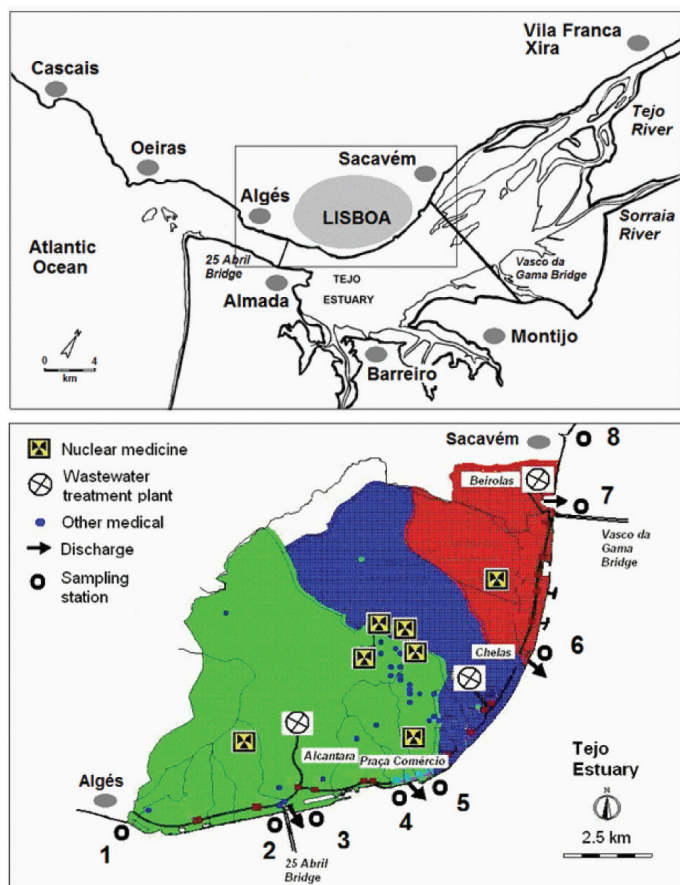


Figure 1. TOP: Tejo River estuary; the rectangle from Algués to Sacavém encompasses Lisboa and indicates the area of this study. BOTTOM: blow up of the area indicated above with a rectangle. Sampling stations are numbered from 1 to 8; in Lisboa city the coloured areas indicate the sewage collection zones and their respective wastewater treatment plants; main wastewater discharges from Lisboa sewerage system into the estuary are indicated with arrows.

Figura 1. CIMA: estuário do Rio Tejo; o rectângulo de Algués a Sacavém inclui Lisboa e indica a área deste estudo. EM BAIXO: ampliação da área indicada acima com um rectângulo. As estações de amostragem são numeradas de 1 a 8; na cidade de Lisboa as áreas coloridas indicam as zonas de recolha de águas residuais urbanas e respectivas estações de tratamento; as setas indicam as principais descargas de águas residuais do sistema de esgotos de Lisboa para o estuário.

may vary from 2 to 5 meters, directly into polyethylene cans, using a nylon net filter (300 μm mesh) to avoid large particles. Sub tidal sediment samples were collected at the same locations, using a bulk sediment sampler operated from the boat and collecting the top 10 cm sediment layer. Mussels were collected always at the same beds. Mulletts were collected with a cast net. Typical size of samples collected was 2-4 litres of sediment, 10 litres of water, 3 kg of mussels and 12 specimens of adult mulletts. Samples were conditioned in plastic containers, identified, and transported in ice chests to the laboratory for immediate processing and radiometric analysis. Not every month all desired sample types could be collected due to occasional adverse weather and tide conditions in the estuary, but the rate of successful sampling was high (>90%).

Radioactivity analysis

Analyses of two types were performed. One, was immediately performed by gamma spectrometry in order to allow for measuring the short-lived gamma emitting radionuclides used in nuclear medicine; the other was performed on oven-dry (60°C) and homogenized sample materials, and included alpha and gamma spectrometry for determining naturally occurring radionuclides with longer physical half-lives ($T_{1/2}$). Immediate gamma radiation measurements were performed on fresh material (*in toto*) placed in 1.0 L capacity Marinelli beakers on HpGe solid state detectors (Canberra). Gamma spectra were acquired for 1 hour and radionuclides identified and quantified using the software package Génie2000 (Canberra). These measurements allowed the determination of radionuclides such as ^{131}I and $^{99\text{m}}\text{Tc}$.

For deferred analyses, animal samples were treated by dissecting and separating fresh tissues from shells in the case of mussels (*Mytilus galloprovincialis*), and separating muscle, liver, and gonads in the case of mulletts (*Liza ramada*). Following oven drying and homogenization, sample aliquots were taken for gamma ray measurement using long counting times (24 h) for quantification of ^{40}K and ^{137}Cs . Separate aliquots were used for complete dissolution with HCl and HNO_3 acids followed by radiochemical separation and analysis of ^{210}Po and ^{210}Pb by alpha spectrometry. Briefly, ^{210}Po was analyzed after addition of an internal isotopic tracer (^{209}Po) to the sample, and ^{210}Pb was determined after in-growth of ^{210}Po from the parent radionuclide ^{210}Pb and second ^{210}Po plating on a silver disc (Carvalho 1995; Carvalho *et al.*, 2010). Alpha radioactivity measurements of the alpha radiation emitted by the silver discs were performed with ion implanted silicon detectors and alpha spectrometers OctetePlus (ORTEC EG&G) for determination of polonium-210 and lead-210.

