Environmental impact assessment of waste-water: radionuclides use in hospitals (Abruzzo, Italy, 2000-2015)

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Radioactive contamination,
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Waste-water,
Gamma spectrometry.

Summary
Radionuclides are increasingly used in hospitals for diagnostic and therapeutic purposes, such as functional research, diagnostic imaging, and in the performance of radioiodine therapy. Their use produces radioactive waste, and risks environmental contamination. The present study involves 486 samples of radioactive waste produced in hospitals in Abruzzo, Italy, during 2000-2015. Measurements were carried out with the gamma spectrometry technique: Germanium detector (PTG) with 8,000 acquisition channels, power range 59-1,836 keV, resolution 1 keV, overall efficiency 30%, measurement time 60,000 Seconds. The radionuclides involved were as follows: ^{131}I, ^{99m}Tc, ^{67}Ga, ^{201}Tl, ^{123}I, ^{111}In, with substantial activity in 44 samples. Checks allowed us to certify the levels of radioactive concentration in waste-water, plan for their suppression, and optimise the management procedures.

Valutazione dell'impatto ambientale delle acque di scarico: l'uso dei radionuclidi negli ospedali abruzzesi, 2000-2015

Parole chiave
Contaminazione radioattiva,
Medicina nucleare,
Radionuclidi,
Acque di scarico,
Spettrometria gamma.

Riassunto
Negli ospedali i radionuclidi sono utilizzati sempre di più per la ricerca funzionale, la diagnostica per immagini e nella radioiodio terapia. Il loro uso produce scorie radioattive e rischi di contaminazione ambientale. Questo studio esamina 486 campioni di scorie radioattive prodotte negli ospedali abruzzesi tra il 2000 e il 2015. Le misurazioni sono state effettuate con la tecnica della spettrometria gamma: rivelatore al germanio (PTG) con 8000 canali di acquisizione, gamma di potenza 59-1.836 keV, risoluzione 1 keV, efficienza complessiva 30%, tempo di misurazione 60.000 s. I radionuclidi coinvolti sono i seguenti: ^{131}I, ^{99m}Tc, ^{67}Ga, ^{201}Tl, ^{123}I, ^{111}In, attivi principalmente in 44 campioni. I controlli hanno permesso di certificare i livelli di concentrazione radioattiva nelle acque di scarico, pianificare la loro eliminazione e ottimizzare le procedure di gestione.

Introduction

The use of radioactive substances, sealed and unsealed, in hospitals for diagnostics and therapy produces solid, liquid, and gas forms of radioactive waste.

In Italy, according to the Euratom directives, Legislative Decree 17/03/1995 n. 230[1] and its subsequent changes and additions are designed to ensure high levels of protection for public health and the environment.

The objectives of the present study were to investigate the radioactive waste generated by Nuclear Medicine hospitals departments located in Abruzzo, particularly the radionuclides used in unsealed form: ^{131}I, ^{99m}Tc, ^{67}Ga, ^{201}Tl, ^{123}I and ^{111}In, according to a previous study by Piersanti and colleagues (Piersanti *et al.* 1992a).

The radionuclides examined have a T½ (Half-life)

[1] Legislative Decree 17/03/1995 n. 230. Attuazione delle direttive 89/618/Euratom, 90/641/Euratom, 96/29/Euratom e 2006/117/Euratom in materia di radiazioni ionizzanti. *Off J*, **136**, 13/06/1995 (S74).

less than 75 days and are categorised as 'waste with other hazardous characteristics - short-lived radionuclides' in accordance with art. 154, Legislative Decree n. 230 of 17/03/1995, in which there is no reference to sealed radioactive sources. These sealed radioactive sources are used in cell sterilising processes, in calibration sources, and for the calibration of radiation therapy performance as, for example, of ^{60}Co sources. The use of these sources does not produce, as a rule, radioactive waste; and the procedures are strictly disposed of through authorised companies.

Unsealed radioactive substances are used in functional studies and in diagnostic imaging techniques; in liquid form are also employed for in vitro radiommunoassay investigations. Radioactive substances in liquid or solid form are administered to patients during the performance of radiometabolic therapy.

Solid radioactive waste includes: supply tanks for individual radio-labelled solutions, contaminated material following elution operations, administration, and investigation; contaminated test tubes, syringes, needles, blotting paper, work gloves; and any other potentially contaminated materials due to patients, workers, and environmental applications. Among solid wastes potentially contaminated, there are also those originated from radiometabolic therapy.

According to operating procedure rules, radioactive waste is managed in special storage tanks, by specialised companies that, in compliance with rules, are required to issue a statement for the withdrawal of materials as well as its final destination.

Radioactive waste in liquid form is originated by cleaning surfaces of controlled zones, such as controlled sinks, areas of radioactive substances use, zones of decontamination of patients and workers. It can further be found in the urination and excreta of patients, and in controlled baths. Liquid waste resulting from radioimmunology investigations is placed in a specific container and managed in the same way as solid waste.

