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# Occurrence of and radioanalytical methods used to determine medical radionuclides in environmental and biological samples. A review

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ABSTRACT

Medical radionuclides are widely used in nuclear medicine practices today. Their production, handling and administration have different impacts on the environment and society due to the radioactive waste generated. Over recent years authors have taken an interest in the monitoring and safe disposal of this radiopharmaceutical waste, mainly in environmental and biological samples, and consequently a variety of radioanalytical methods for these matrices have been developed. The present review aims to outline the state of the art and the latest trends reported in the literature from 2007 to the present, focusing on the occurrence and determination of medical radionuclides in environmental and biological samples. Special attention is given to critically discussing the strengths and weaknesses of the different steps involved in determining medical radionuclides in these types of matrices. The methodologies presented are accompanied by examples.

## 1. Introduction

Nuclear medicine (NM) is a branch of medicine that uses non-encapsulated or unsealed radiopharmaceuticals to diagnose and treat various diseases. These radioactive compounds comprise a specific organic molecule labelled with a radionuclide. The radioisotopes used need to be selected taking into account a number of ideal requirements such as a) easy availability, b) relatively short effective half-life, and, depending on their final application, i.e. therapy or diagnostics, c) the type of radiation emitted. Table 1 lists the most commonly used radionuclides for diagnosis and therapy, with $^{99m}$Tc and $^{131}$I being the most widely administered for these purposes (Chain and Illanes, 2015; Orsini et al., 2010; Qaim, 2017; Saha, 2004; Zimmermann, 2006).

The handling of these unsealed radionuclides – as far as storage of radiation sources, internal transportation, production, preparation of radiopharmaceuticals and administration to patients are concerned – results in the generation of radioactive waste. Waste containing short half-life radioisotopes can be classified in a number of ways, such as a) airborne waste, b) liquid radioactive waste and c) solid radioactive waste, depending on the radionuclide itself, its use and its chemical and physical forms. All these types of waste may potentially be present not only in NM department areas but also outside these medical facilities due to hospital and out-patient discharges (ARPANSA, 2008; Krawczyk et al., 2013; Ravichandran, 2017). Waste management is of the utmost importance since medical staff and the general public could potentially be exposed to external or internal radiation. Thus, the determination of medical radionuclides in environmental and biological samples originating from NM practices is a matter of concern for various researchers due to their increased use. It is therefore necessary to work continuously to develop monitoring methods aimed at improving the evaluation of possible occupational intake and the assessment of possible worker exposure.

The aim of this review is to present recent advances in the methodologies used to determine medical radionuclides in environmental and biological samples, from 2007 to the present. It includes descriptions of the different approaches presented in the literature aimed at determining these kinds of radionuclides in several types of matrices and a detailed discussion of the main advantages and drawbacks of the strategies reported. A number of examples are then given to illustrate current trends in the strategies reported in the literature, focusing especially on collection, pretreatment and measurement. Finally, the radiological content of the samples analysed are presented in terms of activity concentrations.

The review is divided into two parts including associated up-to-date tables. The first focuses on those radionuclides present in envi-

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**Table 1**
Usual radionuclides used for diagnostics and therapy, indicating their type of emission and half-life (Biersack and Freeman, 2007).

| Use | Type of emission | Radioisotope | $T_{1/2}$ |
|---|---|---|---|
| **Diagnostic** | **γ** | $^{67}$Ga | 3.26 d |
| | | $^{99m}$Tc | 6.0 h |
| | | $^{111}$In | 2.83 d |
| | | $^{123}$I | 13.10 h |
| | | $^{201}$Tl | 73.10 h |
| | **β$^+$** | $^{11}$C | 20.48 min |
| | | $^{13}$N | 9.97 min |
| | | $^{15}$O | 2.04 min |
| | | $^{18}$F | 109.74 min |
| **Therapy** | **β$^-$** | $^{32}$P | 14.30 d |
| | | $^{89}$Sr | 50.50 d |
| | | $^{131}$I | 8.00 d |
| | | $^{186}$Re | 3.70 d |
| | **α** | $^{223}$Ra | 11.44 d |
| | **Auger electron** | $^{67}$Ga | 3.20 d |

ronmental samples: a) air samples from inside the NM department work environment, which are used to assess the radiation hazard to exposed medical staff, and b) samples from areas surrounding a medical centre, such as sewage, wastewater treatment plant (WWTP) samples, surface water or sediments and biota, which are frequently analysed to monitor unsealed liquid radioactive waste from hospitals and its impact on the environment. Such discharges are released into the sewage system a) directly as controlled disposal, or b) after decay in abatement systems, although there is no common disposal strategy for the European Union (European Commission, 2000; Janssens et al., 2013; Petrucci and Traino, 2015). Patients' home excretions also contribute to the discharges that should be taken into account (Andrés et al., 2011; Barquero et al., 2008a, 2008b). The second part deals with biological samples, which can come from a) medical staff, these being used to evaluate the related committed effective dose at which they can be exposed, and b) treated patients, these being useful for assessing the radiation safety of family and medical workers helping these patients. One of the most commonly analysed biological samples is urine, since this is the main excretory pathway from the human body (International Atomic Energy Agency (IAEA), 2018). However, there are other biological samples that are also of interest, such as the air exhaled by patients, saliva and skin swabs, which are also taken into consideration in the present review (Castellani et al., 2013).

## 2. Environmental samples: radioanalytical methods

As explained in the Introduction, the medical radionuclides used in NM departments are usually present in environmental samples because of their handling and administration, and consequently they can be found in the waste generated. It is for this reason that over the past twelve years there has been much interest in their determination. The following sections (2.1 and 2.2) contain a detailed discussion focusing on the strengths and limitations of the analytical methods proposed in the literature for this purpose. The most relevant papers published over this period as regards indoor air samples are summarized in Table 2, which shows the specific collection, pretreatment and measurement strategies commonly applied with these types of sample, along with the radiological content in terms of activity concentrations of the different radionuclides found. All this information is also presented in Tables 3 and 4, but, referring to outdoor liquid and solid samples respectively. Generally speaking, the radionuclides most frequently found in all these types of sample are a) $^{131}$I, because of its use in thyroid disease treatment and its highly volatile nature, and b) $^{99m}$Tc, due to its administration in radioaerosol form in diagnostic treatments. Other short half-life emitters such as $^{18}$F, $^{223}$Ra and $^{133}$Xe have also been determined in indoor air samples.

### 2.1. Indoor air samples

In the recent literature, one of the researchers' main concerns is the analysis of air samples from NM department work environments as part of internal dose assessment programmes. In general, the analytical procedures presented in published studies focusing on the quantification of different radionuclide activities in these samples are quite similar and commonly involve two steps: a) sample collection and b) measurement.

The most usual strategies applied for air collection to determine the most widely used medical radionuclides are based on active sampling (AS) (Brudecki et al., 2018; Carneiro et al., 2015; Damien et al., 2015; Ferdous et al., 2017; Ferrand et al., 2010; Hoi et al., 2017, 2016; Jiemwutthisak et al., 2012; Kopisch et al., 2011; Leners et al., 2011; Unpublished results, J. Martínez; Mietelski et al., 2005; Schomäcker et al., 2017), passive sampling (PS) (Jiménez et al., 2012; Mróz et al., 2018) and *in situ* monitoring (De Massimi et al., 2017; Giardina et al., 2015; Kawase et al., 2015; Suhariyono and Bunawas, 2015; Yamamoto et al., 2018).

In the case of AS, different filter media have been used depending mainly on the radionuclide to be determined. Regarding $^{131}$I, the selection of materials depends on the form in which this radionuclide is present in the air. Particulate filters are used to retain the elemental or different iodine compounds associated with the particles, while a wide range of solid adsorbent surfaces are used to trap $^{131}$I, such as gaseous molecular iodine, organic and inorganic compounds (Schomäcker et al., 2017). A review was recently published providing the main advantages and drawbacks of various porous materials used to extract the radioactive iodine present in air samples (Huve et al., 2018). Of the different materials used to collect the $^{131}$I present in indoor air samples, the most commonly used is activated charcoal (AC) (Brudecki et al., 2018; Carneiro et al., 2015; Damien et al., 2015; Hoi et al., 2017, 2016; Jiemwutthisak et al., 2012; Kopisch et al., 2011; Mietelski et al., 2005; Schomäcker et al., 2017). It has been reported that, due to its physical characteristics such as high surface area and pore volume and size (Nandanwar et al., 2016), AC is capable of retaining $^{131}$I gaseous fraction. Other impregnation substances have also been used to improve the adsorption capacity of this material (González-García et al., 2011; Gourani et al., 2014). González-García et al. (2011), for example, reported that triethylenediamine (TEDA) improves AC methyl iodide affinity by forming stable ammonium salts. In the case of $^{99m}$Tc, the most commonly used filter materials are paper filters (Ferdous et al., 2017; Ferrand et al., 2010), borosilicate fibre filters (Unpublished results, J. Martínez) and HEPA H14 filters (Bombardier et al., 2012). The reported papers demonstrate that all these materials are capable of trapping 0.1–0.3 μm particles with an efficiency as high as 99% (Ferrand et al., 2010; Unpublished results, J. Martínez).

