OPTIMIZATION OF MANAGEMENT OF LIQUID RADIOACTIVE WASTE GENERATED IN RESEARCH AND EDUCATION CENTRES

Macías[1] Mª T., Pérez[2] J., Pulido[2] J., Sastre[2] G., Sánchez[3] A., Usera[4] F.

[1]Instituto de Investigaciones Biomédicas "Alberto Sols" (CSIC-UAM)

[2]Universidad de Alcalá de Henares

[3]Centro de Biología Molecular "Severo Ochoa" (CSIC-UAM)

[4]Centro Nacional de Biotecnología (CSIC)

ABSTRACT

The correct adaptation and disposal of the radioactive waste generated is one of the most important issues in the management of radioactive installations of the research and education centres. Some technical guidelines have been published in Spain with the aim to improve the management of this waste. However, some issues had not been resolved completely. Therefore four important research and education centres started one research project, with a grant of ENRESA (the public company for radioactive waste management in Spain) trying to resolve them. The radiological and chemical characterization of the generated waste has been carried out in the indicated project, and the management model for different waste has been proposed (solid and liquid waste, scintillation vials filled with liquid scintillation and uranyle salt waste).

This work shows the proposed management for aqueous and organic liquid waste considering it as one the most important issues of the project. For aqueous liquid waste, the management protocol proposed clearly shows the necessary calculations to determine the maximum discharge concentration for the aqueous liquid waste that does not exceed the annual limit on intake for ingestion for members of the public. Regarding the organic liquid waste, the discharge via the sewer system is not possible taking into account their chemical composition. In this case the disposal way proposed is the incineration. The management protocol for organic liquid shows the necessary calculations to determine the reference levels applicable to the incineration of this waste than not exceeding the constrain dose of 10 μSv per year in an individual member of the public. This model proposes that the hazardous waste company perform the incineration.The present objective is to show the management model proposed in a technical guide facilitating the management waste to the radiation protection professionals of the research and education field.

KEY WORDS: radioactive liquid waste management, classification, characterization, clearance.

1. INTRODUCTION

The radioactive installations of Research and Education Centres generate residual materials with very heterogeneous radioactive content. The International Agency of Energy Atomic (IAEA)[1] names this waste as low and medium activity waste. The Radiation Protection developments in these installations consider, within different activities, the correct management of these materials to minimize its radiological impact and the possible detriment that may cause the human health and the environment. One of the most important issues is to reduce the volume of the waste, carrying out clearance procedures proposed in different technical reports[2,3,4].

Technical recommendations exist to specify the activity levels of waste in order to establish a quantitative reach to define a radioactive waste[4] considering as such these materials that have no any application [3]. Some technical guidelines have been published in Spain[5,6] with the aim to improve the management of this waste. However, some issues had not been resolved completely[7]. Taking as the base the indicated document[6], the research project "Procedimientos para optimizar la gestión de materiales residuales generados en centros de investigación y docencia" has been performed by four important research and education centres, with a grant of ENRESA (the public company for radioactive waste management in Spain), trying to resolve them. Among its objectives the radiological and chemical characterization of the generated waste in the chosen techniques and the management proposal for different waste (solid and liquid waste, scintillation vials filled with liquid scintillation and uranyle salt waste) have been considered.

This work shows a proposal management for aqueous and organic liquid waste considering it as one the most important issues of this project. Measurement procedures[6] exist, of easy application, for these waste, to quantify its radioactive concentration. However, there is not a clear legal framework in relation to the clearance values compared to solid waste[8]. With the aim to harmonize the work in research and education installations, one methodology has been developed, proposing the release ways for liquid waste in accordance with chemical composition.

2. MATERIAL AND METHODS

2.1. MEASUREMENT PROCEDURES FOR LIQUID WASTE ACTIVITY CONCENTRATION

The liquid waste activity has been measured using scintillation counting techniques, liquid scintillation counting was used to β^- emitters and solid scintillation to γ emitters. The absolute activity and the radioactive concentration were determinate applying the Protocols indicated in the document [6].