A rigorous analytical quality control was maintained through the analysis of certified reference materials using the same methods and through participation in interlaboratory comparison exercises (Pham *et al.* 2006; Povinec *et al.*, 2007). Results are expressed in Bq kg^{-1} wet weight (w.w.) for mussels and fish, Bq kg^{-1} dry weight (d.w.) for sediment, and in Bq L^{-1} for water, unless stated otherwise.

The global assessment of radionuclide concentrations in several environmental compartments was made averaging all results per radionuclide and per sample type (Table 1). For computation of the mean concentration for each radionuclide, samples with radionuclide concentrations below the lower limit of detection were ascribed a value of zero and used in the mean computation.

3. RESULTS AND DISCUSSION

Gamma spectrometry analyses of fresh materials (biotic and abiotic) consistently allowed the identification and quantification of several artificial and natural radionuclides (Figure 2). The spectrograms show the gamma peaks of ^{131}I , and $^{99\text{m}}\text{Tc}$, short-lived gamma emitting radioisotopes used in nuclear medicine. ^{137}Cs (artificial radionuclide from fallout of nuclear weapon tests and nuclear accidents) and ^{40}K (naturally-occurring radionuclide) were detected as well. Long lived artificial gamma emitting radionuclides other than ^{137}Cs generally were not detected in sediments.

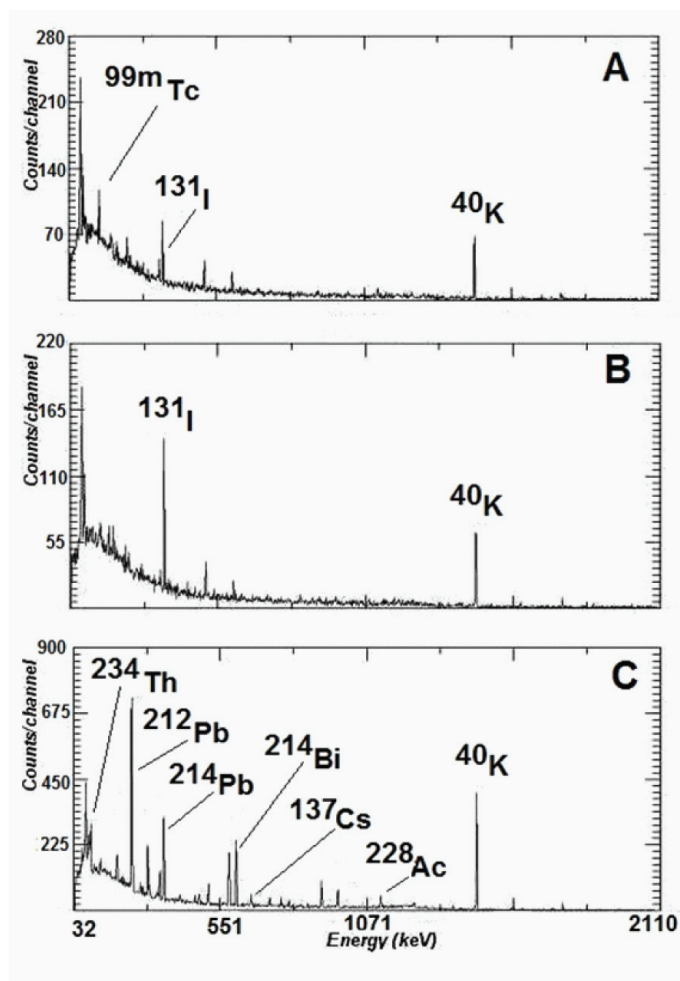


Figure 2. Examples of gamma spectra of estuarine samples: A, Water; B, Mussels; C, Sediment. The peaks of main radionuclides are identified. Radionuclides of anthropogenic origin are ^{131}I , $^{99\text{m}}\text{Tc}$ and ^{137}Cs . All other gamma-ray peaks correspond to naturally-occurring radionuclides.

Figura 2. Exemplo de espectros de radiação gama de amostras do estuário do Tejo: A, água; B mexilhões; C, sedimento. São identificados os picos principais de radionuclídeos emissores de radiação gama. Os radionuclídeos de origem antropogénica são o ^{131}I , $^{99\text{m}}\text{Tc}$ and ^{137}Cs . Os restantes picos correspondem a radionuclídeos de origem natural.

Artificial gamma emitting radioisotopes used in nuclear medicine were found in variable concentrations both in time and space. In all sampling stations, the two artificial radionuclides more consistently found were ^{131}I and $^{99\text{m}}\text{Tc}$. Station 3 (Alcântara Mar dock), station 4 (Praça do Comércio wharf), and station 6 (Chelas dock) were the locations displaying more frequently positive results in more sample types (Figure 3). Samples from other stations showed fewer cases of artificial radioactivity presence and in fewer types of environmental samples.

Round-the-year, samples from Alcântara Mar dock (station 3) showed the presence of ^{131}I , particularly in mussels. $^{99\text{m}}\text{Tc}$ was also often detected but generally in concentrations lower than those of ^{131}I . Samples from Praça do Comércio (station 4) also showed ^{131}I contamination, but less often than in

Alcântara. These two sampling stations were located closer to urban wastewater discharges. During the years of sampling at Alcântara Mar dock, a sewer outlet discharging treated urban wastewater from Alcântara wastewater treatment plant, and at Praça do Comércio a sewer outlet discharging untreated urban wastewaters, were fully operational (Figure 1).