Normally, the hospital activities listed above do not constitute an appreciable source of radioactive waste in gaseous form, however it is necessary to periodically test air conditioning dilution cells and filters.

The Nuclear Medicine hospital Unit, in Abruzzo Region, are equipped with a radioactive waste management system that allows potentially contaminated liquids to flow into septic tanks, so-called 'imhoff tanks' for primary sedimentation, and in other tanks to collect liquid radioactive waste. Local and remote control checks allow to verify the management systems and monitor the levels of each tank.

In diagnostic activity, the main use of radionuclides is mostly linked to medium-short-lived radionuclides such as ^{99m}Tc (T½ 6 h) and ^{111}In (T½ 2,8 d), which in the case of outpatient therapy involvs the ^{131}I (T½ 8 d) and any other radionuclides.

A specific waste management system is implemented in case of radionuclides administered for therapeutic purposes during hospitalisation. It includes 'imhoff tanks' for primary sedimentation and tanks for the collection and decay of liquid waste, in order to optimise the process for liquid radioactive waste involving the use of radionuclides with longer average life which the ^{131}I (T1/2 8d) and higher radioactive concentrations. All organic waste from patients, washing liquids, and decontamination coming from radionuclides administered for therapeutic purposes is collected in tanks.

It is crucial to evaluate the appropriate size of tank systems in order to ensure the efficiency of hospital structures that are implicated in nuclear medicine in relation to costs, number of patients to be treated and quality of services. It also depends on the concentration and quantity of radioactive substances that have been used.

Hospitalised patients who are under diagnostic investigations of the Nuclear Medicine Unit, reported that liquid waste and contaminated excretions were released into the hospital sewage system. However, the controlled baths used in Nuclear Medicine Unit lower the amount of radioactive substances released into the sewer system through liquid waste such as urination.

This type of waste disposal in the environment, particularly for liquids, is supported within the exemption terms of article 154 c. 2 (Legislative Decree 17/03/1995 n. 230). In particular:

- stakeholdes radioactive isotopes must have a T½ of less than 75 days;
- concentration must be less than 1 Bq/g (ref. Art. 1 and Annex 1 to Legislative Decree. 230/95) for a single radionuclide and the mixture of several radionuclides;
- disposal must occur in accordance with Decree. N. 22 of 5 February 1997.

Methods and procedures relating to disposals must be recorded and forwarded to the supervisory bodies.

There is therefore a need to both verify and certify the radioactive concentration of the materials found in the tanks of liquid waste and septic tanks before leaving any waste in the environment, as well as a need to periodically review the radioactive concentration in the hospital sewer system derived from patients using services outside of controlled environments.

The aim of the paper is the environmental impact of waste-water in Abruzzo hospitals.

Materials and methods

This study involved 486 waste-water samples conferred by hospitals to Istituto Zooprofilattico Sperimentale dell'Abruzzo e del Molise 'G. Caporale' (IZSAM) during 2000-2015, in the framework of an hospital self-monitor on the concentration of radioactivity in the tanks, and before they were disposed in the outer sewer system.

Samples were received in the laboratory accompanied by a cover sheet with the following information: applicant's name, date, amount of sample taken, type of material, mode of transport (ADR Code UN 2910), packaging of the sample, and specifications of the radionuclides to be analysed.

The removal of effluent materials was carried out by the hospitals, directly from storage tanks and storage and the sewage outlets, and before they were placed into the public sewer.

The IZSAM lab, which was responsible for carrying out the analysis of Bromatology and residues in foods for human and animal consumption, is part of the RESORAD Network for radiation monitoring and participates in reliability programmes organised by the Italian National Institute for Environmental Protection and Research (ISPRA) (Calvarese *et al.* 1997, Calvarese *et al.* 2009, De Crescenzo 2012, ENEA 1991, Piersanti *et al.* 1992).

This study covered concentrations of following radionuclides that were used by hospitals: ^{131}I, ^{99m}Tc, ^{67}Ga, ^{201}Tl, ^{123}I, ^{111}In.

One-litre samples were placed in a Marinelli beaker to proceed to the gamma spectrometry in order to determine the activity of the involved radionuclides. The shielding of measuring wells was 10 cm of lead. Samples were measured for 60,000 seconds.

Until 2006, measurements were carried out with a germanium detector (PGT) with an overall efficiency of 25% with 4,000 acquisition channels. Since 2007, High Purity Germanium detectors (HPGe) (Ortec®, South Illinois, USA) with a total efficiency of 30% and 8,000 acquisition channels have been used. The detection energy range was between 59 to 1,836 keV. Emission lines were identified with a resolution of 1 keV.

Instrument calibrations were performed with aqueous sources in metrological centres certified by the National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA) until 2006, and subsequently by Deutscher Kalibrierdienst (DKD).

