SURVEILLANCE OF RADIOACTIVE DISCHARGES FROM THE CENTRE OF ISOTOPES OF CUBA

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ABSTRACT

The handling of unsealed radioactive sources in the Centre of Isotope of Cuba leads to radioactive discharges which are under radiological surveillance as a guaranty for comply with the authorized limits for the Regulatory Authority. This paper summarizes the findings from control and monitoring of these releases applying the international standards and recommendations, covering the period of 15 years. A radiometer Berthold LB 2040 with a Geiger Müller detector is used for measurement activity concentrations of ^{131}I in airborne releases. A spectrometric system with HPGe detector is used for measurement concentrations of gamma radionuclides in waste waters. The energy and efficiency calibrations of this system are carried out with a set of solid point radioactive sources and cylindrical ^{133}Ba and ^{152}Eu liquid volume sources (with 425mL), respectively. Solutions used for elaboration of cylindrical sources are certificated by the National Metrological Institute of Hungary. Pure beta emitters are measured using a method of liquid scintillation. ^{131}I and ^{32}P are the most representative radionuclides in waters. The measurement method employed allows in a relative short time and with smaller than 10% of uncertainty to detect any deviation of the safety procedure and good practices. In the 61% of the measurements of liquid effluents we have to apply the reducing sources principle, due to the very conservative character of the authorized clearance levels. A prospective dose assessment for airborne discharges is used to characterize the annual radiological impact to public. The results obtained show a good agreement with the dose constrains.

Keywords: radioactive liquid discharges, airborne discharges, radiological surveillance.

Introduction

The liquid effluents arising from radiochemical laboratories, change-rooms and showers of the Centre of Isotopes (CENTIS) in Cuba are collected in a special canalization system which has 93 generation points [1]. This system is provided with storage facilities so that short lived radionuclides can decay before release in compliance with the clearance levels established by Regulatory Authority [2]. The airborne discharges are sampled during handling unsealed radioactive sources. In this paper findings of their control and monitoring in accomplishment to the applicable regulations [2] are summarized.

Materials and methods

A quality assurance program is established for control and monitoring liquid releases [2]. Requirements relating to representative samples are implemented [2]. A quality assurance program is established for control and monitoring these effluents [3]. Requirements for assuring representative samples are implemented and fulfilled [4÷7]. Source monitoring program is designed to measure the discharge rates of radionuclides [8]. The measurement of the activity concentration of gamma emitters with energy of (59 to 1500) keV is carried out in a spectrometric system of high resolution with HPGe detector, according to established procedure [9] and recommendations [10]. Liquid reference materials used for quantitative analysis were obtained by addition of known aliquot of both ^{152}Eu and ^{133}Ba solutions, provided by the National Metrological Institute of Hungary (MKEH) [11], to aqueous solutions in plastic containers, of the same type as used for performing the routine analysis. Previously, the activity concentration of ^{152}Eu and ^{133}Ba was determined by the method described elsewhere [12]. These reference materials are utilized for carrying out the efficiency calibration of the spectrometric system according the procedure [13].

For beta emitters monitoring is used a method of scintillation for determine ^{32}P, ^{90}Sr and ^{90}Y since 2009. Detection limits (LLD) in terms of concentration minimum detectable, with a significance level of 0.05, are calculated in correspondence with [10].The uncertainties values of measurements are calculated according to the recommendations [14÷15] and are less than 20%.

For effluents management it is rejected the dilution effect and the recommended liquid discharges pH values are used [7].

The traceability of samples and effluents are settled down using a unique identification number for tank and the samples in the records [1] and [9]. On the other hand, for carrying out the analysis two samples with identical geometry are taken from the tank, after the liquid has been homogenized using a proper system [4], and measured. For accepting the results of gamma activity concentration determined in both duplicated samples, requirements of established internal quality control have to be fulfilled [16÷17].

Data from effluents measurements are compared with the values of unconditional clearance levels (UCL) established by Regulatory Authority [2]. These values are the same for clearance and discharges limits.

The dose criterion used to derive clearance levels limits is based on the Schedule I of the Basic Safety Standard [18] of the International Atomic Energy Agency (IAEA), the use of which has been agreed upon by the Cuban Regulatory Authority.

This means the primary radiological basis for establishing activity concentration for radionuclides of man-made origin is that the effective doses to individuals should be of the order of 0.01 mSv or less in a year. Either the collective effective dose committed by year of performance of the discharge is no more than about 1 man-Sv or an assessment for the optimization of protection shows that clearance is the optimum option. This approach is consistent with that used in establishing the exemption levels for small amount of solid material [18].

