Presence of radioactivity in a sewage system:
A proposal for radioactivity control

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Abstract. Most hospitals use radioisotopes in diagnostics and to a lesser extent in therapy. The liquid residues thus generated are usually subjected to treatment before being discharged into the sewage system. Nevertheless, a certain amount of these residues escape from the treatment system and are poured directly into the sewer. In addition, other radioactive products used for research and industrial purposes may also be disposed of in the same way.
The waste waters in many sewage systems can therefore be expected to be radiologically contaminated and the need for at least a basic control system in such situations seems obvious.
When designing a procedure to measure radioactivity, certain conditions should be borne in mind:

- The control program has to be simple and inexpensive.
- Samples must be taken from the appropriate places.
- Short life radionuclides will probably be present in significant amounts, so that specific recipes should be prepared.
- Iodine is also frequently present. Special precautions should be taken to keep it in solution.

In recent years, the Environmental Radioactivity Laboratory of the *Universidad Politécnica de Valencia* (Spain) has carried out a series of tests on the Valencia city sewage system and sewage treatment plant in order to design a permanent program to control radioactive contamination of the city's sewage system.
This paper presents a proposal which we believe can provide the answer to this problem.

KEYWORDS: waste water, radioactivity, contamination.

1. Introduction

A clean environment is a whish for everybody and also a need for a healthy life and even is a legal requirement. Air, water and food are exhaustively monitored to detect the presence of chemical, biological or radioactive pollutants, but this is not the case for waste water systems.

Radioactive isotopes are widely used for different purposes in medicine, research and industry. Some of these activities use isotopes in the liquid phase and residues are often discharged into sewage systems, in which case they can be expected to contain radioactive pollutants. [1]

This pollution will affect the end products of sewage treatment plants. As these products are often recycled, radioactivity control should be a requirement.

This paper presents the results of previous studies carried out on a hospital waste outlet and in the Valencia sewage system and the results of a study on a Sewage Treatment Plant (STP) with the objective of designing an easy, effective and economically affordable radioactive control program.

2. Where should samples be taken?

Once recognized the interest of the presence of radioactive pollutants, the question arises of where the samples should be taken for analysis.

Hospitals with nuclear medicine service use radioactive isotopes for diagnosis and treatment with residues being discharged into public sewage systems. There is therefore a clear need for such systems to be monitored to avoid excessive levels of radioactivity.

As radioactive wastes are discharged into sewage systems at irregular intervals, sampling has to be continuous in order to get a representative value over a certain time period. Each hospital needs a surveillance program and it should also be remembered that hospitals may not be the only source of radioactivity in the system.

Radioactivity surveillance of the sewage system should be able to control the radioactive pollutants no matter where they come from, but again continuous sampling should be used at a number of points in the system. Should this type of arrangement prove too difficult or too costly to carry out, consideration can be given to focusing attention on the sewage treatment plant (STP).

As all waste waters converge in the sewage treatment plant, a radiological characterization of this point will make it possible to identify the best place to take samples.

The STP studied for the purpose of the present work is situated in Pinedo, Valencia (Spain). It was designed for a population of approximately 600,000 inhabitants, and employs a mixture of different treatments (physical-chemical and biological) [2]. Fig. 1 shows the flow diagram corresponding to the Pinedo STP. The flows corresponding to the water line are blue, while those of the sludge line are brown. Both entrance flows and mean residence times of each of the purification phases are indicated.

The following materials can be identified in the complete treatment process [3,4]:

(a) Water line

— Very large, large and fine solids
— Grit and greases
— Raw water
— Settled water
— Treated water

(b) Sludge line

— Thickened sludge from primary sedimentation
— Thickened sludge from secondary sedimentation
— Sludge from primary digester
— Sludge from secondary digester
— Dewatered sludge

Not all of these materials were characterized, some for obvious reasons given their nature (very large and large solids, grits and greases) and others because they only formed a small part of the sedimentation process (thickened sludge from secondary sedimentation). Only the secondary sludge from the digesters was analyzed as it was produced at the end of the digestion process.