The active sampling collection methodologies also vary according to the operating flow rate applied. In this regard there are strategies based on low volume (LVS) and high volume samplers (HVS). The sampling approaches based on LVS are the most widely employed, and noteworthy in this group are personal air samplers and static air samplers (Fig. 1, a and b respectively) (Carneiro et al., 2015; Damien et al., 2015; Ferrand et al., 2010; Hoi et al., 2017, 2016; Jiemwutthisak et al., 2012; Kopisch et al., 2011; Leners et al., 2011; Unpublished results, J. Martínez; Schomäcker et al., 2017). In both cases researchers collect air samples at approximately 1.5 m above the floor to simulate the breathing zone (Brudecki et al., 2018; Ferrand et al., 2010; Hoi et al., 2017, 2016; Jiemwutthisak et al., 2012; Unpublished results, J. Martínez), following IAEA (1999) recommendations. Regarding personal air samplers (Fig. 1, a), these are usually used to collect representative samples of occupational inhaled air in order to estimate indi-

**Table 2**
A summary of reported analytical methods for determining medical radionuclides in indoor air samples.

| Isotope | Sampling method | | | Detector | MDA | Sampling site | Activity concentration | Ref. |
|---|---|---|---|---|---|---|---|---|
| | Type | Flow [$m^3$/h] | Support | | [Bq/$m^3$] | | [Bq/$m^3$] | |
| $^{131}$I | **AS - HVS** | 30 | Whatman aerosol glass filter<br>Charcoal filter impregnated with KI | HPGe | n.r. | Air above septic tank | 0.748[a]<br>0.0163[b] | Mietelski et al. (2005) |
| | | 51 | Paper filter | HPGe | n.r. | Hot laboratory | 0.0064[b] | Ferdous et al. (2017) |
| | | 30 | Petryanov filter FPP-15-1.5<br>AC impregnated with KI | HPGe | n.r. | Hot room<br>Nurses station | 492 ± 4[a]<br>7.2 ± 0.6[b]<br>208 ± 3[a]<br>8.5 ± 0.9[b] | Brudecki et al. (2018) |
| | **AS - LVS** | 0.06 | AC sandwich | HPGe | 0.016 (Bq) | Radiopharmacy laboratory | 7.4[c] | Carneiro et al. (2015) |
| | | 0.18 | Carbon impregnated cellulose filters | 3 × 3″ NaI (Tl) | 0.18 (Bq) | Fume hood | 31.6 ± 16[c] | Jiemwutthisak et al. (2012) |
| | | | | 2 × 2″ NaI (Tl) | 0.37 (Bq) | Therapy room | 31.9 ± 22.3[c] | |
| | | n.r. | Charcoal cartridge | Well counter | n.r. | Preparation and administration room | 2400[a] | Kopisch et al. (2011) |
| | | 2 | TEDA-AC cartridge | NaI (Tl) | n.r. | Patient room | 73.15–7563.81[a] | Damien et al. (2015) |
| | | 4.2 | TEDA-AC cartridge | HPGe | n.r. | Distilling, packing and processing and hot cell room | 814.96 ± 37.18[a] | Hoi et al. (2017) |
| | | 4.2 | TEDA-AC cartridge | HPGe | n.r. | Distilling room<br>Packing room<br>Hot cell room | 2580 ± 1011.7[a]<br>1986.5 ± 1754.7[a]<br>726.4 ± 169.6[a] | Hoi et al. (2016) |
| | | 4.5 | CP100 (TEDA-AC)<br>GF50 (glass fiber filter)<br>2 BE110 ($CdI_2$ on Chromorb-P)<br>GY-130 (silver zeolite filter) | NaI (Tl) | 0.1 | Exhaust air from laboratory | $33.36 \cdot 10^6$ (Bq)[c,d] | Schomäcker et al. (2017) |
| | **PS** | – | PicoRad™ | LSC | 0.14 | In-patient room<br>Dose preparation room<br>Dose administration room<br>Shielded cell | 0.037–0.498[a]<br>0.622[a]<br>0.202[a]<br>0.516[a] | Jiménez et al. (2012) |
| | | – | PicoRad™ with Petryanov filter | HPGe | n.r. | Hot room<br>Nurses station | 4.71 ± 0.82 (Bq)[a]<br>0.59 ± 0.36 (Bq)[a] | Mróz et al. (2018) |
| | **Online monitoring** | n.r. | – | Portable $LaBr_3$ | n.r. | Radioisotope production stack | 321.16 | Suhariyono and Bunawas (2015) |
| $^{99m}$Tc | **AS - HVS** | 51 | Paper filter | HPGe | n.r. | Hot laboratory | 0.0042[c] | Ferdous et al. (2017) |
| | **AS - LVS** | n.r. | Paper filter<br>AC cartridge | γ-spectrometry | n.r. | Administration room<br>Gammacamera room | $4.6 \cdot 10^3$<br>1.5 | Ferrand et al. (2010) |
| | | n.r. | n.r. | γ-spectrometry | n.r. | Dose administration room | 7–191 | Leners et al. (2011) |
| | | 5.4 | Borosilicate fibre filter | HPGe | 0.028 | Dose administration room<br>Gammacamera room<br>Corridor | $2.1 \cdot 10^5$–$6.4 \cdot 10^5$<br>360–1470<br>$2.4 \cdot 10^4$–$5.4 \cdot 10^4$ Bq/$m^3$ | (Unpublished results, J. Martínez) |
| $^{18}$F | **Online monitoring** | n.r. | – | NaI(Tl) with shielding | n.r. | Hot wait room<br>Hot laboratory<br>Hall<br>Radiopharmacy laboratory | 137<br>129<br>141<br>194 | De Massimi et al. (2017) |

**Table 2** (*Continued*)

| Isotope | Sampling method | | | Detector | MDA | Sampling site | Activity concentration | Ref. |
|---|---|---|---|---|---|---|---|---|
| | Type | Flow [$m^3$/h] | Support | | [Bq/$m^3$] | | [Bq/$m^3$] | |
| | | 10 | – | NaI(Tl) with shielding $LaBr_3$:Ce | n.r. | Radioisotope production stack | < 20 (Bq/L) | Giardina et al. (2015) |
| $^{223}$Ra | **Online monitoring** | n.r. | – | Electrostatic collecting-type detector | n.r. | Imaging room | n.r. | Yamamoto et al. (2018) |
| $^{133}$Xe | **Online monitoring** | n.r. | – | CsI(Tl) | n.r. | Administration room | 37 ± 2.1 μSv/h | Kawase et al. (2015) |

AS Active sampling.
HVS High volume sampler.
LVS Low volume sampler.
PS Passive sampling.
n.r. not reported.
[a] $^{131}$I gaseous fraction.
[b] $^{131}$I aerosol fraction.
[c] $^{131}$I total activity.
[d] $^{131}$I organic fraction.

vidual $^{131}$I intake (European Commission, 2018), and the flow rates at which they usually operate are below 0.2 $m^3$/h (Carneiro et al., 2015; Jiemwutthisak et al., 2012; Kopisch et al., 2011). Carneiro et al. (2015) recently used this type of sampler at a flow rate of 0.06 $m^3$/h to collect air samples from above a hand-made AC sandwich filter in a radiopharmacy laboratory which was capable of assessing the internal dose received due to the radionuclide inhalation mentioned earlier. In contrast, static air samplers (Fig. 1, b) working at flow rates of 2.0 $m^3$/h and 4.2 $m^3$/h are commonly used to monitor air conditions in the workplace (Damien et al., 2015; Hoi et al., 2017, 2016; Schomäcker et al., 2017). For example, LVS have been used by Schomäcker et al. (2017) as an effective tool for evaluating the $^{131}$I retention capability of fume hoods in NM facilities and for determining and distinguishing the $^{131}$I compounds present in air. Nowadays European standard EN 14175-4 (2004) tests the escape of hazardous agents from fume hoods, but the cited study suggests that, according to the air measurements taken, this standard is not sensitive enough to properly evaluate the protective function relating to radioactive iodine.

HVS (Fig. 1, c) has recently been introduced as a sampling methodology, working at flow rates of between approximately 30.0 $m^3$/h and 50.0 $m^3$/h (Brudecki et al., 2018; Ferdous et al., 2017). The main advantage of using HVS rather than LVS is the relatively short collection time involved, which enables a greater amount of air to be sampled, making it possible to take a considerable number of samples from different locations within a short period of time. However, LVS based methodologies are extensively used especially for the ease with which their equipment can be handled and for their portability compared to methodologies based on HVS.

As far as the detection system is concerned, various radiometric detectors have been used to quantify $^{99m}$Tc and $^{131}$I on active sampling filter media, but the usual choice is gamma-ray spectrometry. High-purity germanium detectors (HPGe) are the most usual detection technique employed in the literature (Brudecki et al., 2018; Carneiro et al., 2015; Ferdous et al., 2017; Hoi et al., 2017, 2016; Unpublished results, J. Martínez; Mietelski et al., 2005). Thallium-activated sodium iodide crystals (NaI(Tl)) have been also used in some cases (Damien et al., 2015; Jiemwutthisak et al., 2012; Schomäcker et al., 2017). For both types, the filter media require minimum handling before measurement. In addition, the particulate filter and the solid adsorbent materials used for sampling can be measured together, as long as the aim of the measurement is to quantify the total $^{131}$I activity in air (Carneiro et al., 2015; Jiemwutthisak et al., 2012; Schomäcker et al., 2017). However, other researchers prefer an independent measurement so as to be able to differentiate between and quantify the activity of various forms of $^{131}$I in the atmosphere through their specific adsorption in different materials (Brudecki et al., 2018; Ferdous et al., 2017; Hoi et al., 2017, 2016; Mietelski et al., 2005). Using these strategies, as can be observed in Table 2, a number of different authors in the literature have found the highest $^{131}$I activity concentration in the gaseous fraction (Brudecki et al., 2018; Mietelski et al., 2005). When comparing the results of the different studies in terms of activity concentrations there is much variability, which can be explained by differences in the NM departments themselves and their own particular ways of administering doses to patients, among other reasons. The presence of $^{131}$I in the air is high in places where high concentrations of this radionuclide are produced, handled and administered, as would be expected (Brudecki et al., 2018; Carneiro et al., 2015; Damien et al., 2015; Ferdous et al., 2017; Hoi et al., 2017, 2016; Jiemwutthisak et al., 2012). The activity concentrations found in these areas ranged from 32 Bq/$m^3$ (Jiemwutthisak et al., 2012) to 7563.8 Bq/$m^3$ (Damien et al., 2015). There is also a wide dispersion in the results for $^{99m}$Tc in air samples, since each medical centre uses different doses and different tracer molecules. Generally speaking, the highest activity concentrations have been found in the administration room, where the radioaerosol is produced for *in situ* administration (Ferrand et al., 2010; Unpublished results, J. Martínez). Leners et al. (2011) found the presence of $^{99m}$Tc here at levels of 7–191 Bq/$m^3$ despite the application of protection measures such as portable filtration units installed above the patient and the use of FFP2 face masks by the patient and medical staff. Martínez et al. (Unpublished results) recently determined the occurrence of this radionuclide in other areas of an NM department such as the corridor and the gamma camera room at levels of $2.4{\cdot}10^4$–$5.4{\cdot}10^4$ Bq/$m^3$ and 360–1470 Bq/$m^3$ respectively. This occurred during the performance of a diagnostic treatment when, due to the generation of a radioactive aerosol, it could have easily spread throughout the NM environment.