A quality assurance program is established for control and monitoring airborne releases [19]. Requirements relating to representative samples are implemented [19]. An isokinetic sampling probe is used in the duct where is a reasonable degree of mixing in the exhaust air stream [20]. Source monitoring program is designed to measure the discharge rates of radionuclides [21]. The most volatile and frequently radionuclide used is ^{131}I and for this reason F&J radioiodine collection cartridges (model TE2C 30x50) and 47mm glass fiber filters (FP-47) are used with an analogue low volume air sampler (model VS 23-0523CV) from HI-Q Environmental Products Company, USA [22].

A routine exhaust air sampling and monitoring with a weekly filter exchange frequency are currently executed; using a radiometer Berthold LB 2040 with a Geiger Müller detector (model GZ-7).

The efficiency calibrations of this system are carried out with a source of ^{90}Sr type capsule with Aluminium as backing, from Isotopes Products Laboratories with NIST traceability. The nominal cubic flow rate from stack is 67 554 $m^3 h^{-1}$.

The established unconditional clearance levels (UCL) for airborne discharges of ^{131}I are: 5.94E+01 Bq m^{-3} and 1E+08Bq y^{-1} [3]. These values are the same for clearance and discharges limits as we before explained.

The averaged activity concentration and release rate by discharge are calculated for each year. Their trends are presented and analyzed over the 11 y period from 2001 to 2011. Operations with ^{131}I in a hot cell and manufactured practices for obtaining labelled compounds in a glove box are studied.

Results

Results from monitoring of liquid effluents mainly show the presence of ^{131}I, ^{32}P, ^{125}I and ^{99}Mo. For this order, the mean measured radionuclide concentrations during the total control period of 1998-2011, in Bq L^{-1} are: 8.10E+01, 6.43E+03, 2.68e+01 and 9.18E+01. The two first radionuclides have indeed the greatest influence on the management of these effluents. The use of UCL indicates the necessity of a specific prospective dose assessment where the concept of the representative person should be used [23] and implies an increasing of management costs.

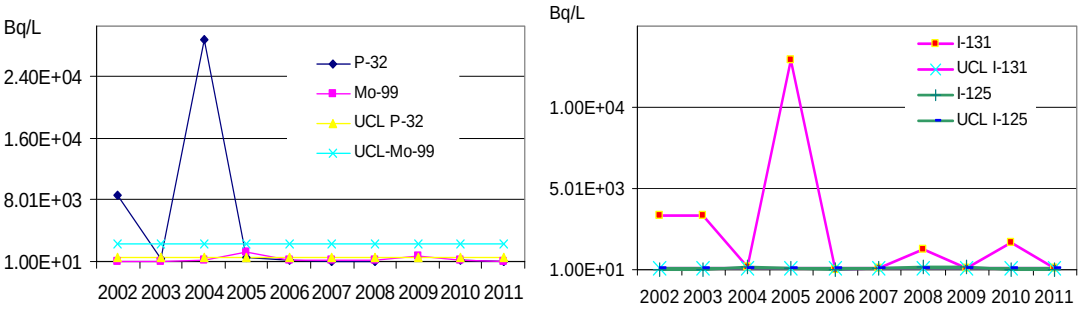

Figure 1. Maximum activity concentration radionuclides measured in liquid effluents vs. unconditional levels (UCL)

The mean monitoring frequency is 13 days and the averaged released volume is 2 m^3.The uncertainty obtained is about 10%. LLD for ^{131}I is 9 Bq L^{-1} and for ^{32}P is 2E+02 Bq L^{-1}. These values are determined with a 95% of confidence. Chemical control of the waters reflects an averaged value of pH equal to 7; for this reason its readjustment was not required [7].

In the table 1 the annual handling activity and total activity for ^{131}I by year in the effluents are presented. The percentage of effluents retained for the reduction of the source term is also shown.

Preliminarily, the 50% of annual effluents´ volume was considered as radwastes [24]. As can be seen in table 1, there is an important reduction of generation of liquid radwastes since 2007.

Training of the staff allows obtaining this purpose. Nevertheless, additional measures are implemented for decreasing the effluent generation. For instance, the close of water supply when handling radioactive materials operations are finished.

Mean handling activity and total activity in liquid effluents by year are reflected in table 1 (^{131}I) and table 2 (^{32}P). In the case of the last since 2007 to 2009 this radionuclide was not measured because we have a problem with its determination in presence of ^{90}Sr. The mean value of release fraction for the first is about 4E-05 and for the second is around 9E-05. Despite the bigger volatility of ^{131}I, this is an evidence of the very contaminant characteristic of Orthophosphoric Acid with ^{32}P, which is the radioisotopic matter for Sodium Phosphate production. A high frequency of the staff's contamination with ^{32}P has been registered and influenced this result.