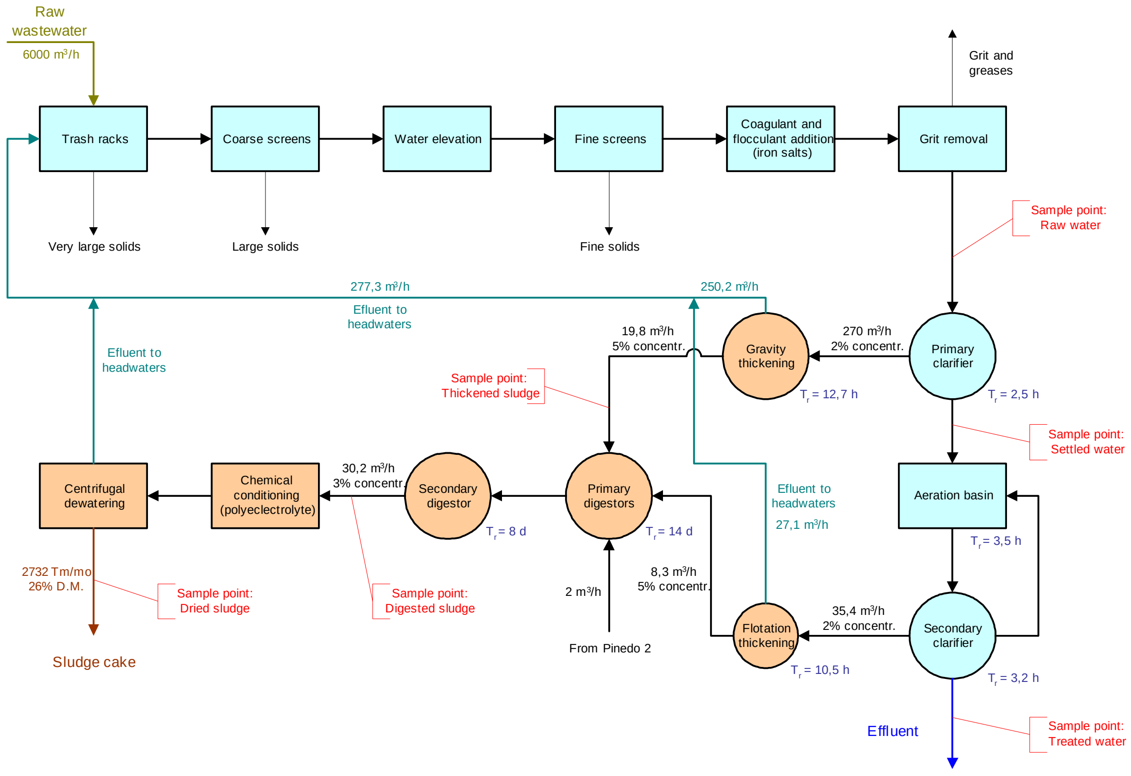


Figure 1. Flow diagram of the Pinedo STP.

Fig. 1 shows where the different types of samples analyzed were collected. These points were chosen to permit identification of radioemitters present in the different stages of the treatment. Given the origin of the pollutants (waste from homes, industries and hospitals), wide variations of the pollutant concentrations in time are to be expected.

Nonetheless, the waste water cleaning process concentrates pollutants in certain materials which at the same time present a long mean residence time in a particular stage of the process [5]. This is a considerable help in finding a good sampling point.

3. Preparation of samples

Water samples from the sewer network were collected discontinuously, while samples from hospitals outlets were taken continuously. Water samples (raw, settled and treated) from the STP were taken continuously for 24 hours. Those corresponding to the analyzed sludge (thickened, digested and dried) are individual samples. Thickened and digested sludge are liquids with high solids content (5-3%), while the dried sludge samples are solids with a moisture content between 18 and 30%.

On arrival in the laboratory, before being subjected to treatment, water samples, thickened and digested sludge were sterilized with formaldehyde (20 ml/l) in order to avoid biological contamination [6]. Samples were then prepared as follows:

(a) **Water samples**: The sample is sterilized and homogenized and a litre is poured into a Marinelli beaker for the measurement of short period emitters for 1 hour. Another litre is evaporated in an oven at 80ºC on a plastic sheet which is folded according to a protocol and placed in a 90 cc Petri beaker.