^{131}I ($T_{1/2} = 8.0$ d) has a longer half-life than $^{99\text{m}}\text{Tc}$ ($T_{1/2} = 6.01$ h), which allows longer-lasting transport by tidal currents from the discharge points a few km along the shore either upstream or downstream. This was shown by positive radioactivity results from Algés to Beirolos, although radionuclide concentrations displayed decreasing frequency and decreasing concentration values from the stations at Alcântara and Praça do Comércio to both ends of the city area (Figures 1 and 3). At station 8, near Sacavém, no biota samples were available and only water and sediments could be analysed. $^{99\text{m}}\text{Tc}$ was the only radionuclide of artificial origin detected there, and in low concentrations.

Concentrations of ^{131}I in sediments were in average lower than those of technetium, which is in line with known K_d sediment-water partitioning coefficients ($K_d = \text{Bq kg}^{-1}$ in sediment dry weight/ Bq L^{-1} in water) reported in literature: K_d values are of about 70 for iodine and 10^2 - 10^3 for technetium in the marine environment (IAEA, 2004). Cesium-137 was rarely detected and always in low concentrations in sediments; furthermore it would be very difficult to measure ^{137}Cs in small volume water samples due to high sorption onto sediments of this radionuclide, with K_d values of 10^4 - 10^5 (IAEA, 2004).

At the same sampling station radionuclide concentrations showed wide variations over time, reflecting also the fluctuation of radioactivity in wastewater discharges (Figure 4). Mussels were the environmental compartment that showed the highest number of positive results (*i.e.*, displaying radionuclide concentrations above detection limit and quantifiable) for artificial radioactive contamination. Practically, in most stations there were always mussels contaminated with ^{131}I , and often with $^{99\text{m}}\text{Tc}$ also. This is due to the filter-feeding activity of these molluscs that filter large water volumes to feed upon suspended organic particles. It is interesting to notice that the mussels concentrated $^{99\text{m}}\text{Tc}$ from contaminated water to values sometimes much higher than those determined in mussels (Figure 4).

Radioactive iodine and technetium are present also in waste discharges from nuclear fuel reprocessing plants in UK and France into the marine environment, and have been subject to research and environmental surveys (Keogh and al., 2007; Smith et al., 2001). For example, $^{99\text{m}}\text{Tc}$ from these waste discharges was consistently measured over several years along the East coast of Ireland in concentrations from 19-45 mBq/L in seawater. Seaweeds accumulate technetium to high concentrations with concentration factors relative to water of about 1.2×10^5 on dry weight (d.w.) basis ($\text{CF} = \text{Bq kg}^{-1}$ d.w. seaweed/ Bq L^{-1} filtered sea water), while the concentration factor on a wet weight basis for the edible part of mussels was from about 500 to near 1000, and for fish muscle was around 12 (Smith et al., 2001). While these isotopes measured in the Irish sea are inorganic forms of Tc and I, the radioisotopes in samples from the Tejo estuary are most likely still bound to the radiopharmaceutical organic molecules. This organic