2.2 CALCULATION OF REFERENCE VALUES OF ACTIVITY CONCENTRATION FOR LIQUID WASTE

2.21. CALCULATION OF REFERENCE VALUES OF ACTIVITY FOR ORGANIC LIQUID WASTE

Considering the incineration as disposal way of organic liquid waste, this paper proposes reference values for this procedure. Its has been established using as base the study carried out in the Unidad de Protección Radiológica del Público y del Medio Ambiente del Ciemat[7]. In this study were took into account next assumptions:

- This study focuses for H^3 y/o C^{14} waste, given that these waste have some problems for their long half-lives.
- The dose limit established is that the effective dose to individuals should be of 10 μSv per year
- Pathways of exposure considered:
  - Inhalation of discharge material.
  - Ingestion of contaminated food.
  - External irradiation due to the immersion in the gaseous cloud and deposition on the ground.
- Twenty-four hours daily have been considered during which the individual may be exposed, during 365 days of the year.
- The scenery contemplates the following situations:
  - Atmospheric standard conditions.
  - The point of gaseous discharge is 1 m high on a building 10 m.
  - The wind speed is 1 m/s in the direction where an individual lives a 25 % in time.
  - The distance from the gaseous discharge to the individual is 100 m.

2.2.1. CALCULATION OF REFERENCE VALUES OF ACTIVITY CONCENTRATION FOR AQUEOUS LIQUID WASTE

The regulatory body in Spain (Consejo de Seguridad Nuclear –CSN-) limits the annual amount of discharge to the normal sewerage system to a maximum of 10 GBq of ^3H, 1 GBq of ^{14}C and 1

GBq as the sum of the rest of radionuclides, ensuring that in the final point of discharge the activity concentration does not exceed the concentration limits obtained between the ALI_{ing} (age group older than 17 years) and the annual ingestion rate of water for the adult individual, 600 l [9,10].

Taking into account the discharge to the normal sewerage system as final disposal way for aqueous liquid waste, it is necessary to have release concentration values to allow this clearance. This work shows the methodology to determine these values, establishing the maximum limit of activity concentration of the waste to discharge in the initial point of the normal sewerage system, ensuring that if an individual intakes water at the final point of the normal sewerage system, he does not exceed the Annual Limits on Intake (ALI_{ing})

It has been developed on the basis of the CSN consideration, previously indicated, and the criteria proposed for the Grupo de Trabajo de Efluentes del Foro de Protección Radiológica del Medio Hospitalario [11] (SEPR-SEFM), calculating the following parameters:

a) Calculation of Annual Limits on Intake by ingestion (ALI_{ing})

The Annual Limits on Intake by ingestion (ALI_{ing}) has been calculated as the amount of radioactive material that one worker can ingest each year, which will deliver an annual effective dose of 1 mSv. It has been calculated applying the dose coefficient per unit intake for ingestion $e(g)_j$ Sv/Bq for members of the public (age group older than 17 years) for each radionuclide shown as Sv/Bq[12].

$$LIA_{ing} = \frac{1 \bullet 10^{-3}}{e(g)j} \frac{Sv}{Sv/Bq} = Bq \quad \text{Expression 1}$$

b) Calculation of the maximum limit of activity concentration in the final point of the normal sewerage system CV_{max},

Using as starting point the ALI_{ing} previously calculated, and the annual ingestion rate of water, 600 l for adult individual [10], it is possible to calculate the maximum limit of activity concentration in the final point of the normal sewerage system CV_{max}, applying the following expression

$$CV_{max} = \frac{LIA_{ing}}{600} \frac{Bq}{l} \quad \text{Expression 2}$$

This value $\boldsymbol{CV_{max}}$ represents the maximum activity concentration allowed for a liquid intake in an individual to not exceed the ALI_{ing}.

c) Calculation of the maximum limit of activity concentration of the waste to discharge in the initial point of the normal sewerage system (Cv_{PI}).

This concentration can be calculated applying the following expression:

$$Cv_{PI} = \frac{CV_{max} \times (V_c + V_{ec})}{V_c} \quad \text{Expression 3}$$

Cv_{PI}: maximum limit of activity concentration of radionuclide in the container (initial point of discharge).
CV_{max}: maximum limit of the activity concentration in the final point of the normal sewerage system.
V_c: liquid waste volume in the container.

V_{ec}: water volume released in the centre daily.