(b) **Thickened and digested sludge**: formaldehyde is added and the sample is homogenized. One litre of sample is measured in a Marinelli beaker for 1 hour. The remainder of the sample is dried at 110ºC to constant dry weight, then ground and placed in an appropriate laboratory ware for its measurement.

(c) **Dried sludge**: The entire sample is dried in an oven at 110 ºC to constant dry weight, then ground and placed in an appropriate geometry for measurement.

Measurements of the gamma-ray emitting nuclides were performed with a multichannel analyser equipped with a Ge(HP) detector. A library including isotopes used in medicine and industry was used.[7]

4. Measurements and comments

During a preliminary study of the sewage system and liquid waste from hospital outlets, multiple samples were taken from various points and analyses performed in order to characterize the waste waters in the sewage system of Valencia (Spain) [8,9,10].

Table 1 shows the results of samples from a hospital outlet. Data correspond to samples collected continuously during a period of 8 hours. As can be expected, the lower Tc-99m and ^{131}I values belong to the overnight period.

Table 1. Maximum values activity (Bq/m^3) from a hospital outlet.

Period of time	^{99m}Tc	^{131}I
14,00-23,30	(6,53±0,16)E+05	(2,46±0,04)E+04
0,00-7,30	(2,92±0,24)E+04	(1,17±0,02)E+04
7,30-13,30	(6,97±0,08)E+05	(1,35±0,02)E+04

Table 2 presents maximum values for ^{131}I, ^{99m}Tc, ^{67}Ga and ^{111}In found during the characterization of the whole sewage system. ^{99m}Tc shows a relatively high value, but it has to be remembered that these values correspond to discontinuous sampling, while the great majority of the values found in samples taken from the sewage system are much lower and frequently zero.

Table 2. Maximum values found in samples from sewage system (Bq/m^3).

Isotopes	North Sewage system	South and West Sewage system
^{131}I	(1,75±0,08)E+05	(13,07±0,02)E+05
^{99m}Tc	(5±2)E+07	(17,36±0,14)E+05
^{67}Ga	(1,91±0,09)E+04	(4,30±1,37)E+03
^{111}In	(6,98±0,49)E+02	(3,30±0,89)E+02

As can be expected, the waste water flowing through the sewer lines show permanent radioactive pollution.

As all sewer lines converge in the sewage treatment plant; this is probably the right place to locate radioactivity control. This kind of plant presents a wide variety of locations to be monitored. Thus, as a first step, simultaneous daily samples were collected from the different stages of the process. Table 3 shows the mean activity values of some radionuclides frequently used in nuclear medicine for five days of the week.

Table 3. Mean activity values (Bq/m^3)

Samples	Isotope	Monday	Tuesday	Wednesday	Thursday	Friday
Raw water	^{131}I	121	4912	4082	2192	1345
	^{99m}Tc		12983	12070	9180	6446
Settled water	^{131}I	148	2412	5124	2097	1391
	^{99m}Tc		7696	6459	6340	5540
Treated water	^{131}I	193	3026	3159	2072	1345
	^{99m}Tc		2486	3062	4381	2637
Thickned sludge	^{131}I	5650	6832	3636	7264	6188
	^{99m}Tc		290345	256700	233010	212355
	^{67}Ga	1695	4078	3955	7960	8820
	^{111}In	216		183	23774	3341
Digested sludge	^{131}I	5644	5879	5924	5582	4871
	^{67}Ga	771	670	821	628	548
	^{111}In	183	139	112	86	-
Dewatered sludge (Bq/Kg)	^{131}I	30.5	44.5	27.2	31.8	33.3
	^{67}Ga	1.7	7.9	2.5	5.3	5.2
	^{111}In	-	2.8	0.4	1.0	0.8

Figure 2. Weekly evolution of ^{131}I in raw, settled and treated water.