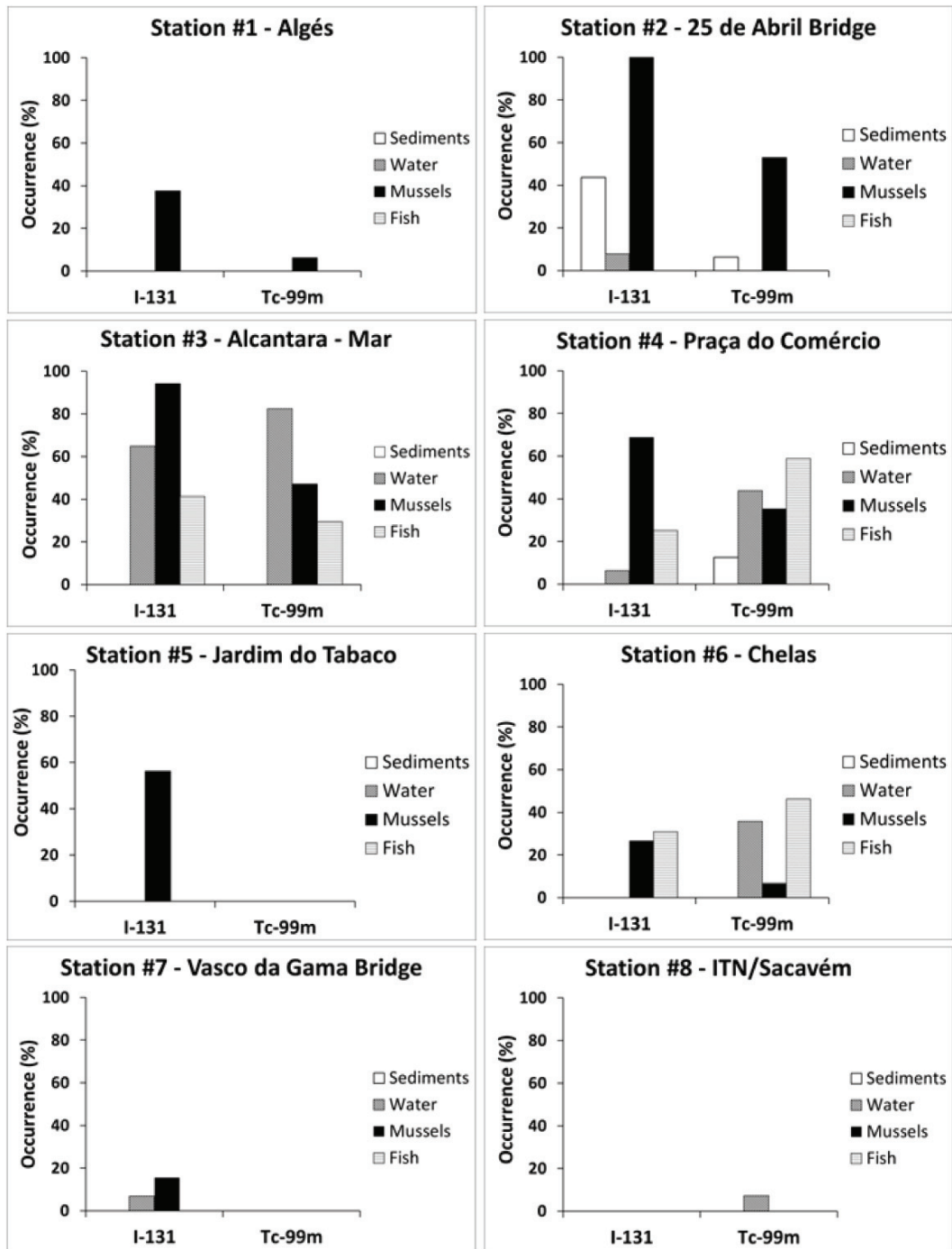


Figure 3. Frequency of occurrence (%) of artificial radioisotopes in estuarine samples for each sampling station in the Tejo estuary during the 19-month survey period (2004-2005).

Figura 3. Frequência de ocorrência (%) de radioisótopos artificiais em amostras do estuário em cada estação de amostragem no estuário do Tejo durante o período de 19 meses do estudo (2004-2005).

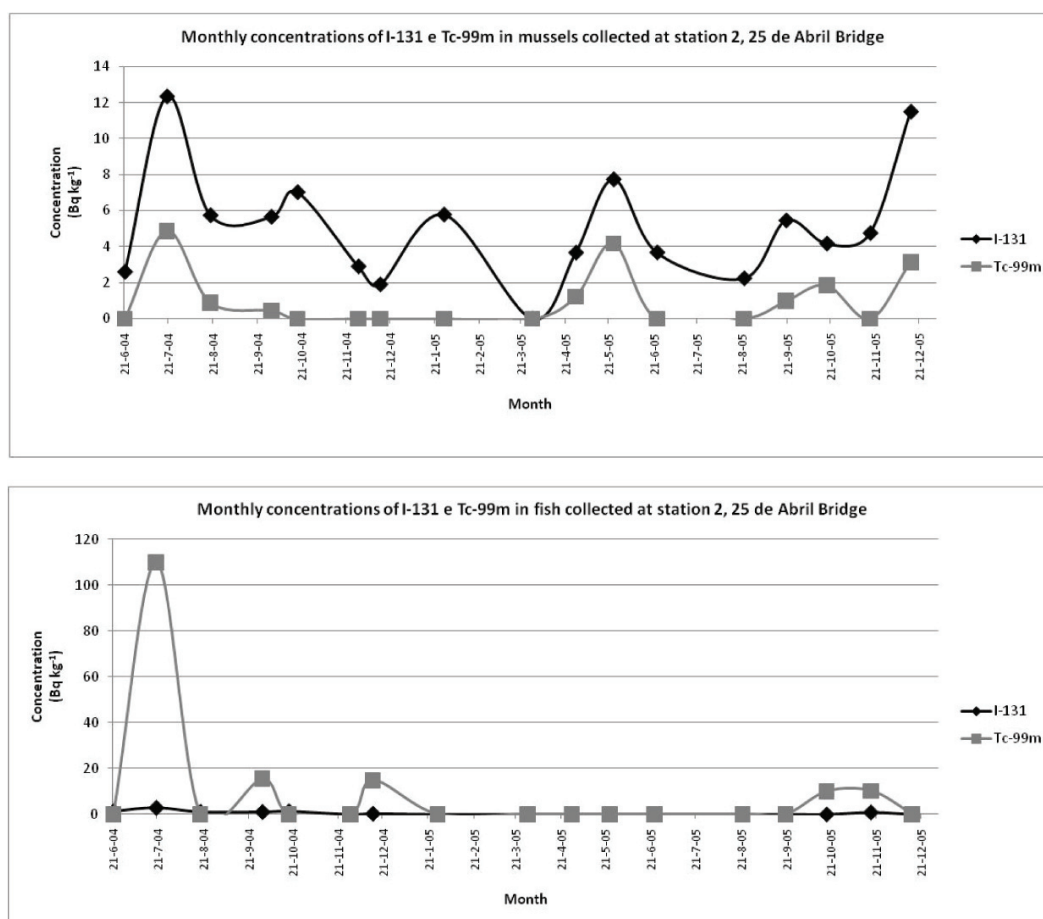


Figure 4. Concentrations of ^{131}I and $^{99\text{m}}\text{Tc}$ (Bq kg $^{-1}$, in toto) in mussels and fish collected at the sampling station 2 (25 April Bridge) during this survey.

Figura 4. Concentrações de ^{131}I e $^{99\text{m}}\text{Tc}$ (Bq kg $^{-1}$, in toto) no mexilhão e peixe colhidos na estação 2 (Ponte 25 de Abril) durante o período deste estudo.

binding might contribute to a much higher accumulation in mullet than in mussels, *i.e.*, the reverse than observations made in Irish Sea on the bioaccumulation of inorganic technetium and iodine. This observation indicates that bioaccumulation of radiopharmaceutical residues by aquatic biota and food chain transfer in the estuarine environment might be different than bioaccumulation of inorganic forms of the same radioelements. Most likely, current knowledge obtained with studies on the environmental behaviour of inorganic I and Tc does not apply to residues of radiopharmaceuticals labelled with these radionuclides. These aspects need further investigation.