If it is necessary to discharge a mixture of radionuclides the expression 4 would be applied.

$$\sum_{i=1}^{n} \frac{Cv_i}{Cv_{PI\ i}} \leq 1 \qquad \text{Expression 4}$$

Cv_i: activity concentration in the container
Cv_{PIi}: maximum limit of activity concentration of radionuclide in the container (initial point of discharge).

3. RESULTS AND DISCUSSION

3.1. CHARACTERIZATION OF LIQUID WASTE

The table nº 1 shows the obtained results of characterization of liquid waste in the analyzed techniques. The activity percentage in the waste has been calculated applying 100 % of initial activity used in each technique[6].

These data indicate that the characterized waste have low activity concentration. Therefore, at the start, they are clear candidate for clearance, some of them may be directly and another should be stored for decay if they have long half-lives. The cleared waste could be release to the normal sewerage system (aqueous liquid) or incineration.

Table nº 1 Radiological characterization of liquid waste of different techniques

Nº	Group of techniques	Technique	Radionuclide	Radioactive concentration (Bq/ml)	Total %
1	Enzymatic Assays	Leucine decarboxilation	^{14}C	0,338	0,006
		Methionine adenosyl transferase	^{3}H	244,3	51,50
		Deoxiglucose Intake	^{3}H	590	13,71
		Telomeric amplification	^{32}P	181,9	12,29
2	Cell culture assays	Ethanolamine kinase	^{14}C	2.989,6	33,7
		Ceramides quantification	^{32}P	76.487,8	87,18
		Intracellular calcium release	^{45}Ca	102.378,2	75,33
3	Nucleic Acid Hybridisation	Northern blot	^{32}P	186,3	5,08
4	Nucleic Acid Characterization	Electrophoretic mobility shift assay	^{32}P	27,3	1,19
5	Radioinmunoassay	Radioimmunoassay for GMPc	^{125}I	85,6	82,07
		Radioimmunoassay for methionine	^{125}I	324,7	18,84
6	Binding assays	Binding somatostatin-receptor	^{125}I	49.107,4	64,14
		Binding VIP-receptor	^{125}I	720,1	96,33
7	Radioiodination	Radioiodination of somatostatin	^{125}I	173,3	40,57

3.2 REFERENCE CLEARANCE LEVELS PROPOSED TO ORGANIC LIQUID WASTE

The considerations shown in the point 2.2.1 have been used to propose the reference values in terms of radioactive concentration for the incineration. In this way, to obtain the effective dose of 10 μSv/year it is necessary a continual emission of:

^{3}H	12.262 Bq/s	$3{,}9 \cdot 10^{11}$ Bq/year
^{14}C	171 Bq/s	$5{,}4 \cdot 10^{9}$ Bq/year

When both radionuclides are incinerated, so that the effective dose to individual is lower than 10 μSv/year the expression 5 must be fulfilled.

$$\sum \frac{A(H-3)(Bq/a)}{3.9 \cdot 10^{11}(Bq/a)} + \frac{A(C-14)(Bq/a)}{5.4 \cdot 10^{9}(Bq/a)} \leq 1$$ Expression 5

The calculation of reference clearance values has been performed using the indicated data and the volume of liquid waste generated in Spain for the whole radionuclides. This amount was 495 l and it was provided by ENRESA for de year 2008. The proposed values are given in table nº 2.

Table nº 2: Reference clearance levels proposed to organic liquids

^{3}H	$3{,}9\ 10^{11}$ Bq /year of ^{3}H / 495 l = $7{,}8\ 10^{8}$ Bq/ l
^{14}C	$5{,}4\ 10^{9}$ Bq /year of ^{14}C / 495 l = $1{,}09\ 10^{7}$ Bq/l

The liquid waste with radioactive concentration lower than these values may be incinerated with the Regulatory Body approval.

If it is necessary the incineration of both radionuclides simultaneously, the expression 6 must be achieved.

$$\sum \frac{A(H-3)(Bq/l)}{7.88 \cdot 10^{8}(Bq/l)} + \frac{A(C-14)(Bq/l)}{1.09 \cdot 10^{7}(Bq/l)} \leq 1$$ Expression 6

3.3 REFERENCE CLEARANCE LEVELS PROPOSED TO AQUEOUS LIQUID WASTE

The ALI_{ing} and the maximum limit of activity concentration for radionuclides used frequently in the cited installations have been calculated, showing the results in table nº 3.