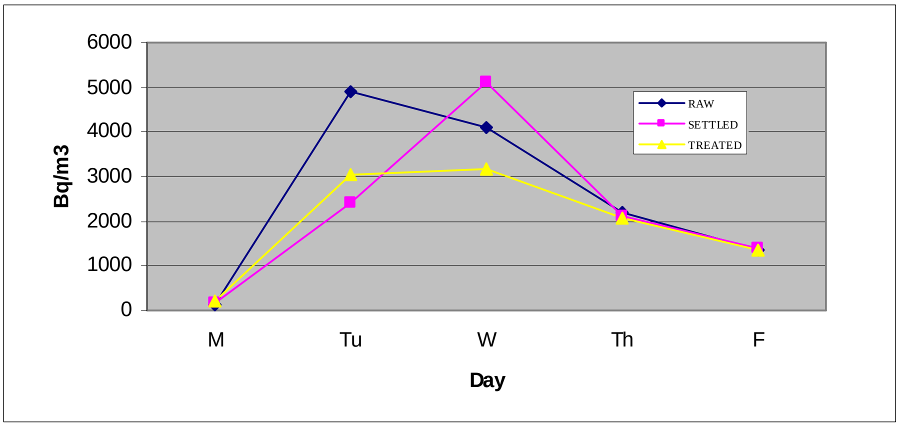


Figure 3. Weekly evolution of ^{131}I in sludge (Bq/m^3).

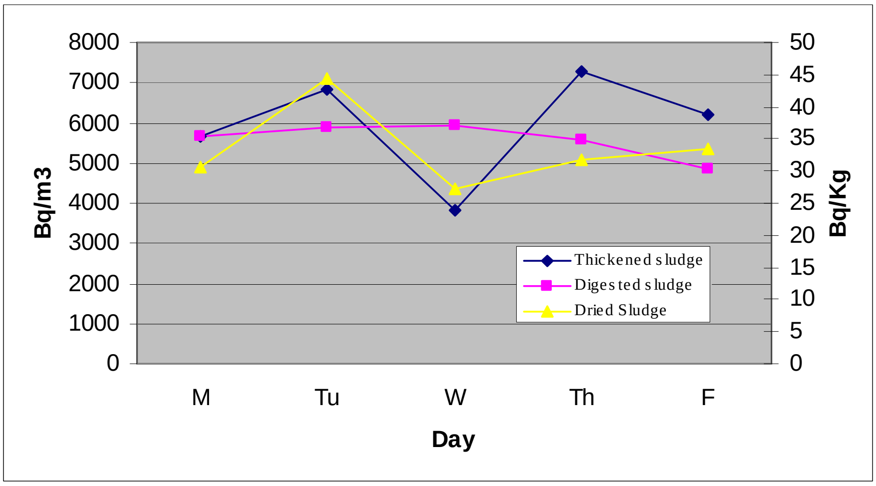


Fig. 2 clearly shows the time evolution of radioactive pollutants in the different kinds of waters, which in all cases present a very low residence value, increasing from the lowest value on Monday, due to absence of activity in the nuclear medicine departments at the weekend, to maximum values on Tuesday and Wednesday, later declining again to values similar to those of Monday. Tuesday and Wednesday will therefore be the best days in the week for taking samples.

Fig. 3 helps us in another aspect to be considered in sampling, which is, where the samples should be taken. The best place will probably be the one which has the longest residence time. Fig. 3 shows that samples of digested sludge (the step with the highest residence time) show very stable values for specific activity.

Of course, the very short-period radioactive contaminants will have decayed. If the program also includes very short-period radioactive emitters, samples should be taken from raw water and analyzed immediately.

5. Proposal of radioactive control program

From all of the foregoing, it can be concluded that it is possible to establish an inexpensive, effective and feasible program to control radioactive contaminants in a city's waste water system. The steps involved in the procedure can be summed up as follows.