Po-210 concentrations determined in mussels' soft tissues and in tissues of the mullets *Liza ramada* were not identical (Figure 5). Instead, they varied with the tissue and between species, as expected. The fish muscle displayed the lowest concentrations measured in fish tissues, which for fish file consumers reduces the transfer of radionuclides, although consumption of other organs such as gonad would transfer much higher activity. It must be said that concentrations determined for naturally-occurring radionuclides, such as ^{210}Po , ^{210}Pb and ^{40}K in biota from the north bank of the Tejo estuary are comparable to values reported in similar species

from non-contaminated areas (Cherry 1974; Carvalho 1995; Dahlgard 1996). This indicates that in the Lisboa area there is no significant enhancement of these radionuclide concentrations above normal background values, as opposed to observations made at the south bank around Barreiro peninsula due to discharge of industrial wastes (Carvalho *et al.*, 2013).

The mean and range of radionuclide concentrations measured in estuarine samples throughout the sampling period, including artificial and main naturally-occurring radionuclides, is shown in Table 1. These radionuclides have different sources. The naturally-occurring radionuclides in mussels and fish tissues did not originate particularly in the urban wastewater discharges. Actually, ^{210}Po originates in the radioactive decay of uranium series radionuclides, which are ubiquitous in nature. In mussels and fish the concentrations of natural ^{210}Po and ^{210}Pb varied over the months, most likely under the effect of the organism physiological conditions. Indeed, it was recently shown in mussels that animal size and accumulation/expenditure of fat are parameters that explain the fluctuation of ^{210}Po concentrations in mussels throughout the seasons of the year (Carvalho *et al.*, 2010). The artificial radionuclides ^{131}I and $^{99\text{m}}\text{Tc}$ displayed average concentrations

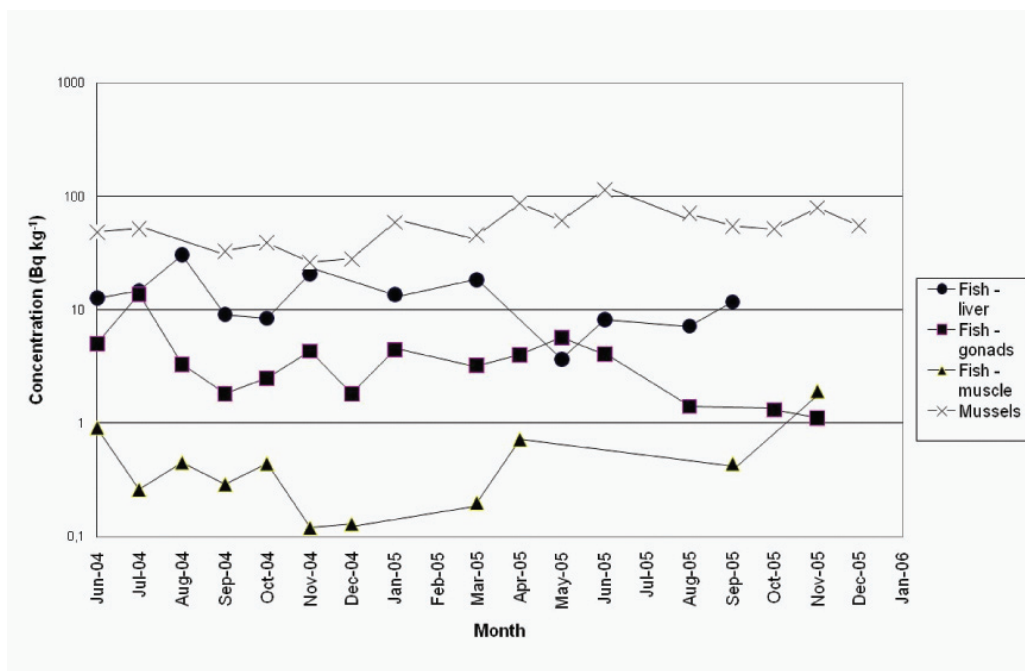


Figure 5. Polonium-210 concentrations (Bq kg^{-1} wet weight) in mussels' soft tissues and fish tissues collected at the sampling station 1 (Algés).

Figura 5. Concentrações de polónio-210 (Bq kg^{-1} peso fresco) nos tecidos moles de mexilhões e tecidos de peixes colhidos na estação 1 (Algés).

lower than those of the naturally occurring radionuclides for the entire data set. Notwithstanding, one should keep in mind that concentrations of natural radionuclides are nearly the same in most areas of the estuary while concentrations of artificial radionuclides range from zero in some areas to high values near the sewers discharges (Carvalho et. al, 2013).

Absorbed radiation doses due to these natural and artificial radionuclides in mussels and in whole body mullets were computed for the maximum concentrations measured, as reported in Table 1 (Pröhl 2003). It may be noticed that in spite of the absorbed radiation dose imparted to aquatic biota by artificial radionuclides, the main contribution to the overall absorbed dose is still originated by the naturally-occurring radionuclides. Amongst these, ^{210}Po gave the main contribution (95.5%) to the total absorbed dose in mussels while ^{40}K gave the main contribution (73%) in fish (Figure 6). Therefore, despite adding radiation dose to the aquatic organisms and to eventual human consumers of aquatic biota, the contribution of radioisotopes from hospital releases did not represent much in comparison with the radiation dose from naturally occurring radionuclides present in the estuarine environment. Notwithstanding, as discharges fluctuated widely throughout the year the contribution of artificial radionuclides to the radiation dose may also vary over time.