3.3.1. Annual Limits on Intake by ingestion (ALI_{ing})

To calculate the maximum activity that an individual may intake in order to not exceed the Annual Effective Dose Limit (1 mSv) the expression 1 should be applied (Materials and Methods). The values of ALI_{ing} are presented in table nº 3.

3.3.2. Maximum limit of activity concentration in the final point of the normal sewerage system CV_{max},

The maximum limit of activity concentration in the final point of the normal sewerage system, CV_{max}, has been calculated using the expression 2 (Materials and Methods). The results are shown in table nº 3.

Table nº 3. ALI $_{ing}$ and maximum limit of activity concentration

		ALI_{ing} (Bq)	CV_{max} (kBq/l)		ALI_{ing} (Bq)	CV_{max} (kBq/l)
^{3}H	H_2O tritiated	$5,56 \cdot 10^{7}$	92,593	^{55}Fe	$3,03 \cdot 10^{6}$	5,051
	OBT	$2,38 \cdot 10^{7}$	39,683	^{59}Fe	$5,56 \cdot 10^{5}$	0,926
^{14}C		$1,72 \cdot 10^{6}$	2,874	^{65}Zn	$2,56 \cdot 10^{5}$	0,427
^{22}Na		$3,13 \cdot 10^{5}$	0,521	^{75}Se	$3,85 \cdot 10^{5}$	0,641
^{32}P		$4,17 \cdot 10^{5}$	0,694	^{86}Rb	$3,57 \cdot 10^{5}$	0,595
^{33}P		$4,17 \cdot 10^{6}$	6,944	^{111}In	$3,45 \cdot 10^{6}$	5,747
^{35}S	inorganic	$7,69 \cdot 10^{6}$	12,821	^{125}I	$6,67 \cdot 10^{4}$	0,111
	organic	$1,30 \cdot 10^{6}$	2,165	^{131}I	$4,55 \cdot 10^{4}$	0,076
^{36}Cl		$1,08 \cdot 10^{6}$	1,792	^{133}Ba	$6,67 \cdot 10^{5}$	1,111
^{45}Ca		$1,41 \cdot 10^{6}$	2,347	^{153}Gd	$3,70 \cdot 10^{6}$	6,173
^{51}Cr		$2,63 \cdot 10^{7}$	43,860			
		$2,70 \cdot 10^{7}$	45,045			

3.3.3. Maximum limit of activity concentration of the waste to discharge in the initial point of the the normal sewerage system (Cv_{PI})

Considering the dilution of the discharged liquid waste in the water volume released in the centre, the concentration in the initial point of discharge could be greater than the concentration in the final point of the normal sewerage system.

It is possible to determine the Cv_{PI} values for the different radionuclides used in each installation, being necessary to set a maximum volume waste to dispose each time and water volume released in the centre daily. The waste which activity concentration is lower than the Cv_{PI} values could discharged to the the normal sewerage system with the Regulatory Body approval

To undergo this, it is necessary to perform some previous actions to verify the possible release of this waste.

- Measure the activity concentration of one radionuclide into the initial container Cv, applying the protocol given in point 2.1.
- Compare the obtained result with the maximum limit of activity concentration Cv_{PI} .

Different situations could appear:

- If the value of Cv, is lower than Cv_{PI} the waste could be discharged to the normal sewerage system with the regulatory body approval.
- If the value of Cv, is greater than Cv_{PI} the waste could not be discharged. Three possibilities exist in this situation:
  - When the waste contain radionuclides with half-lives < 100 days, the necessary time to reach a Cv lower than $Cv_{PI,}$ it could be stored for decay.
  - The waste could be diluted to obtain a Cv lower than Cv_{PI} (then it may be discharged).
  - When the waste contain radionuclides with half-lives > 100 days, and the dilution is not possible, it must be transferred to radioactive company waste.

3.4. PRACTICAL EXAMPLE ASSUMING THE CLEARANCE OF AQUEOUS LIQUID WASTE

One storage of the residual materials with radioactive content has two drums with aqueous liquid waste, one of them has ^{3}H and another with ^{14}C.