- Gamma analysis should be the first step, carried out on samples obtained from different stages of the sewage treatment plant for each day of the week for at least two weeks.
- The day of maximum radioactivity values must be determined.
- Once the day has been determined, a stage of the plant should be selected for sampling. Special characteristics will be taken into account: e.g. ease of sampling and residence time of materials in the step. In our case, the secondary digestor was the stage chosen for taking samples of digested sludge.
- Samples should be taken for measurement on the same day of the week from the same stage of the treatment plant along the year.

The data collected can supply fairly accurate evidence of how radioactive contaminants have evolved in the period studied.

Such a program was established at the sewage treatment plant of Pinedo, Valencia, whose flow diagram is shown in Fig. 1. Samples were taken every Tuesday from the digested sludge tank. After preparation as described in Section 3, samples were gamma analyzed with a high efficiency semiconductor detector.

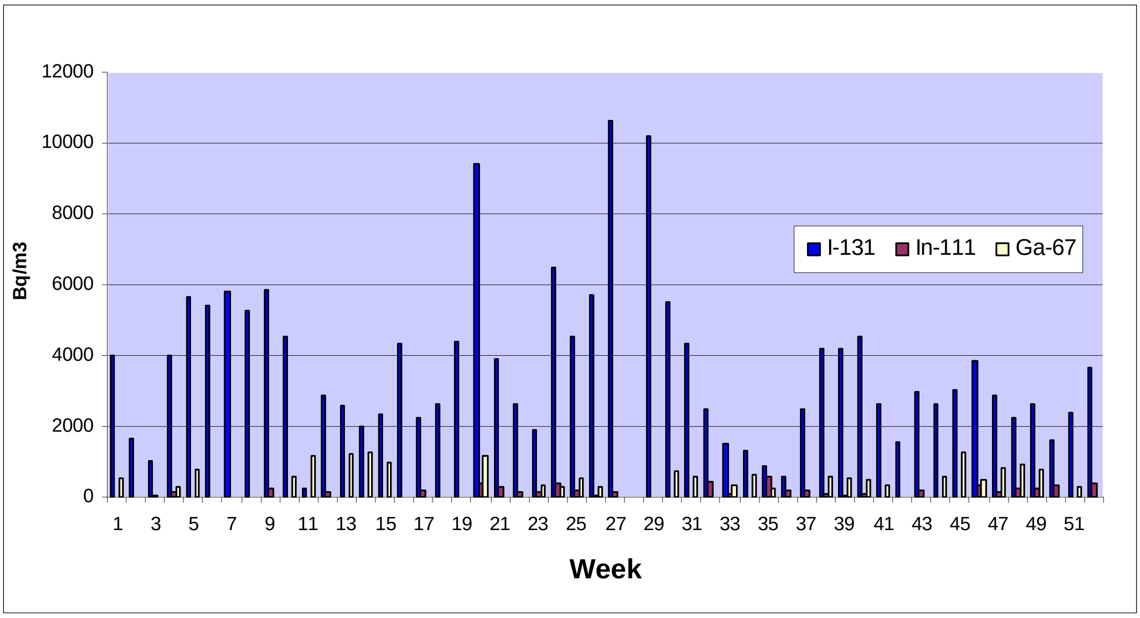
The results throughout 2006 are given in Fig. 4.

The following comments should be borne in mind:

- ^{99m}Tc was not measured. Although it is almost always present, due to its very short period it will be depleted between arrival of waste water at the plant and sampling time.
- ^{131}I is present in all samples, and without doubt, after ^{99m}Tc, is the second most commonly used isotope in Valencia hospitals.
- ^{67}Ga and ^{111}In appear in sample measurements with less frequency and in smaller amounts.
- ^{131}I values in Fig. 4 oscillate mainly between 1000 and 5000 Bq/m^3, although higher values can be observed for weeks 20, 27 and 29. The explanation could be that patients were treated in advance of Easter and summer holidays.

A program like this will detect the presence of radioactive pollutants in waste water provided that the radioactive period is of one day or longer. Shorter periods will be ineffective. Pollutants originating from locations other than hospitals will also be identified.

Figure 4. Evolution of ^{131}I, ^{67}Ga and ^{111}In activity (Bq/m^{3}) during 2006.



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