PS is an alternative sampling strategy used for determining radionuclides in air, i.e. $^{131}$I, and is another way of estimating this radionuclide's contribution to the effective occupational dose. There is a dearth of literature that uses this approach (Jiménez et al., 2012; Mróz et al., 2018) even though it has several advantages over AS insofar as it is a) cheaper, b) easy to handle, c) noise-free, d) unobtrusive while sampling, and e) requires no electrical current. Passive sampling is based on the deposition of the $^{131}$I present in indoor air onto AC, which can

**Table 3**
A summary of reported analytical methods for determining medical radionuclides in outdoor liquid samples.

| Sample | Sampling method | | Sample pretreatment | Detector | MDA | Activity concentration | | Ref. |
|---|---|---|---|---|---|---|---|---|
| | Type of sample | Volume [L] | | | [Bq/L] | $^{99m}$Tc [Bq/L] | $^{131}$I [Bq/L] | |
| **WWTP influent waters** | 24-h composed | 1 | Addition of cellulose powder | HPGe | n.r | 1.18–19.0 | 0.17–0.86 | Fischer et al. (2009) |
| | n.r. | n.r. | – | HPGe | n.r | 0.37–0.98 | 0.33–3.5 | Chang et al. (2011) |
| | Spot | 1 | $Ag^{131}I$ precipitation in acidic medium<br>Precipitation with $NH_4OH$ | LSC | 0.011 (Bq/kg) | | 0.49–2.6 (Bq/kg) | Jiménez et al. (2011b) |
| | Spot | 1 | – | HPGe | 0.06–0.08 (Bq/$kg_{ww}$) | | 0.65 (Bq/$kg_{ww}$) | Cosenza et al. (2015) |
| | Spot | 5 | 2-step iodine extraction procedure:<br>1- organic fraction<br>2- inorganic fraction<br>3- residual iodine fraction | HPGe | 0.030–0.1 Marinelli)<br>0.002–0.015 (PMMA dish) | | 5–10% AEI<br>70% BEI | Hormann and Fischer (2017) |
| | 24-h composed | 2 | 2. Vacuum filtering, $Ag^{131}I$ precipitation in acidic medium, using NaOH to increase pH | HPGe | 0.004 (filters) | | 0.07–0.39 | Martínez et al. (2018) |
| | 24-h composed | 5 | – | HPGe | 0.2 $^{131}$I<br>4.9 $^{99m}$Tc | <5–50 | <0.2–4.4 | Mulas et al. (2019) |
| **WWTP effluent waters** | n.r. | 4 | – | BEGe | 0.2 | | 0.25 | Smith et al. (2008) |
| | 24-h composed | 1 | Addition of cellulose powder | HPGe | n.r. | 0.07–3.89 | 0.04–0.98 | Fischer et al. (2009) |
| | n.r. | | – | HPGe | n.r. | < MDA | 0.41–1.49 | Chang et al. (2011) |
| | Spot | 1 | $Ag^{131}I$ precipitation in acidic medium<br>Precipitation with $NH_4OH$ | LSC | 0.011 (Bq/kg) | | 0.18–0.93 (Bq/kg) | Jiménez et al. (2011b) |
| | 24-h composed | | Acidify sample to < pH 2 with $HNO_3$ | HPGe | 0.1 | | < MDA - 4.7 | Veliscek Carolan et al. (2011) |
| | Grab | 1 | –<br>Filtering by using 47 mm 0.7 μm glass fiber filter.<br>Dry at 40 °C filters overnight | LEGe | 1.7<br>50,000 (Bq/kg)[c] | | 1.8–217<br>2.5–227[a]<br>61 - 2801 (Bq/g)[c] | Rose et al. (2012) |
| | Spot | n.r. | Vacuum filter, 47 mm 0.7 μm glass fiber filter | LEGe | 0.2 | | 0.9–8.1[a] | Rose et al. (2013) |
| | Spot | 1 | – | HPGe | 0.08–0.9 (Bq/$kg_{ww}$) | | 0–0.17 (Bq/$kg_{ww}$) | Cosenza et al. (2015) |
| | Spot | 5 | 2-step iodine extraction procedure:<br>1- organic fraction<br>2- inorganic fraction<br>3- residual iodine fraction | HPGe | 0.030–0.1 (Marinelli)<br>0.002–0.015 (PMMA dish) | | 70–80% AEI | Hormann and Fischer (2017) |
| | 24-h composed | 0.8 | – | HPGe | 0.03 | | 0.2–0.5 | Montenero et al. (2017a) |
| | 24-h composed | 5 | – | HPGe | 0.2 $^{131}$I<br>4.9 $^{99m}$Tc | <5–50 | <0.2–4.4 | Mulas et al. (2019) |
| **Hospital wastewater** | 24-h composed | | Acidify with HCl until pH 2 | HPGe | 0.2 | 0.9–1789.9 | | Martínez et al. (2018) |
| | Spot hourly | 1 | Addition of formaldehyde<br>Evaporation of 1 L | HPGe | n.r. | 6519 | 3625 | Krawczyk et al. (2013) |
| **Surface water** | Spot | 500 | Filter through a 0.45 μm filter cartridge | BEGe | 50 (Bq/kg)[c] | | 22 (Bq/kg)[c] | Smith et al. (2008) |
| | Spot | 10 | 1. Decante 1 L and add $H_2SO_4$<br>2. Reduce by evaporation at 70 °C to 50 mL<br>3. Decante | HPGe | n.r. | | 0.2–1.4 | Gilfillan and Timmers (2012) |
| | Spot | 10 | 1. Evaporate sample volume < 100 °C<br>2. Rinse any residue with 2.5% $HNO_3$<br>3. Vacuum filter the sample | LEGe | 0.03 | | 0.08–6.07 | Rose et al. (2013) |
| | Spot | 180 | Filter through 0.7 μm pore size filter | HPGe | $1{\cdot}10^{-4}$ | | $4{\cdot}10^{-3}$[b] | Montenero et al. (2017a) |
| | | | Addition of BioRad, filter | | 30 (Bq/kg)[c]<br>$1.7{\cdot}10^{-3}$[d] | | $3{\cdot}10^{-4}$[c] | |
| | Spot | 180 | Filter through 0.7 μm pore size filter<br>Sequential series of BioRad addition and treatment with iron precipitate | HPGe | n.r. | | $1.5{\cdot}10^{-3}$ - $7.5{\cdot}10^{-3}$<br>$2{\cdot}10^{-5}$ - $6.5{\cdot}10^{-4}$ | Montenero et al. (2017b) |

n.r. Not reported.
AEI iodine in organic fraction.
BEI iodine in inorganic fraction.
[a] Filtered effluent.
[b] Dissolved.
[c] Suspended particulate material.
[d] Water column.