Table 1. Mean handling activity and total activity of ^{131}I in the effluents and the percentage of these treated as radwastes.

Year	Mean handling activity of ^{131}I (Bq y^{-1})	Total activity of ^{131}I (Bq y^{-1})	Release fraction	Volume of liquid effluents (m^3 y^{-1})	Effluents treated as radwastes (%)
1998	4.90E+12	5.68E+06	1.16E-06	38.0	65.0
1999	4.87E+12	9.79E+06	2.01E-06	73.3	81.3
2000	1.03E+11	2.45E+06	2.38E-05	30.0	46.7
2001	9.97E+10	3.04E+06	3.05E-05	38.0	21.1
2002	7.80E+10	9.75E+06	1.25E-04	26.0	55.6
2003	8.21E+10	1.23E+06	1.50E-05	22.0	100.0
2004	1.13E+11	1.34E+06	1.19E-05	27.8	95.5
2005	9.06E+10	2.65E+07	2.92E-04	32.0	87.5
2006	7.19E+10	1.00E+05	1.39E-06	22.0	88.9
2007	1.04E+11	4.60E+05	4.42E-06	36.0	28.6
2008	9.09E+10	3.38E+06	3.72E-05	36.0	31.3
2009	1.34E+11	8.90E+05	6.64E-06	14.0	57.14
2010	1.48E+11	3.66E+06	2.47E-05	18.0	40.00
2011	2.18E+11	1.15E+06	5.28E-06	42.0	9.50

In table 3 we can see the maximum activity concentration of liquid discharges and their respective value of UCL for all of existing radionuclides. There are only 2 deviations with respect to ^{32}P (2002 and 2004). Really, this result is not reliable because we considered only the presence of this radioisotope and the measurement did not allows determine its concentration with respect of ^{90}Sr. In the other hand this water is mixed with the rest of 25 m^3 of CENTIS' waters. As we can appreciate, for the rest of radioactive inventory in discharges there is no problem.

In the light of operating experience and as it was considered in [24], radioiodines (^{131}I and ^{125}I) have the biggest source term in airborne releases, allowing for the kind of operations with them in CENTIS. On the other hand, the first is used with biggest activity and frequency.

Table 4 lists the mean annual handling activity of ^{131}I and its maximum activity concentration by discharge and annual release rate. As can be observed in this, the maximum radioactive concentration and annual release rate of ^{131}I registered are about 30 Bq m^{-3} and 6.82E+07 Bq y^{-1}, which are 0.5 and 0.68 times lower than their respective values of UCL.

Table 2. Mean handling activity and total activity in the effluents for P-32

Year	Mean handling activity of ^{32}P (Bq y^{-1})	Total activity of ^{32}P in effluents (Bq y^{-1})	Release fraction
2002	2.35E+11	2.90E+07	1.23E-04
2003	2.35E+11	1.01E+07	4.30E-05
2004	1.93E+11	4.29E+07	2.23E-04
2005	9.75E+10	6.23E+06	6.39E-05
2006	5.45E+10	1.90E+06	3.48E-05
2010	1.51E+10	1.20E+06	7.95E-05
2011	1.30E+10	1.03E+06	7.95E-05

Table 3. Maximum activity concentration of radioisotopes in liquid discharges

	Maximum activity concentration ^{131}I (Bq L^{-1})	Maximum activity concentration ^{125}I (Bq L^{-1})	Maximum activity concentration ^{32}P (Bq L^{-1})	Maximum activity concentration ^{99}Mo (Bq L^{-1})	Maximum activity concentration $^{90}Sr/^{90}Y$ (Bq L^{-1})
UCL (Bq L^{-1})	6.23E+01	9.13E+01	5.71E+02	2.28E+03	4.89E+01
Year					
1998	4.66E+01	No measured	No handling	No measured	No handling
1999	4.26E+01		No measured		
2000	4.17E+01				
2001	4.55E+01				
2002	4.12E+01		8.64E+03		
2003	2.24E+00		3.80E+02		
2004	1.70E+01	6.96E+01	2.88E+04	1.19E-01	No measured
2005	1.17E+01	8.22E+01	5.71E+02	4.35E-02	
2006	7.83E+00	1.19E+01	2.72E+02	3.38E-03	
2007	1.58E+01	5.02E+01	No measured	1.09E-02	
2008	2.63E+01	8.13E+01		2.82E-03	
2009	5.05E+01	6.39E+01		1.19E-01	3.00E+01
2010	3.65E+00	1.16E+01	1.47E+02	1.16E+01	4.00E+01
2011	5.04E+01	1.58E+01	3.70E+01	1.58E+01	4.60E+01

The averaged values of activity concentration and release rate by discharge taking into account all operations with ^{131}I are shown in table 5. Besides the labelling of compounds has a biggest value of source term and radionuclide concentration than the practice in the hot cell.