Data:
V_c: liquid waste volume in the drum (30 l)
Cv: activity concentration of the drum is calculated applying as indicated in point 2.1.
V_{ec} **:** water volume released in the centre daily (45.000 l)

Results obtained:
Cv of ^{3}H drum was 215 kBq/l
Cv of ^{14}C drum was 229,72 kBq/l

Procedure
Firstly, the values of ALIing and the maximum activity concentration in the final point of the sewerage system (**CV_{max})** for ^{3}H and ^{14}C were calculated applying the expression 1 and 2 (Materials and Methods). The results are shown in table nº 4.
Cv_{PI} (the maximum limit of activity concentration of radionuclide in the container, initial point of discharge) was calculated applying the expression 3 (Materials and Methods). The results of which are given in table nº 4.

Table nº 4. Maximum limit of activity concentration to discharge in the initial point of the normal sewerage system (*Cv_{PI}*) for some radionuclides

		ALI (Bq)	*CVmax* (kBq/l)	*Cv_{PI}* (kBq/l)
^{3}H	$^{3}H_2O$	$5,56 \cdot 10^{7}$	92,593	138.981,48
	OBT	$2,38 \cdot 10^{7}$	39,683	59.563,49
^{14}C		$1,72 \cdot 10^{6}$	2,874	4.313,22

These results allow to confirm that the radioactive concentration of the waste to discharge, is lower than the values of Cv_{PI} shown in table nº 4.

Cv of ^{3}H drum is 215 kBq/l is lower than ^{3}H Cv_{PI}: 59.563,49 kBq/l.
Cv of ^{14}C drum is 229,72 kBq/l, is lower than ^{14}C Cv_{PI}: 4.313,22 kBq/l.

Considering the joint discharge of two radionuclides, it is necessary to verify that the expression 4 is accomplished. The obtained result is:

$$\frac{215}{59.563,49} + \frac{229.72}{4.313,22} = 0,0036 + 0,0532 = 0,0568$$

This result is lower than 1, therefore it is possible to discharge the aqueous liquid waste contained in the indicated drums to the normal sewerage system, taking into account the regulatory body considerations given in point 2.2.2.

4. CONCLUSIONS

This work considered two options in relation with the release way to liquid waste, the transfer to radioactive waste company or their clearance and ulterior release attending to their chemical composition, organic or aqueous.

The non-aqueous liquid waste represent a small volume in relation with the total liquid waste, the final release way proposed is the incineration prior handing over to hazardous waste company. Therefore, it is essential that their radioactive concentration is lower than the reference levels given by the regulatory authorities.

To determine the derived values of activity concentration, it was used the theorical model made by Unidad de Protección Radiológica del Público y del Medio Ambiente del Ciemat[7]. The results obtained show that the organic liquid whit radioactive concentration lower than $7{,}8\cdot10^{9}$Bq/l de ^{3}H ó $1{,}09\cdot10^{7}$Bq/l de ^{14}C would be incinerated assuring a public dose constrain of 10 μSv/year.

The ^{3}H and ^{14}C radioactive concentrations of organic liquid waste generated in the characterized techniques, shown in table nº 1, are lower than the values indicated above. Therefore, It is feasible to make the clearance of this waste and transfer it to an hazardous waste company to its subsequent incineration. However, it is necessary to take into account that the proposed values have been obtained considering an annual production of 495 l of organic liquid waste (information provided by ENRESA for the year 2008). This quantity could be changed as from now, implying a modification of the proposed radioactive concentration values.

In relation with the aqueous liquid waste, this study suggests the discharge to the normal sewerage system as a final disposal way. Thus, the radioactive concentration values of this waste should be lower than the calculated concentration values in the initial point of discharge (CVpi). In this work the CVpi values have been determined for different radionuclides used in a standard radioactive installation of research and taking into account a water volume released in the centre of 45.000 l /day. The measured radioactive concentration of aqueous liquid waste of ^{3}H and ^{14}C, presented in table nº 1, are lower than the calculated CVpi values for these radionuclides, therefore these liquids could be discharged directly to the normal sewerage system.

As regards the radioactive concentration of aqueous liquid waste of other radionuclides, as ^{32}P or ^{125}I, the obtained results are greater than the calculated CVpi. However, taking into account their short half-lives, this waste could be stored for decay the time necessary to reach the corresponding CVpi values and then be released.