**Table 4**
A summary of reported analytical methods for determining medical radionuclides in outdoor solid samples.

| Sample | Sample pretreatment | Detector | MDA | Activity concentration | | Ref. |
|---|---|---|---|---|---|---|
| | | | | $^{99m}$Tc | $^{131}$I | |
| **WWTP sludge** | Addition of cellulose powder | HPGe | n.r. | 138–6040 Bq/$Kg_{dw}$[a]<br>0.19–252 Bq/$Kg_{dw}$[b] | 11.8–574 Bq/$Kg_{dw}$[a]<br>0.14–201 Bq/$Kg_{dw}$[b] | Fischer et al. (2009) |
| | – | HPGe | 26 Bq/kg | < MDA<br>< MDA | 203–231 Bq/kg[b]<br>1961–2410 Bq/kg[c] | Chang et al. (2011) |
| | Dry at 50 °C for 20 h and ground to fine powder | HPGe | n.r. | | 8.1–45 Bq/$kg_{ww}$[b] | Veliscek Carolan et al. (2011) |
| | Homogenisation and dry at 100 °C for 1 h | HPGe | 0.1 Bq/kg | | 74.8–1238 Bq/$kg_{dw}$ | Jiménez et al. (2011a) |
| | Dry at 130 °C, crushed and siefted | HPGe | 67 Bq/$kg_{dw}$ | | 58–883 Bq/$kg_{dw}$ | Camacho et al. (2012) |
| | – | LEGe | 10–500 Bq/$kg_{dw}$ | | 27 - 148,000 Bq/$kg_{dw}$[b] | Rose and Swanson (2013) |
| | n.r. | HPGe | 0.02–0.3 Bq/$kg_{ww}$[e]<br>0.06–0.3 Bq/$kg_{ww}$[f]<br>0.07–0.35 Bq/$kg_{ww}$[c] | | 12.3 Bq/$kg_{ww}$[e]<br>n.r.<br>150 Bq/$kg_{ww}$[c] | Cosenza et al. (2015) |
| | Formaldehyde addition[a,b,h] | HPGe | 0.2 Bq/L | 119.6 Bq/L[a]<br>2.6 Bq/L[g]<br>3.4 Bq/L[b] | 0.5–4.2 Bq/L[a]<br>4.2–28.7 Bq/L[g]<br>2.1–7 Bq/L[b] | Martínez et al. (2018) |
| | Dry at 100 °C, crushed in a ball mill and siefted in a 250 μm sieve[c] | | 1,1 Bq/kg[c] | n.d. | 176.5–213.9 Bq/$kg_{dw}$[c] | |
| | 2-step iodine extraction procedure:<br>1 organic fraction<br>2 inorganic fraction<br>3 residual iodine fraction | HPGe | 0.030–0.1 Bq/L (Marinelli)<br>0.002–0.015 Bq/L (PMMA dish) | n.d. | AEI > BEI | Hormann and Fischer (2017) |
| | 2-step iodine extraction procedure:<br>1 organic fraction<br>2 inorganic fraction<br>3 residual iodine fraction | HPGe | n.r. | n.d. | 80% AEI and 10% BEI[g]<br>60% AEI and 20% BEI[b] | Martínez et al. (2019) |
| | Dehydrate[c] | HPGe | 13 Bq/$kg_{dw}$ $^{131}$I[a]<br>4 Bq/$kg_{dw}$ $^{131}$I[c]<br>154 Bq/$kg_{dw}$ $^{99m}$Tc[a]<br>98 Bq/$kg_{dw}$ $^{99m}$Tc[c] | 154–9530 Bq/$kg_{dw}$[a]<br>< MDA – 5000 Bq/$kg_{dw}$[c] | < MDA - 700 Bq/$kg_{dw}$[a]<br>60–1000 Bq/$kg_{dw}$[c] | Mulas et al. (2019) |
| | Dehydration and homogenization | HPGe | n.r. | | 202 - 1908 Bq/kg | Zannoni et al. (2019) |
| **Sediments** | – | BEGe | 0.7 Bq/kg | | 3.3–6 Bq/kg | Smith et al. (2008) |
| | – | HPGe | n.r. | | 0.10–106 Bq/$kg_{dw}$ | Fischer et al. (2009) |
| | – | HPGe | n.r. | <0.3–1.6 Bq/kg ww | <0.3–0.8 Bq/$kg_{ww}$ | Malta et al. (2013) |
| | Dry at 80 °C and homogenise using a mortar and pestle | LEGe | 3.6 Bq/kg | | 1.3–117 Bq/L | Rose et al. (2013) |
| | Homogenize and still wet | HPGe | 0.35 Bq/kg | | n.d. - 68 Bq/$m^2$ | Montenero et al. (2017a) |
| **Biota** | Algae: Unwash, patted dry and chopped | HPGe | 0.039 Bq/$kg_{ww}$ | | 0.37 ± 0.010 Bq/$kg_{ww}$ | Morita et al. (2010) |
| | Carp entrails | HPGe | n.r. | | 9–11 Bq/kg | Gilfillan and Timmers (2012) |
| | Mussels: Separate tissue from shells; muscle, liver and gonads | HPGe | n.r. | n.d. - 5 Bq/kg | n.d. - 12 Bq/kg | Malta et al. (2013) |
| | Mullets: Dry, homogenize | | | n.d. - 20 Bq/kg | n.d. | |
| | Algae: Dry at 50 °C for 20 h Ground to fine powder | HPGe | n.r. | | 1.2–73 Bq/$kg_{ww}$ | Veliscek Carolan et al. (2011) |
| | Algae: Fresh analysed | HPGe | 0.4 Bq/kg | | n. d. - 91 Bq/$kg_{dw}$ | Montenero et al. (2017a) |

[a] Primary sludge.
[b] Digested sludge.
[c] Dewatered sludge.
[e] Mixed liquor.
[f] Recycled sludge.
[g] Secondary sludge.

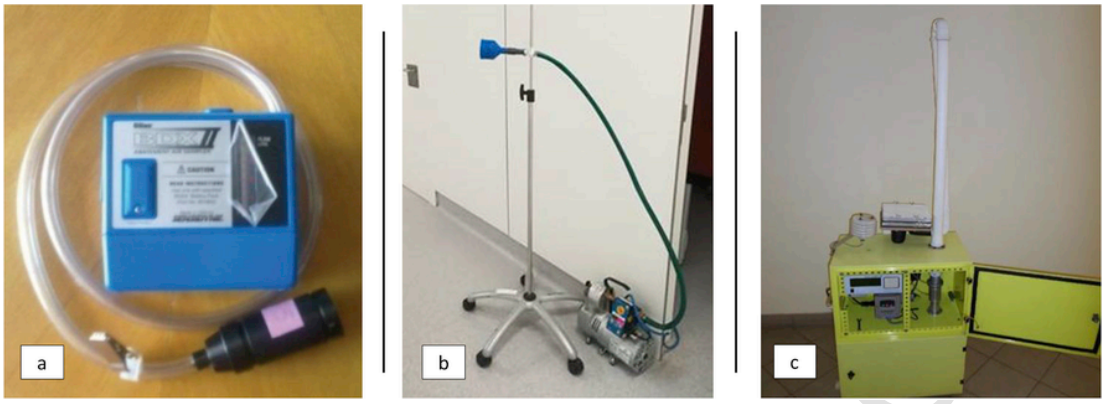


**Fig. 1.** Active sampling equipment. a) Personal air sampler. Reproduced from (Jiemwutthisak et al., 2012); b) Static air sampler. Personal image; c) High volume sampler. Reproduced from the open access study (Brudecki et al., 2018) (http://creativecommons.org/licenses/by/4.0/).

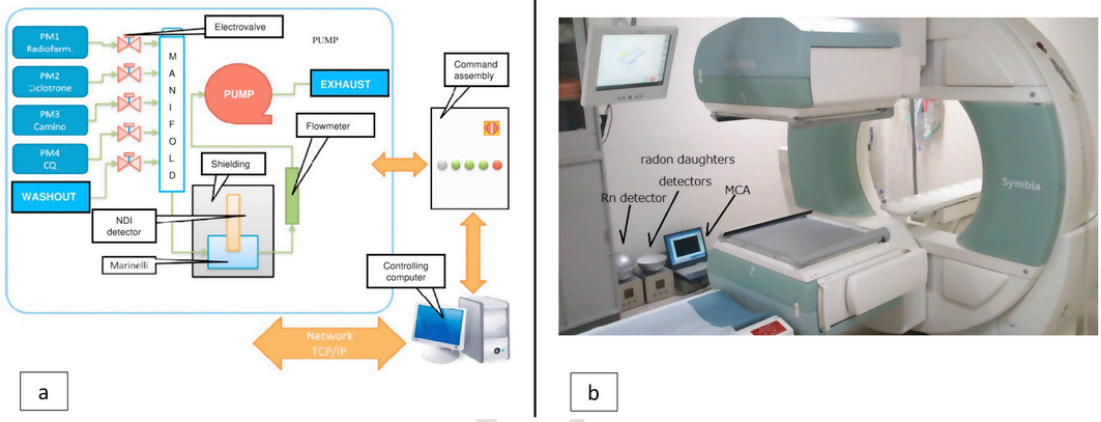


**Fig. 2.** Examples of continuous air sampling systems. a) Air sample pumped through a Marinelli beaker externally coupled to a scintillation detector. Reproduced permission from (De Massimi et al., 2017), Copyright 2017, Elsevier; b) $^{222}$Rn detector and $^{222}$Rn and $^{219}$Rn daughter detectors. Reproduced from the open access study (Yamamoto et al., 2018) (http://creativecommons.org/licenses/by/4.0/).

be contained in commercial PicoRad™ canisters (commonly used for $^{222}$Rn collection) (Jiménez et al., 2012). Recently a pilot study performed by Mróz et al. (2018) tried to increase the affinity of AC for radioactive iodine species by impregnating the porous surface with KI or NaOH, since they expected that trapping efficiency would be improved with these reagents. However, the authors reported that further work needed to be done in this area because the strategy was unsuccessful due to an unexpected reduction in the AC active surface.

One of the major drawbacks of these two studies is that the canisters are exposed for a lengthy time, which varies from three days (Jiménez et al., 2012) to one week (Mróz et al., 2018). The results obtained represent time-weighted average concentrations. With this sampling approach it is not possible to detect $^{131}$I activity concentration fluctuations in indoor air samples during the sampling time. The determination of this radionuclide in PS supports has been performed using the liquid scintillation technique (LSC) (Jiménez et al., 2012) and by HPGe (Mróz et al., 2018).