4. CONCLUSIONS

In the Lisboa area, the Tejo estuary received urban wastewater discharges that carried artificial radioisotopes originated in medical facilities. Besides the risk of radiation

exposure of workers of the city sewerage system and wastewater treatment plants, there is also the potential for exposure of members of the public to radioisotopes present in discharges into the estuary. Those radioisotopes were short lived and the temporary storage of contaminated effluents in tanks prior to the discharge should be sufficient to allow for radioactive decay. Nevertheless, there are either facilities without interim storage radioactive effluents or this interim storage was too short and, once released, a rapid transit from the medical facilities to the estuary (a few hours at the maximum) occurred in a systematic manner and allowed the round-the-year detection of ^{131}I and $^{99\text{m}}\text{Tc}$, in estuarine water, sediment, mussels, and fish.

The highest radionuclide concentrations were consistently measured near Alcântara and Praça do Comércio where two major sewers drained the urban wastewater discharges into the estuary. Discharges at these sites were of treated and untreated wastewaters, respectively. Results for Alcântara indicate that radionuclides from hospital effluents were not removed in the Alcântara wastewater treatment plant, as could be expected because waste water treatment plants are designed to treat nutrient organic loads and pathogens. Dispersal of sewage discharges by the river flow and tidal currents allowed for transport of radioisotopes up and down along the North bank and accumulation of ^{131}I by mussels in all sampling stations from Algés (estuary mouth) to Beiroas (mid estuary). Fish accumulated radionuclides, $^{99\text{m}}\text{Tc}$ in particular, but fish mobility renders less obvious than the sessile mussels the exact place of radionuclide uptake. Notwithstanding, $^{99\text{m}}\text{Tc}$ was consistently detected in water, mussels and fish in the stations of Alcântara, Praça

Table 1. Concentrations of artificial radionuclides and main naturally-occurring radionuclides (^{210}Po , ^{210}Pb and ^{40}K) measured in samples of the Tejo estuary. Mean of all data per radionuclide (n values) and range observed during the 19-month survey period.**Tabela 1.** Concentrações dos radionuclídeos artificiais e dos principais radionuclídeos naturais (^{210}Po , ^{210}Pb and ^{40}K) determinados em amostras do estuário do Tejo. Média e gama de todos os dados por radionuclídeo (n valores) determinados durante o período de monitorização de 19 meses.

Radionuclide		Sediments (Bq kg ⁻¹ ww)	Unfiltered water (Bq L ⁻¹)	Mussels (<i>in toto</i>) (Bq kg ⁻¹ ww)	Fish (<i>in toto</i>) (Bq kg ⁻¹ ww)		
^{131}I	n	88	120	110	47		
	mean	0.47	0.56	1.79	0.3		
	maximum	0.82±0.1	10±2	12±2	2.9 ±0.4		
	minimum	< 0.3	< 0.31	< 0.27	< 0.27		
$^{99\text{m}}\text{Tc}$	n	88	120	110	47		
	mean	0.76	0.62	0.56	11.6		
	maximum	1.6±0.3	13±2	9±1	136±20		
	minimum	< 0.3	< 0.27	< 0.28	< 0.31		
^{137}Cs	n	6	6	6	1		
	mean	1.6±0.2	< 0.35	0.1±0.03	< 1.9		
	maximum	1.8±0.2	-	-	-		
	minimum	1.3±0.2	-	-	-		
^{40}K	n	5	6	6	1		
	mean	328±53	20±5	35±18	618±21		
	maximum	429±10	27±5	66±13	-		
	minimum	283±10	14±2	18±5	-		
Mussels (soft tissues) (Bq kg ⁻¹ ww)					Fish (Bq kg ⁻¹ ww)		
					Liver	Gonads	Muscle
^{210}Po	n	-	-	16	14	14	11
	mean	-	-	56±23	13±7	3.8±3.1	0.53±0.52
	maximum	-	-	114±5	30±2	14±0.5	1.9±0.1
	minimum	-	-	26 ±1	3.66±0.19	1.1±0.1	0.12±0.01
^{210}Pb	n	-	-	16	14	14	11
	mean	-	-	1.09±0.56	1.39±0.87	0.32±0.34	0.10±0.09
	maximum	-	-	2.18±0.22	3.46±0.38	1.43±0.17	0.33±0.03
	minimum	-	-	0.39±0.03	0.33±0.04	0.07±0.01	0.03±0.002

<x, lower than the detection limit x, which value is indicated in each case.