To calibrate sources it was used a mixture containing the following gamma-emitting radionuclides: ^{241}Am, ^{113}Sn, ^{109}Cd, ^{85}Sr, ^{57}Co, ^{137}Cs, ^{139}Ce, ^{60}Co, ^{203}Hg, ^{88}Y, with calibration energy from 59.5 to 1,836 keV.

The following software were used for the evaluation of spectra: Up until 2006, Range 2000 (Silena International S.p.A., Cernusco Sul Naviglio, Italy), and since 2007, GammaVision 32 (Ortec®).

Tests for the detection of gamma-emitting radionuclides were adopted in accordance with ISO/IEC 17025 .

Check results were sent to requiring hospitals so that they could organise the unloading of liquid waste tanks and septic tanks in accordance with local regulations.

In the laboratory, samples were managed in compliance with the standards for protection, safety, and working instructions on radioactive waste management, and according to the accreditation process. Samples detecting an exceeding activity beyond the limits of the Legislative Decree 17/03/1995 n. 230, have been stored for physical decay.

Results

Between 2000-2015, 486 waste-water samples were analysed. The significant increase in analysis during 2009-2015, reflects the period in which the Nuclear Medicine Unit activity increased significantly (Figure 1).

Table I reports the distribution of single radionuclide and findings for the distribution of activity, for each sample, of any implicated radionuclide. Forty-four samples contained values that require attention, as the sum of the concentrations of the radionuclides that were analysed was close to the critical value. We detected radioactive concentration values between 1,000 and 10,000 Bq/l in 18 samples; in two samples found in 2015, values of between 10,000-50,000 Bq/l

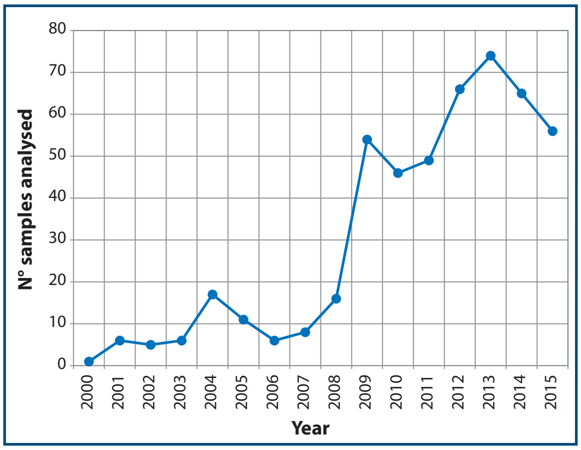

Figure 1. *Number of waste-water samples analysed by IZSAM.*

Table I. *Number of samples analysed by IZSAM.*

Activity range		N. of analysed samples						
≥ Bq/l	< Bq/l	Activity in the range for single radionuclide						for total radionuclides
		^{131}I	^{99m}Tc	^{111}In	^{67}Ga	^{123}I	^{201}Tl	
0	1	331	377	425	70	51	338	3
1	5	43	55	17	386	67	118	17
5	10	15	27	4	21	243	13	154
10	50	51	12	14	8	120	12	214
50	100	17	3	8	1	4	4	33
100	1,000	17	8	13	0	1	1	44
1,000	10,000	9	4	5	0	0	0	18
10,000	50,000	2	0	0	0	0	0	2
50,000	100,000	1	0	0	0	0	0	1

for ^{131}I were detected, and in a sample dated 2013, a value of 70,184 Bq/l always for ^{131}I was detected.

Discussion and conclusions

Sundell-Bergman and colleagues (Sundell-Bergman *et al.* 2008) report on the complexity of radioactive waste management in Sweden. The risk of radioactivity spreading through the sewer system and the subsequent use of sludge in agricultural land has been highlighted in this study,.

Another study of Krawczyk and colleagues (Krawczyk *et al.* 2013) shows as the storage of waste water in tanks to control the concentration of radionucluids prevents the dispersion of radioactivity into the environment. This conclusion descends from studies carried out in two large public hospitals in Granada (Spain) with gamma spectrometry investigations.

Barquero (Barquero *et al.* 2008) carried out a study on the evaluation of exposure for'worker[s] assigned to the treatment of waste water' in a hospital that utilises radioactive substances in Nuclear Medicine.

The results of this study demonstrate that the control of the radioactive concentrations of potentially contaminated liquid waste from hospitals is crucial.

The checks that we carried out have allowed us to certify the levels of radioactive concentration in the effluent resulting from the storage tanks from the hospital and, going forward, to plan for the elimination of waste, in accordance with local regulations and limits.

In several cases, significant activity was detected. This prompted new evaluations, including a methodological review of procedures in order to improve internal performance of the hospitals and environmental safety.

Since 2000, IZSAM has confirmed the following needs:

- a comprehensive system for radioactive waste (liquid) management with storage tanks and decay;
- a specific assessments not only limited to theoretical calculations, but based on measurements made with dedicated instrumentation and procedural protocols.

Radioactive waste concentration control in hospitals promotes the management of risk for workers, populations, and the ecosystem in general.

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