Continuous air sampling systems have also recently been used in the literature to monitor specific radionuclides such as $^{131}$I (Suhariyono and Bunawas, 2015), $^{18}$F (De Massimi et al., 2017; Giardina et al., 2015), $^{223}$Ra (Yamamoto et al., 2018) and $^{133}$Xe (Kawase et al., 2015). Using this strategy and depending on the detector, it is possible to obtain the radionuclide activity levels or air dose rates in real time. Continuous air sampling has a number of advantages over active and passive sampling. The sample collection and measurement steps are performed together, for instance, and the system can be managed with a laptop computer or even a smartphone. Moreover, the determination of short half-life emitters can be performed accurately using easy-to-handle equipment that can operate for long periods of time. Thus, continuous measurement can provide real time information about activity fluctuations and possible incidences of high peak activity. This sampling strategy has been applied in the literature for different purposes. One of the *in situ* monitoring applications, as illustrated in Fig. 2 a, is to detect the presence of $^{18}$F in the work environment of an NM department and assess occupational exposure (De Massimi et al., 2017). Another purpose is to monitor the stack emissions of $^{131}$I and $^{18}$F in production centres (Giardina et al., 2015; Suhariyono and Bunawas, 2015). In all these three studies, the methods presented are quite similar, involving pumping the air sample continuously through a Marinelli beaker (shielded or not) externally coupled to a scintillation detector such as NaI(Tl) or lanthanum bromide ($LaBr_3$), the latter having a better resolution. The study by Suhariyono and Bunawas (2015) compared the continuous air measurements of $^{131}$I present in air from a stack following an active sampling strategy using an LVS equipped with a paper filter and AC cartridges. They concluded that the continuous air sampling approach can be used instead of active sampling because it is accurate, provides immediate results, and is portable and easy to operate. However, the continuous strategy cannot distinguish the $^{131}$I compounds (i.e. $CH_3{}^{131}I$, $^{131}I_2$ or $HO^{131}I$) present in the sample because of the lack of any filter media in the system to trap these separately.

**Table 5**
A summary of reported analytical methods for determining medical radionuclides in biological samples.

| Sample | Isotope | Sampling method | Sample pretreatment | Detector | MDA | Activity concentration | Ref. |
|---|---|---|---|---|---|---|---|
| **Urine** | $^{131}$**I** | Spot sample | Store at 4 °C until measure | NaI | 72 Bq/L | n.d. | Shoji et al. (2007) |
| | | 24-h sample | Homogenization and determination of initial volume. | HPGe | 0.48–2.21 Bq/L | 1.2 ± 0.1–105 ± 1 Bq/L | Lucena et al. (2007) |
| | | Spot sample | – | HPGe | 4.8 Bq/kg | 5045 ± 45 Bq/kg maximum activity | Jeong et al. (2011) |
| | | Spot sample | | HPGe | | 6.47 ± 0.03 Bq/L – 283 ± 0.27 Bq/L | Ferdous et al. (2012) |
| | | 24-h sample | | HPGe | 0.1 Bq | n. r. | Bitar et al. (2013) |
| | | Spot sample | – | HPGe | n. r. | 5.78 ± 0.86 Bq/L - 389.95 ± 11.0 Bq/L | Ferdous (2016) |
| | | n.r. | – | HPGe | 0.047 Bq/L | | Bitar et al. (2016) |
| | | 24-h sample | Addition of 10% (v/v) concentrated $HNO_3$ | NaI | 0.88 Bq | 0.37 ± 0.09 Bq/L | Peekhunthod et al. (2017) |
| | | | | HPGe | 0.05–0.86 Bq | | |
| | | Spot sample | | NaI (Tl) | 1.7 Bq | n. r. | Matheoud et al. (2018) |
| | | 48 h post incorporation | – | Gammacamera | 272 Bq[a] | n. r. | Rodríguez-Laguna et al. (2010) |
| | | 48 h post incorporation | – | Gammacamera | 852 Bq[a] | n. r. | Díaz-Londoño et al. (2017) |
| | | Spot samples | – | HPGe | n.r. | 17–574 Bq/d | Kim et al. (2018) |
| | $^{99m}$**Tc** | Spot sample | – | HPGe | | 8.58 ± 0.03 Bq/L - 314 ± 1.92 Bq/L | Ferdous et al. (2012) |
| | | n.r. | – | HPGe | 0.14 Bq/L | | Bitar et al. (2016) |
| | | Spot sample | – | HPGe | n. r. | 24.47 ± 2.14 Bq/L - 1529.5 ± 36.5 Bq/L | Ferdous (2016) |
| | | Spot samples | – | HPGe | 3 Bq/L | 279.3 ± 80.4–18,652.4 ± 1218.7 Bq/L | (Unpublished resutls, J. Martínez) |
| | | 24-h sample | | | 1 Bq/L | ~ 2100 Bq/d | |
| | | Spot sample | | NaI (Tl) | 1 Bq | | Matheoud et al. (2018) |
| | $^{18}$**F** | Spot sample | | HPGe | 0.0270–0.0580 Bq/g | 0.138 ± 0.007 Bq/g –1.472 ± 0.027 Bq/g | Noh et al. (2016) |
| | $^{99}$**Tc** | n.r. | Evaporation and TEVA disk filtration | LSC Ultima Gold-LLT | n.r. | n.r. | Eichrom Technologies (2014) |
| | | n.r. | SPE TEVA resin automatic LOV | LSC Optiphase Hisafe | 1 Bq/L | n.r. | Villar et al. (2013) |
| | | n.r. | Reduction of salt effect Automatic DLLME | LSC Ultima Gold | 21 Bq/L | n.r. | Villar et al. (2015) |
| | | n.r. | Filtration: 0.45 µm syringe filter | LSC ULG-LLT | 0.023 Bq | n.r. | Khan and Um (2015) |
| | | n.r. | Colour removal PSr with Aliquat® 336 | LSC | 0.0036 Bq | n.r. | Barrera et al. (2016) |
| **Skin** | $^{131}$**I** | Wipe with alcohol 10 $cm^2$ of surface | – | Thyroid uptake probe | n. r. | 103 Bq/$cm^2$ front skin<br>214 Bq/$cm^2$ palm skin | Hamizah et al. (2012) |
| | | Wipe with alcohol 10 $cm^2$ of surface | | HGPe | n. r. | 125 Bq/$cm^2$ hand<br>< 50 Bq/$cm^2$ other parts | Guillot et al. (2016) |
| **Saliva** | $^{131}$**I** | Salivette© | | Activimeter | n. r. | in order of MBq/g | Guillot et al. (2016) |
| **Urine** | $^{131}$**I** | Spot sample | | Well counter | n. r. | 2928 ± 124 MBq | Demir et al. (2013) |
| **Exhaled breath** | $^{131}$**I** | AS - LVS<br>Aerosol filter<br>2 stages of DSM 11<br>2 layers of AC | | γ-spectrometry | | 10 kBq/$m^{3a}$ (after 24 h)<br>0,1 kBq/$m^{3a}$ (after 25 d) | Gründel et al. (2008) |
| | | AS - LVS<br>GF50 (borosilicate fibre)<br>BE110 ($CdI_2$ on Chromorb-P)<br>CP100 (TEDA on animal charcoal) | | NaI (Tl) | | 1–90 kBq/$m^3$ (after 1 h) | Schomäcker et al. (2011) |

n.r. Not reported.
AS - LVS Active sampling - low volume sampler.
[a] MDA for 1.6 L of urine.

Other specific continuous sampling approaches have also been applied. For instance, an electrostatic collecting-type detector was used to detect the alpha emitter daughter radionuclides of $^{119}$Rn in air samples when $^{223}$Ra is used therapeutically (Yamamoto et al., 2018) (Fig. 2, b). Another interesting strategy found in the literature is based on the determination of *in situ* dose rates. This method was applied during $^{133}$Xe

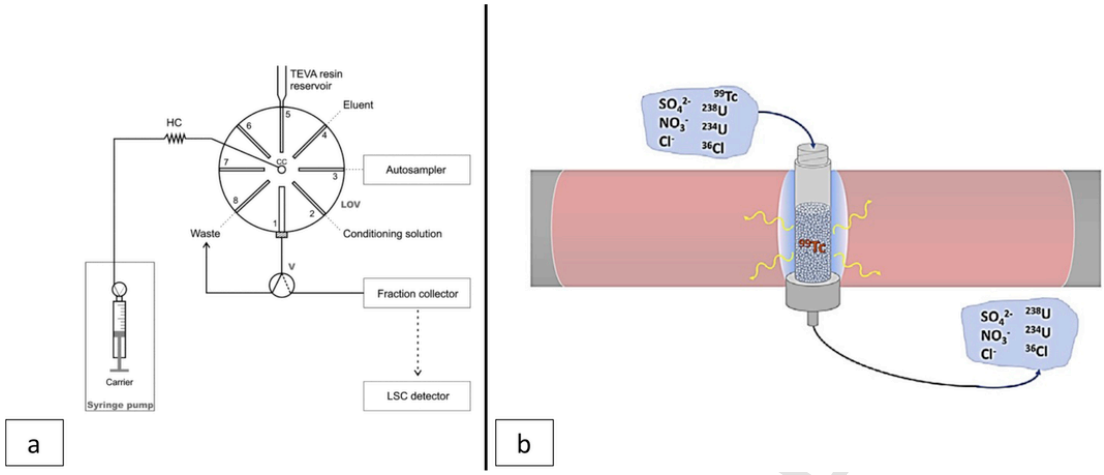


**Fig. 3.** Approaches for determining $^{99}$Tc in urine samples. a) Lab-on-valve system with TEVA integrated column. Reproduced with permission from (Villar et al., 2013). Copyright 2013, American Chemical Society; b) Plastic scintillation resin approach with PSr cartridge. Reproduced with permission from (Barrera et al., 2016). Copyright 2016, Elsevier B. V.