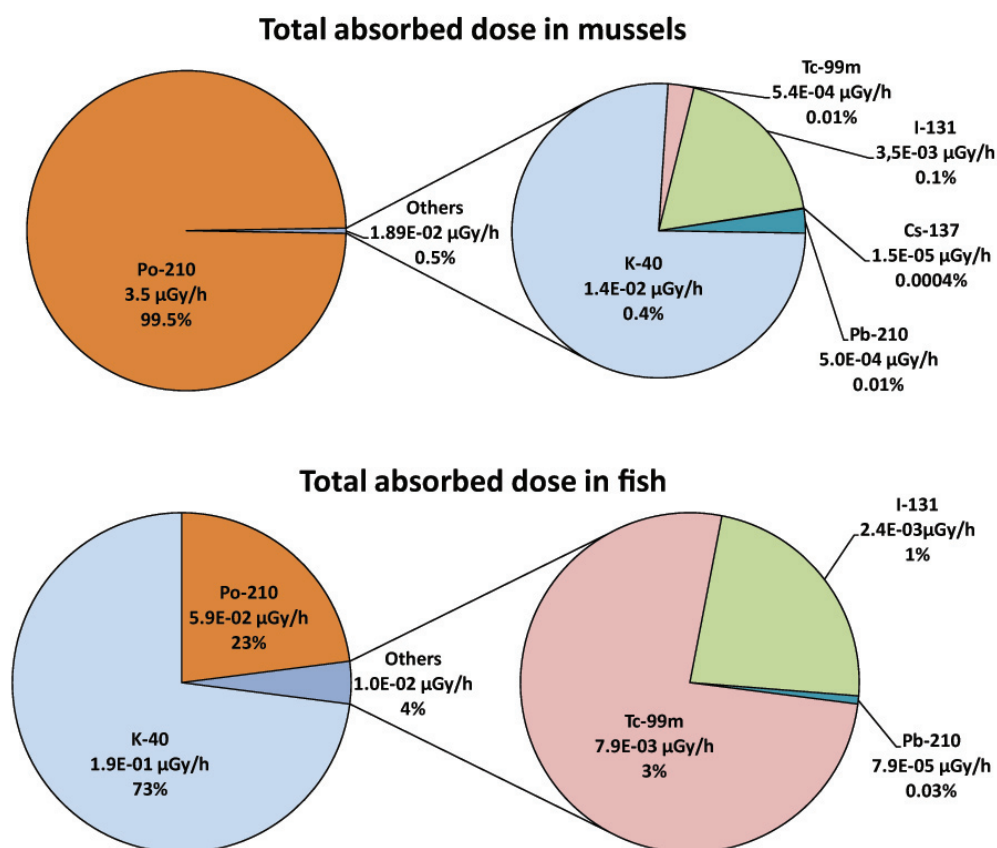


Figure 6. Total absorbed radiation dose from the main radionuclides measured in the tissues of mussels (*Mytilus galloprovincialis*) and mullets (*Liza ramada*) of the Tejo estuary.

Figura 6. Dose de radiação absorvida total dos principais radionuclídeos determinados nos tecidos de mexilhão (*Mytilus galloprovincialis*) e tainha (*Liza ramada*) do estuário do Tejo.

do Comércio and Chelas. Therefore, suitable measures to better control radioactivity in hospital effluents and in urban wastewater discharge are desirable in order to abate contaminants in the estuarine environment.

Recent improvements in the sewage system of Lisboa and in the Alcântara wastewater treatment plant aiming at the full treatment of urban wastewaters may have modified the pattern of waste discharges and contaminant distribution in the estuary described herein, and improved the quality of wastewater discharges. Nevertheless, this potential improvement was not checked for radioactivity as yet. Furthermore, it is likely that other discharges of radioisotopes from medical facilities exist around the estuary and the assessment of radioactive contamination of the estuary presented here probably is not complete. In addition, the use of radioisotopes in medicine and the number of treatments dispensed increase every year and new hospitals have been recently built in the Lisboa area and in other cities around the Tejo estuary and in its catchment area.

As professional fisheries along with sport fishing and leisure activities take place in the Tejo estuary, the implementation of a regular programme for radioactivity monitoring in the estuary seems much needed to keep under review the radioactivity levels and to assess and update the radiological

risk assessment to members of the public. Results from radioactivity monitoring may provide also useful feedback to better control effluent discharges and to improve radioactive waste management.

ACKNOWLEDGEMENTS

Are due to the Department of Environment and to the Lisboa Fire Department (Câmara Municipal de Lisboa) for the support and excellent collaboration with the sampling programme.

REFERENCES

- Barquero, R.; Agulla, M.M.; Ruiz, A. (2008) – Liquid discharges from the use of radionuclides in medicine (diagnosis). *Journal of Environmental Radioactivity*, 99(10):1535-1538. DOI: 10.1016/j.jenvrad.2007.12.009
- Bergman, S.S.; Cruz, I.; Avila, R.; Hasselblad, S. (2008) – A new approach to assessment and management of the impact from medical liquid radioactive waste. *Journal of Environmental Radioactivity*, 99(10):1572-1577. DOI: 10.1016/j.jenvrad.2007.12.005
- Carolan, J.V.; Hughes, C.E.; Hoffmann, E.L. (2011) – Dose assessment for marine biota and humans from discharges