In conclusion, it is possible confirm the clearance of the generated waste in the analyzed techniques considering the obtained results on their characterization. This paper suggests that each radioactive installation need to perform their own calculations in order to obtain the corresponding values of activity concentration of the waste to discharge in the initial point of the normal sewerage system (Cv_{PI}). The use of presented procedures is suggested in this study, this fact could allow a homogeneous management of the indicated waste in the research field.

Nowadays the organic liquid waste management is unsolved. Therefore this work emphasizes the necessity to establish the reference clearance values for organic liquid waste, suggesting the use of the obtained results to determine the indicated values. These reference clearance values should be approved by the regulatory body and published in a technical document. In this way, an official framework could be used, making easier the waste management in all installations, allowing an important decrease of the transferred waste to the radioactive waste company. On the other hand, it would be resolved the lack of the legislation in relation with the transfer of these waste to the hazardous waste companies and ulterior treatment by incineration.

ACKNOWLEDGEMENTS
The authors wish to thank Mr J.C. Mora and Ms B. Robles (Unidad de Protección Radiológica del Público y del Medio Ambiente .Ciemat) for their collaboration in these studies.

REFERENCES

[1] IAEA Safety Standards. Clasification of radioactive waste. 2009. IAEA Safety Standards Series .General Safety Guide Nº GSG-1. (International Atomic Energy Agency. Vienna)
[2] IAEA Safety Standards. Radiation Protection and Safety of Radiation Sources. International Basic Safety Standards. Interim Edition. 2011. No. GSR Part 3. (International Atomic Energy Agency. Vienna)
[3] IAEA Safety Standards. Application of the Concepts of Exclusion, Exemption and Clearance. 2004. IAEA Safety Standards Series No. RS-G-1.7. (International Atomic Energy Agency. Vienna)
[4] IAEA TECDOC. Clearance of materials resulting from the use of radionuclides in medicine, in industry and research. 1998. IAEA-TECDOC-1000. (International Atomic Energy Agency. Vienna)
[5] Castell, A., Macías, M.T., Olivares, P., Ortíz, M.T., Plaza, R., Quesada, J.D., Sánchez, A y Téllez, M.1996. Guía de Gestión de Material Radiactivo en Instituciones Médicas y Laboratorios de Investigación Biomédica. Sociedad Española de Protección Radiológica, publicación nº 2. (Senda. Madrid)
[6] Macías, M.T., Pulido, J., Ruiz, A., Sánchez, M., Sánchez, A. y Usera, F. 2002.Guía técnica de gestión de materiales residuales con contenido radiactivo en centros de investigación y docencia. Sociedad Española de Protección Radiológica, publicación nº 7. (Senda. Madrid).
[7] Macías, M.T.,Pulido,J.,Pérez, J., Sastre, G., Sánchez, A., Usera, F.2010.Optimization of Management of Radioactive Waste Generated in Research and Education Centres. Proceedings of the 3th European IRPA Congress- Helsinky (Finland) 14-18 June 2010. (Stuk. Finland). 962-971
[8] ORDEN ECO/1449/2003, de 21 de mayo, sobre gestión de materiales residuales sólidos con contenido radiactivo generados en las instalaciones radiactivas de 2ª y 3ª categoría en las que se manipulen o almacenen isótopos radiactivos no encapsulados. (BOE num. 134 de 5 de junio de 2003)
[9] IAEA TECDOC. Management of radioactive waste from the use of radionuclides in medicine. 2000. IAEA-TECDOC-1183. (International Atomic Energy Agency. Vienna)
[10] ICRP 23. International Commission on Radiological Protection. Report of the task group on reference man.1975. ICPR Publication 23 (Pergamon Press.Oxford)
[11] Chapel, M., Ferrer N., Ramos, L.,Sánchez,M.2002.Grupo de efluentes del foro de protección radiológica en el medio hospitalario. (Sociedad Española de Física Médica. Madrid)
[12] Texto comentado del Reglamento de Protección Sanitaria contra Radiaciones Ionizantes. (Real Decreto 783/2001, de 6 de Julio).2001.Sociedad Española de Física Médica. Sociedad Española de Protección Radiológica y Sociedad Nuclear Española. (Senda. Madrid)