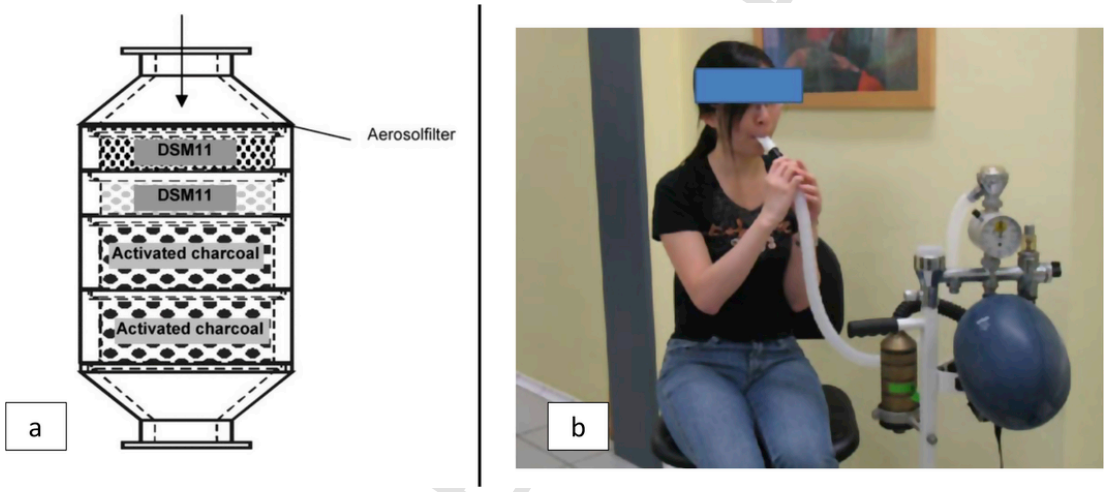


**Fig. 4.** $^{131}$I collection strategies for exhaled air from treated patients. a) Composition of a modular support system. Reproduced with permission from (Gründel et al., 2008). Copyright 2007, Oxford University Press; b) Illustration of a collection strategy. Reproduced from the open access study (Schomäcker et al., 2011) (http://creativecommons.org/licenses/by/4.0/).

administration in a lung-ventilation scintigraphy by employing a thallium-activated caesium iodide detector CsI(Tl) equipped with a module to convert the signal to dose equivalent (Kawase et al., 2015). The results of the applied strategy, expressed as air exposure dose levels, provide useful information to guarantee the safety of the patient and medical staff.

### 2.2. Outdoor samples

The administration of radiopharmaceuticals as unsealed sources in NM means the generation of radioactive effluents that reach the general sewage system, commonly discharged by one of two ways: a) from out-patients who release the excreta from home, and b) from their administration and use in medical centres whether or not they have radioactive decay tanks (Barquero et al., 2008b; Krawczyk et al., 2013). From these points the radioactive effluents can reach WWTP facilities. Once there, some radionuclides may be retained in the generated sludge, while others may be expelled into the environment (Martínez et al., 2018; Mulas et al., 2019; Palomo et al., 2010). In spite of the current good practices applied involving the reduction or elimination of radioactive waste from hospital effluents (Banerjee and Mitra, 2013; Rodríguez, 2012; Sudbrock et al., 2017), various studies have detected the presence of medical radionuclides in environmental samples from the areas surrounding medical centres (Punt et al., 2007; Sundell-Bergman et al., 2008). These samples can be classified according to their physical state as a) liquid samples such as hospital wastewater, WWTP influent and effluent waters and surface water, and b) solid samples such as WWTP sludge, sediments and biota.

In the studies reviewed, the approaches aimed at determining different radionuclides, mainly $^{131}$I and $^{99m}$Tc, in liquid samples generally involve three steps: a) sample collection, b) sample pretreatment, and c) measurement.

The collection of liquid samples is usually performed using polyethylene containers and the sample type may vary depending on the aim of the study: a) 24-h composed samples, which enable the total activity discharged during a day to be studied (Fischer et al., 2009; Martínez et al., 2018; Montenero et al., 2017a; Veliscek Carolan et al., 2011), or b) spot samples, which give an idea of the activity released at a particular point in time (Cosenza et al., 2015; Gilfillan and Timmers, 2012; Hormann and Fischer, 2017; Jiménez et al., 2011b; Krawczyk et al., 2013; Malta et al., 2013; Montenero et al., 2017a, 2017b, Rose et al., 2013, 2012). In general terms, liquid samples can be classified according to their activity concentration levels. Samples such as WWTP influent and effluent waters and hospital wastewater are directly influenced by the radioactive source (discharges from out-patients and medical centres) and usually have high activity concentration levels.

Studies reported in the literature, as shown in Table 3, have presented activity levels ranging from a) 0.07 Bq/L to 4.4 Bq/L in WWTP influent waters, and b) 0.14–217.0 Bq/L in WWTP effluent waters, to c) about 3625.0 Bq/L in hospital wastewater. These samples are not usually subject to pretreatment, although sometimes just a simple one is involved in the reported strategies (Chang et al., 2011; Cosenza et al., 2015; Fischer et al., 2009; Krawczyk et al., 2013; Martínez et al., 2018; Montenero et al., 2017a; Mulas et al., 2019; Rose et al., 2013, 2012; Smith et al., 2008; Veliscek Carolan et al., 2011). For example, Rose et al. (2012) filtered 1 L of WWTP effluent water through a 0.7 μm glass fibre filter, but this pretreatment showed no differences in terms of activity concentrations between the results obtained from the direct gamma-ray spectrometry measurement of the sample, from measurement of the filtered sample and from measurement of the filter used. In those cases reported by Jiménez et al. (2011b) and Martínez et al. (2018), other pretreatments based on silver iodide (AgI) precipitation are applied when the activity concentrations of WWTP effluent waters are in the low range of the corresponding presented levels. These procedures are based on chemical treatment of the sample presented by Baeza et al. (2004) and in UNE-EN ISO 10703 (AENOR, 2016), with some modifications made so as to optimize the measurement time by LSC counting (Jiménez et al., 2011b) and to selectively separate $^{131}$I from other radionuclides measured in a filter geometry using HPGe (Martínez et al., 2018). Other pretreatments are based on the flocculation strategies used in another study from the literature aimed at predicting the $^{131}$I dynamics and behaviour in the different WWTP compartments (Hormann and Fischer, 2017). To this end, the procedure consisted of relating the radioactive iodine content to a) the WWTP influent organic fraction, by using aluminium chlorohydrate based flocculant, b) the inorganic fraction, by precipitating $^{131}$I as silver iodide, and c) the residual fraction.

In the case of surface water samples, activity concentration levels are usually lower than for WWTP samples and hospital wastewater, in the range of $2{\cdot}10^{-5}$ Bq/L to 6.7 Bq/L. Hence the general strategy when analysing these samples includes as a pretreatment, as reported by various authors, a sample preconcentration based on a variety of approaches including evaporation or the use of anionic resins, among others (Gilfillan and Timmers, 2012; Montenero et al., 2017a, 2017b; Rose et al., 2013; Smith et al., 2008). Montenero et al. (2017a), for instance, present a method in which a BioRad AG 1-X8 anionic exchange resin was added to 180 L of surface water samples in order to trap the $^{131}$I. The authors take advantage of the presence of this radionuclide in WWTP effluent water to trace it in a large lake sample (Montenero et al., 2017a).

As regards detection techniques, HPGe is the most widely used radiometric detector for quantifying $^{131}$I and $^{99m}$Tc in liquid samples (Chang et al., 2011; Cosenza et al., 2015; Fischer et al., 2009; Gilfillan and Timmers, 2012; Hormann and Fischer, 2017; Krawczyk et al., 2013; Malta et al., 2013; Martínez et al., 2018; Montenero et al., 2017a, 2017b; Mulas et al., 2019; Veliscek Carolan et al., 2011), although low energy germanium detectors (LEGe) (Rose et al., 2013, 2012) and broad energy germanium detectors (BEGe) (Smith et al., 2008) have also been used. In some cases LSC has also been used for determining $^{131}$I content in WWTP influent and effluent water samples (Jiménez et al., 2011b).

Of all the liquid samples of this type measured in studies in the literature, hospital wastewater is the sample with the highest activities, as expected, achieving maximum values of 1789.9 Bq/L (Martínez et al., 2018) and 6519.0 Bq/L (Krawczyk et al., 2013) for $^{99m}$Tc and 3625.0 Bq/L for $^{131}$I. Almost all of these studies have focused on spot samples (Cosenza et al., 2015; Gilfillan and Timmers, 2012; Hormann and Fischer, 2017; Jiménez et al., 2011b; Krawczyk et al., 2013; Montenero et al., 2017a, 2017b; Rose et al., 2013; Smith et al., 2008). $^{131}$I has generally been found in influent WWTP samples with activity concentrations ranging from 0.07 Bq/L (Martínez et al., 2018) to 2.6 Bq/L (Jiménez et al., 2011b). This variation in the activities obtained could be due to a number of factors, as Mulas et al. (2019) report in their study. These authors concluded from their results that "activities found in the studies carried out in urban areas, where hospitals were equipped with abatement systems, showed a one-order-of-magnitude lower range where abatement systems are not used". In the same study, the presence of other NM radionuclides such as $^{99m}$Tc and $^{123}$I was also confirmed. There has also been much interest in the literature in determining medical radionuclides in WWTP effluent water samples and surface waters. Activity levels of few Bq/L have been found in such samples (Chang et al., 2011; Cosenza et al., 2015; Fischer et al., 2009; Gilfillan and Timmers, 2012; Jiménez et al., 2011b; Malta et al., 2013; Montenero et al., 2017a, 2017b; Mulas et al., 2019; Rose et al., 2012, 2013; Smith et al., 2008; Veliscek Carolan et al., 2011), as in the study by Cosenza et al., for example, in which the $^{131}$I activity concentrations in WWTP effluent samples ranged between 0 and 0.17 Bq/kg$_{ww}$ in the course of a long-term sampling campaign (Cosenza et al., 2015).