- of ^{131}I to the marine environment and uptake by algae in Sydney, Australia. *Journal of Environmental Radioactivity*, 102(10):953-963. DOI:10.1016/j.jenvrad.2009.10.002
- Carvalho, F.P. (1995) – ^{210}Po and ^{210}Pb intake by the Portuguese population: the contribution of seafood. *Health Physics*, 69(4):469-480. DOI: 10.1097/00004032-199510000-00004
- Carvalho, F.P.; Oliveira, J.M.; Alberto, G.; Batlle, J.V. (2010) – Factors affecting ^{210}Po and ^{210}Pb concentration in mussels and implications for bio monitoring programmes. *Marine Pollution Bulletin*, 60(10):1734-1742. DOI: 10.1016/j.marpolbul.2010.06.025
- Carvalho, F.P.; Oliveira, J.M.; Gouveia, J.G.; Figueira, I.; Monteiro, P. (2002) – Radioactividade de origem artificial no estuário do Tejo na área ribeirinha do Município de Lisboa. *Actas do 10º Encontro Nacional de Saneamento Básico*, Universidade do Minho, Braga, Portugal. Relatório ITN/DPRSN, Série A, nº 23/2002, Lisboa, Portugal. <http://www.itn.pt/docum/relat/radiolog/rel-radiol-artif-tejo2002.pdf>
- Carvalho, F. P.; Oliveira, J.M.; Silva, L.; Malta, M. (2013) – *Radioactivity of anthropogenic origin in the Tejo Estuary and need for improved waste management and environmental monitoring*. *International Journal of Environmental Studies*, DOI:10.1080/00207233.2013.845714.
- Cherry, R.D.; Shannon, L.V. (1974) – Alpha Radioactivity of marine organisms. *Atomic Energy Review*, 12(1):3-45. <http://www.ncbi.nlm.nih.gov/pubmed/4598725>
- Dahlgård, H. (1996) – Polonium-210 in mussels and fish from the Baltic-North sea estuary. *Journal of Environmental Radioactivity*, 32(1-2):91-96. DOI: 10.1016/0265-931X(95)00081-K
- EU (1996) – *Directive 96/29/EURATOM*, Council Directive of 13 May 1996 laying down basic safety standards for the health protection of the general public and workers against the dangers of ionizing radiation. http://ec.europa.eu/energy/nuclear/radioprotection/doc/legislation/9629_en.pdf
- Figueira, I.; Monteiro, P. (2001) - *Implicações das descargas de águas residuais hospitalares nos sistemas de saneamento*. Acções de Sensibilização Ambiental em Águas Residuais Hospitalares, Direcção Geral de Infraestruturas e Equipamentos de Saúde, Ministério da Saúde, Lisboa, Coimbra, Porto, Portugal. Unpublished.
- Fischer, H. W.; Ulbrich, S.; Pittauerová, D.; Hettwig, B. (2009) – Medical radioisotopes in the environment - following the pathway from patient to river sediment. *Journal of Environmental Radioactivity*, 100:1079-1085. DOI: 10.1016/j.jenvrad.2009.05.002
- IAEA (2004)- Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment. *Technical Reports Series* No. 422. International Atomic Energy Agency, Vienna, Austria http://www-pub.iaea.org/MTCD/Publications/PDF/TRS422_web.pdf
- Keogh S.M., Aldahan A., Possnert G., Finegan P., Leon Vintró L., Mitchell P.I. (2007) - Trends in the spatial and temporal distribution of ^{129}I and ^{99}Tc in coastal waters surrounding Ireland using *Fucus vesiculosus* as a bio-indicator. *Journal of Environmental Radioactivity*, 95(1):23-38. DOI: 10.1016/j.jenvrad.2007.01.009
- Krawczyk E., Piñero-García F., Ferro-García M.A. (2013) - Discharges of nuclear medicine radioisotopes in Spanish hospitals. *Journal of Environmental Radioactivity*, 116:93-98. DOI: 10.1016/j.jenvrad.2012.08.011
- Mundschenk, H. (1996) – Occurrence and Behaviour of Radionuclides in the Moselle River - Part I: Entry of Natural and Artificial Radionuclides. *Journal of Environmental Radioactivity*, 30(3):199-213. DOI: 10.1016/0265-931X(95)00009-Y
- Parrington, J.R.; Knox, H.D.; Breneman, S.L.; Baum, E.M.; Feiner, F. (1996) – *Nuclides and Isotopes*. 64p., Fifteenth Edition, General Electric Nuclear Energy. ISBN: 978-0002988704
- Pham, M.K.; Sanchez-Cabeza, J.A.; Povinec, P.P.; Arnold, D.; Benmansour, M.; Bojanowski, R.; Carvalho, F.P.; Kim, C.K.; Esposito, M.; Gastaud, J.; Gascó, C.L.; Ham, G.J.; Hegde, A.G.; Holm, E.; Jaskierowicz, D.; Kanisch, G.; Llauro, M.; La Rosa, J.; Lee, S.-H.; Liong Wee Kwong, L.; Le Petit, G.; Maruo, Y.; Nielsen, S.P.; Oh, J.-S.; Oregioni, B.; Palomares, J.; Pettersson, H.B.L.; Rulik, P.; Ryan, T.P.; Sato, K.; Schikowski, J.; Skwarzec, B.; Smedley, P.A.; Tarján, S.; Vajda, N.; Wyse, E. (2006) – Certified reference material for radionuclides in fish flesh sample IAEA-414 (mixed fish from the Irish Sea and North Sea). *Applied Radiation and Isotopes*, 64(10-11):1253-1259. DOI: 10.1016/j.apradiso.2006.02.032
- Povinec, P.P.; Pham, M.; Barci-Funel, G.; Bojanowski, R.; Boshkova, T.; Burnett, W.; Carvalho, F.P.; et al. (2007) – Reference material for radionuclides in sediment, IAEA-384 (Fangataufa Lagoon sediment). *Journal of Radioanalytical and Nuclear Chemistry*, 273(2):383-393. DOI: 10.1007/s10967-007-6898-4
- Pröhl, G. (2003) – *Dosimetric models and data for assessing radiation exposures to biota*. FASSET Deliverable 3 Report for the EC 5th Framework Programme Contract FIGE – CT – 2000 – 00102. Disponível em: https://wiki.ceh.ac.uk/download/attachments/115802176/fasset_d3.pdf?version=1&modificationDate=1263905014000
- Smith V., Fegan M., Pollard D., Long S., Hayden E., Ryan T.P. (2001) - *Technetium-99 in the Irish marine environment*. *Journal of Environmental Radioactivity*, 56:269-284. [http://dx.doi.org/10.1016/S0265-931X\(00\)00209-5](http://dx.doi.org/10.1016/S0265-931X(00)00209-5)