As previously mentioned, solid samples can also be influenced by NM departments. In particular, radionuclides can be found in a) WWTP sludge, since this sample may tend to concentrate the radioisotopes present in the influent water due to the facility's processes, and b) sediments and biota, due to the transference of radioisotopes between the aquatic media and these samples. The collection of these types of sample is usually performed using polyethylene containers. The literature includes several alternative proposals for these kinds of solid sample as pretreatment, although in some cases different authors have preferred direct measurement (Chang et al., 2011; Fischer et al., 2009; Gilfillan and Timmers, 2012; Malta et al., 2013; Martínez et al., 2018; Mulas et al., 2019; Rose and Swanson, 2013). Of the different approaches used as pretreatment, the usual choices generally involve drying, sieving and homogenizing the sample (Camacho et al., 2012; Jiménez et al., 2011a; Martínez et al., 2018; Montenero et al., 2017a; Morita et al., 2010; Veliscek Carolan et al., 2011; Zannoni et al., 2019). However, some of these treatment procedures have certain operational requirements, such as drying the sample, and in this case the temperature applied can be dramatic as regards the determination of $^{131}$I, since this radionuclide can undergo a volatilization process (Gilfillan and Timmers, 2012; Montenero et al., 2017a; Rose et al., 2012). The highest value found in the literature which ensures the preservation of this radioisotope in a sludge sample after drying is around 100 °C (Sundell-Bergman et al., 2009). Other studies in the literature, such as those by Hormann and Fischer (2017) and Martínez et al. (2019), aim to qualitatively evaluate the behaviour of $^{131}$I in the different compartments of a WWTP. The procedures are mainly based on isolating the radioactive iodine content present in three fractions (organic, inorganic and residual) of the different kinds of generated sludge by using flocculants and chemical precipitation in the form of silver iodide. Both the studies mentioned related their results to the WWTP processes, concluding that a) the $^{131}$I present in the secondary sludge is associated mainly with the organic fraction (particles, colloids and organic matter) due to the microbiological process involved in the aeration tank, and b) the presence of $^{131}$I in the digested sludge organic fraction diminished, probably due to oxidation of the organic matter brought about by anaerobic microorganisms.

Concerning detection systems, HPGe is the radiometric detector most frequently used to quantify $^{131}$I and $^{99m}$Tc in solid samples (Camacho et al., 2012; Chang et al., 2011; Cosenza et al., 2015; Fischer et al., 2009; Gilfillan and Timmers, 2012; Hormann and Fischer, 2017; Jiménez et al., 2011a; Malta et al., 2013; Martínez et al., 2019, 2018; Montenero et al., 2017a; Morita et al., 2010; Mulas et al., 2019; Veliscek Carolan et al., 2011; Zannoni et al., 2019), although LEGe (Rose et al., 2013; Rose and Swanson, 2013) and BEGe (Smith et al.,

2008) have also been used. The presence of these two radionuclides – and to a lesser extent others such as $^{67}$Ga, $^{111}$In and $^{123}$I – in WWTP sludge samples is common in almost all the studies reviewed.$^{131}$I has been found in a wide range of activity values in different WWTP sludges, ranging from 0.14 Bq/kg$_{dw}$ (dry weight) to 148000 Bq/kg$_{dw}$ (Camacho et al., 2012; Chang et al., 2011; Cosenza et al., 2015; Fischer et al., 2009; Hormann and Fischer, 2017; Jiménez et al., 2011a; Martínez et al., 2019, 2018; Mulas et al., 2019; Rose and Swanson, 2013; Veliscek Carolan et al., 2011; Zannoni et al., 2019). Activated sludge is the WWTP sludge type that tends to accumulate the highest activity levels of this radionuclide, mainly due to microorganism action during the previous biological treatment and interactions with organic matter (Cosenza et al., 2015; Hormann and Fischer, 2017; Kaplan et al., 2014; Martínez et al., 2019, 2018; Mulas et al., 2019). The radionuclide accumulation in sludge due to the settlement of particles or microbiological treatment is generally thought also to be governed by factors related to the WWTP facility, such as a) the time the wastewater takes to reach the WWTP, b) the population equivalents, c) the hydraulic retention time, and d) the age of the sludge (Mulas et al., 2019; Zannoni et al., 2019). In spite of the high activity concentrations found in this type of sample, the authors conclude that its handling and land use pose no occupational radiological risk (Cosenza et al., 2015; Jiménez et al., 2011a; Martínez et al., 2018; Mulas et al., 2019; Zannoni et al., 2019).

Since the removal of medical radionuclides in WWTP facilities is incomplete (Mulas et al., 2019), they have also been found in sediments and biota at a variety of lower activity levels ranging from $<0.3$ Bq/kg$_{ww}$ to 73 Bq/kg$_{ww}$ (Malta et al., 2013; Veliscek Carolan et al., 2011). The determination of $^{131}$I in these samples is today used to trace WWTP radioactive effluent in the aquatic environment. For example, there are different species of marine alga that are capable of concentrating this radionuclide, and these can therefore be used as biomonitoring organisms of $^{131}$I levels (Morita et al., 2010; Veliscek Carolan et al., 2011).

## 3. Biological samples: radioanalytical methods

Both NM workers and the general public can be radiologically exposed due to the presence of medical radionuclides in the NM department environment. In recent years the individual monitoring of these people to detect any radionuclide intake has been an issue of concern. Although the use of *in vivo* measurements (i.e. whole body counting or thyroid counting (Bitar et al., 2016)) is recommended for almost all short half-life radionuclides, the analysis of biological samples from these individuals is also important to complement the previous information and is useful for estimating internal occupational exposure (ISO, 2016).

Patients undergoing NM treatment can also act as radioactive sources, exposing caregivers and family members (Uhrhan et al., 2014), and therefore any protective measures that may have been taken by these people could be assessed by analysing the patients' body fluids.

The most frequently analysed biological sample in the reviewed studies is generally urine, since it is one of the main radionuclide excretion pathways from the human body and sample collection is easy and non-invasive (Hou, 2011; International Atomic Energy Agency (IAEA), 2009). Urine-based measurements have been commonly used as a) a screening method, b) when *in vivo* measurements cannot be performed, c) to verify possible internal contamination, and d) after a supposed accidental intake (Bitar et al., 2013; Breustedt et al., 2018). According to the literature, the radionuclides most usually found in urine samples are a) $^{18}$F (Noh et al., 2016), $^{99m}$Tc (Bitar et al., 2016; Ferdous, 2016; Ferdous et al., 2012; Unpublished results, J. Martínez; Matheoud et al., 2018) and $^{131}$I (Bitar et al., 2016, 2013; Díaz-Londoño et al., 2017; Ferdous, 2016; Ferdous et al., 2012; Happel et al., 2013; Jeong et al., 2011; Kim et al., 2018; Lucena et al., 2007; Matheoud et al., 2018; Peekhunthod et al., 2017; Rodríguez-Laguna et al., 2010; Shoji et al., 2007) from exposed workers, and b) $^{99}$Tc (Barrera et al., 2016; Eichrom Technologies, 2014; Khan and Um, 2015; Villar et al., 2015, 2013) and $^{131}$I (Demir et al., 2013) from treated patients. In addition, $^{131}$I is also excreted in saliva, sweat and in air exhaled by the patient (Gründel et al., 2008; Guillot et al., 2016; Hamizah et al., 2012; International Atomic Energy Agency (IAEA), 2009; Schomäcker et al., 2011). So far, however, these patient samples have been investigated in very few studies, although the results obtained should be taken into consideration since they may provide valuable information as regards setting radiation safety precautions.

The following sections (3.1 and 3.2) contain a discussion of the different approaches found in the recent literature for determining different medical radionuclides in biological samples. A selection of the most significant studies is given in Table 5, which includes details of the collection, pretreatment and measurement strategies commonly used for biological samples, along with the radiological content in terms of activity concentrations of the different radionuclides found in the samples.

### 3.1. Biological samples from NM workers

The analysis of biological samples, mainly urine, from workers involves individual internal monitoring, the results of which provide information about a) estimated intake, by using the corresponding biokinetic model, and b) the corresponding committed effective dose (Breustedt et al., 2018). ISO 20553 (2006) sets out the need to perform this assessment on those workers who are exposed and receive a relevant dose from unsealed radionuclide intake that is equal to or greater than 6 mSv (Bingham and Etherington, 2018).

The most commonly used bioassays in the literature involve similar steps: a) sample collection, and b) measurement.

The collection of urine samples is a crucial step and there are three important factors to consider, as recommended in the report on the *Medical monitoring of occupational internal exposure to radionuclides in nuclear installations* (French Occupational Health Medicine Society, 2011): a) it should be carried out in uncontaminated areas to avoid sample contamination, b) the volume of the sample should be recorded, and c) the total activity should be assessed per unit of time from the provided sample. In this regard, ISO 16637:2016 (2016) recommends the collection of 24-h urine samples in order to carry out an accurate dose assessment based on daily excretion. Some of the studies reviewed do indeed apply this recommendation (Happel et al., 2013; Lucena et al., 2007; Unpublished results, J. Martínez; Peekhunthod et al., 2017). However, mainly due to technical considerations this practice is not usually carried out and the usual procedure is to collect spot samples (Bitar et al., 2016, 2013; Ferdous, 2016; Ferdous et al., 2012; Unpublished results, J. Martínez; Matheoud et al., 2018; Noh et al., 2016; Shoji et al., 2007). This provides valuable information as regards internal occupational contamination at a particular point in time. These urine samples are commonly taken three to 4 h after handling the relevant radionuclide or once a month, depending on the radionuclide and the aim of the monitoring study. The results obtained, in terms of activity concentrations, are not representative of the whole radionuclide intake, and therefore the specific radionuclide activity needs to be normalized. Two common normalization methods are used for this purpose by the different authors: a) by creatinine content, or b) by volume (European Commission, 2018; Paquet et al., 2015). However, an interesting study published recently by Noh et al. (2016) used urine spot samples without normalization. The main advantage of this procedure is that it is hoped it will serve to monitor internal occupational exposure to $^{18}$F and other short half-life radionuclides by using

*in vitro* bioassay methodology and the newly developed biokinetic models, thereby avoiding the task of sample normalization.

Measurement methods are generally based on gamma-ray spectrometry, which is used to detect $^{131}$I, $^{99m}$Tc and $^{18}$F in urine samples, and a positive aspect of this is that no pretreatment is needed (Bitar et al., 2016; Díaz-Londoño et al., 2017; Ferdous, 2016; Ferdous et al., 2012; Jeong et al., 2011; Kim et al., 2018; Matheoud et al., 2018; Rodríguez-Laguna et al., 2010; Shoji et al., 2007). However, some studies in the literature have applied homogenization and acidification with nitric acid to minimize precipitation and prevent bacterial growth (Peekhunthod et al., 2017). The HPGe detector (Bitar et al., 2016, 2013; Ferdous, 2016; Ferdous et al., 2012; Jeong et al., 2011; Lucena et al., 2007; Unpublished results, J. Martínez; Noh et al., 2016; Peekhunthod et al., 2017) is the preferred choice, although the NaI scintillator detector has also been employed (Matheoud et al., 2018; Peekhunthod et al., 2017). In a novel approach, Díaz-Londoño et al., 2017 and Rodríguez-Laguna et al. (2010) took advantage of the gamma cameras present in NM departments to quantify the radionuclide activities of workers' urine samples and to self-assess internal occupational exposure. Both studies used this equipment without the presence of collimators so as to achieve maximum sensitivity and were able to achieve detection of the annual effective dose limit of 1 mSv. This measurement approach is also advantageous because samples can be analysed *in situ*, meaning they do not need to be sent to external laboratories, thus reducing analysis time.

$^{18}$F, $^{99m}$Tc and $^{131}$I have been found in various studies in which NM workers' urine has been analysed. In these studies $^{131}$I activity levels vary greatly between workers, ranging from 1.2 Bq/L (Lucena et al., 2007) to a maximum of 390.0 Bq/L (Ferdous, 2016). This variability in terms of $^{131}$I activity concentrations depends on a number of aspects including the radionuclide activity administered, the protection measures undertaken and on the rate of urine excretion, which varies between individuals (Kim and Whang, 2009), among others. Studies into $^{99m}$Tc in urine also illustrate this variability in their results, which range from 8.6 Bq/L (Ferdous et al., 2012) to 18652.0 Bq/L (Unpublished results, J. Martínez). Martínez et al. (Unpublished results), for example, evaluated $^{99m}$Tc activity in different NM workers performing different tasks in an NM department. They concluded that those NM nurses involved in the $^{99m}$Tc-based diagnostic study who were present when the radionuclide activity in air reached its maximum presented the highest $^{99m}$Tc activity in 24-h urine samples, as expected.

### 3.2. Biological samples from treated patients

Those patients undergoing exposure due to medical diagnosis or treatments are in themselves a potential radioactive source. In this regard, a patient's body fluids may cause radiological impact on a) the environment, due to the discharge of radioactive urine and faeces, mainly $^{99}$Tc, into the sewage system, and b) caregivers, medical staff and the general public, due to their exposure to the $^{131}$I present in the patient's urine, saliva, sweat or exhaled air.

$^{99m}$Tc is used in approximately 80% of the diagnostic studies performed worldwide. This short half-life radionuclide decays to $^{99}$Tc, which has a long half-life ($T_{1/2} = 2.13{\cdot}10^5$ years) and high mobility in the environment (Barrera et al., 2016; Clases et al., 2018). The reviewed studies from the literature focus on the presence of $^{99}$Tc in patients' urine. Since $^{99}$Tc is a beta emitter, the radioanalytical methods used commonly involve a pretreatment step to isolate it from the matrix before measurement (Barrera et al., 2016; Eichrom Technologies, 2014; Khan and Um, 2015; Villar et al., 2015, 2013). Eichrom Technologies present a procedure based on the use of an extraction chromatographic resin for $^{99}$Tc separation – TEVA resin in disk format (Eichrom Technologies, 2014) – but it is a manual strategy and time-consuming. With the aim of overcoming the drawbacks associated with such an approach, several attempts based on flow analysis techniques have appeared in recent literature, and these have been used to automate the strategies to provide faster, greener methods and improve the analyst's safety (Villar et al., 2015, 2013). In this regard it is important to highlight that a review setting out and comparing the most frequently used automated extraction and preconcentration strategies has been published (Rodríguez et al., 2016). There are various studies that use these types of automated tools for $^{99}$Tc determination in urine samples, such as the two reported by Villar et al. (2015, 2013). In the first of these the authors combined a solid phase extraction (SPE) based on TEVA resin with a lab-on-valve system (Fig. 3, a) (Villar et al., 2013) to isolate and preconcentrate the radionuclide. In order to avoid backpressure problems due to SPE, the second study was based on lab-in-syringe dispersive liquid-liquid microextraction (DLLME) (Villar et al., 2015). This study critically tested and discussed extractants and dispersers for DLLME, concluding that the optimal extractant was Aliquat®336 due to its high $^{99}$Tc extraction efficiency, while the best dispersant was acetone due to its cheapness and the fact that it provides higher reproducibility than others.

Barrera et al. (2016) (Fig. 3, b) recently presented an approach based on the use of plastic scintillation resins (PSr), a strategy that unifies the radionuclide separation and the scintillation measurement, thus reducing analysis time, sample handling and reagent consumption. In the case of $^{99}$Tc isolation, the PSr employed was a combination of plastic scintillation microspheres with Aliquat® 336 as the extractant, which also acts as a scintillator agent reducing the generation of organic waste. Although this is not a novel methodology, further research into the subject would broaden its application to other radionuclides (Tarancón et al., 2017).

Although almost all the strategies found in the literature aiming to determine $^{99}$Tc in urine samples need sample pretreatment, Khan and Um (2015) have developed an interesting, fast, economical method to measure urine samples without radionuclide isolation and sample purification. They use a) a ULG-LLT liquid scintillation cocktail for urine and samples with a high degree of quench, and b) ULG for cleaner samples. This strategy also reduces the precount delay time published in automated methods (Villar et al., 2015, 2013), from 12 h to 2 h.

All the strategies mentioned used LSC to determine $^{99}$Tc in the processed samples.

$^{131}$I has also been determined from different biological samples from treated patients. In this case the most frequently analysed samples are sweat on skin (Guillot et al., 2016; Hamizah et al., 2012), saliva (Guillot et al., 2016) and urine (Demir et al., 2013). The reported sample collection strategies depend on the type of sample, i.e. direct collection in the case of urine, a wipe test for sweat on the skin surface and Salivette© for saliva. The sampling is usually performed some time after the $^{131}$I dose is administered, in the range between 4 h and 120 h. In none of these studies was pretreatment performed, and the samples were measured directly by the use of the different detection systems usually present in NM departments, such as a well counter (Demir et al., 2013), HPGe and activimeter (Guillot et al., 2016) and thyroid uptake probe (Hamizah et al., 2012). The highest activities for $^{131}$I, in the order of magnitude of MBq, have been found in urine and saliva samples.

Another sample of concern is the patient's exhaled air, since caregivers or family members could be exposed to internal contamination via the inhalation pathway. In this case samples have been directly collected from the patient (Gründel et al., 2008; Schomäcker et al., 2011) with the aim of determining the total $^{131}$I activity and also to investigate the activity related to different exhaled iodine compounds. The reviewed studies that focus on this issue have therefore flowed the air sample through modular support systems. Gründel et al. (2008) collected the exhaled air directly from patients on different adsorbents placed in a modular system that can be adapted depending on the aim

of the iodine species separation, as shown in Fig. 4, a. The maximum exhaled concentration was $2.66{\cdot}10^{-5}$ Bq/m$^3$. Similarly, Fig. 4, b illustrates another collection strategy reported in the literature (Schomäcker et al., 2011). In this case patients filled fifteen balloons, the air from which was then conducted by an air pump into a modular filter system. From both studies it was concluded that the proportion of organic iodine species exhaled by a patient was greater than 90% of the total iodine present in the air sample.

## 4. Conclusions

Despite the current good practices applied by hospitals with NM departments, several research studies in recent years have demonstrated the presence of medical radionuclides, mainly $^{99}$Tc, $^{99m}$Tc and $^{131}$I, in environmental and biological samples. This review compiles and summarizes the most recent developments in determining these radionuclides, in almost all cases using gamma-ray spectrometry and liquid scintillation counting.

As regards the samples tested, in order to successfully analyse these radionuclides in a variety of matrices, different collection and pretreatment strategies have been developed.

In the case of indoor air samples, sampling approaches based on AS have usually been performed, but future trends should focus on the application of real-time strategies comprising both collection and detection steps. This would allow for the determination of short half-life radionuclides, the detection of activity fluctuations and possible evaluation of the occupational dose received in real time.

The existing literature based on biological samples, with urine being the most commonly used matrix, includes automated pretreatment strategies to determine $^{99}$Tc so as to minimize waste generation and avoid sample handling. However, further work is needed to develop automated tools suitable for analysing different types of matrices, such as hospital wastewaters.

Finally, bearing all of this in mind, the future challenge is to develop collection, pretreatment and detection strategies to enable the determination of medical radionuclides that are sensitive enough to assess potential internal occupational contamination and to monitor environmental impacts due to medical radioactive releases.

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