Table 4. Mean annual handling activity, maximum activity concentration and annual release rate of ^{131}I

Year	Annual handling activity ^{131}I (TBq y^{-1})	Maximum activity concentration of ^{131}I (Bq m^{-3})	Annual release rate of ^{131}I (Bq y^{-1})
UCL	--	**5.94E+01**	**1E+08**
2001	4.88	2.87E+00	1.12E+07
2002	4.60	2.45E+01	6.79E+07
2003	3.94	4.85E+00	1.99E+07
2004	4.71	1.03E+01	6.32E+07
2005	4.08	4.38E+00	4.10E+07
2006	3.28	1.61E+01	3.04E+07
2007	4.91	1.91E+01	6.82E+07
2008	4.33	2.99E+01	2.61E+07
2009	5.76	1.96E+01	3.46E+07
2010	7.09	2.02E+01	3.07E+07
2011	10.46	1.37E+01	3.62E+07

Table 5. Mean activity concentration and mean activity of I-131 by discharge

Year	Mean activity concentration of ^{131}I (Bq m^{-3})	Mean activity of ^{131}I (Bq)
2001	5.39E+00	2.31E+05
2002	1.06E+01	5.20E+06
2003	1.27E+00	5.71E+05
2004	1.67E+01	1.35E+06
2005	7.14E+00	3.46E+06
2006	1.45E+00	6.34E+05
2007	2.66E+00	1.24E+06
2008	6.90E+00	4.66E+05

This is shown in table 6, where data of 2008÷2011 are reflected. This process took place in these years only as a mean of nine occasions. The compound is concentrated by evaporation and a chemical solution is used for retention of free iodine and the discharge pass through activated charcoal filter, impregnated with triethylenediamine (TEDA).

The maximum value registered for the release fraction (RF) is 7.85E-05 and the mean RF for the 10 years analyzed period is 6.06E-06. The percentage contribution to RF is presented in the

table 7. As can be seen in the majority of the data, RF is about two orders lower than the estimated value (1E-03) in [25]. This difference is in the kind of procedure in the hot cell since radiopharmaceutical compound is not obtained from distillation, like it firstly was projected.

In the figure 2 can be seen as the E distribution has values 10 times lower than individual dose constrain of 10 µSv y^{-1}. Besides, this implies for a committed collective effective dose (S) equal to 1 man-Sv y^{-1} that critical group has 1E+06 infants. This issue is a very faraway boundary condition for CENTIS and means the deterministic approach considered requires a substitution by a specific prospective dose assessment where the concept of the representative person should be used [23].

Table 6. Mean activity concentration and mean activity of ^{131}I by discharge from hot cell and glove box

Workplace	Mean activity concentration by discharge of ^{131}I (Bq m^{-3})	Mean activity by discharge of ^{131}I (Bq)
Hot cell	1.92E+00	6.64E+05
Glove box	2.81E+00	5.55E+05

Table 7. Percentage of the release fraction in the airborne discharges

Release fraction	1.00E-05	1.00E-06	1.00E-07
%	77.5	8.4	14.1

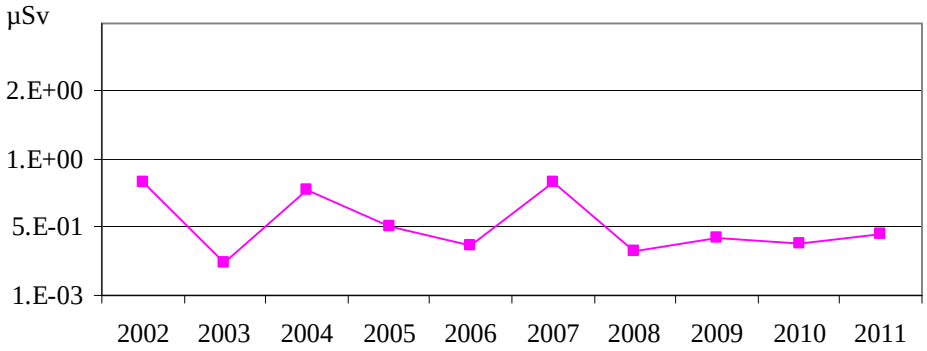

Figure 2. Plots of annual effective dose to public due to airborne releases from CENTIS

Conclusions

The main operating findings from liquid and airborne effluents management in CENTIS are: the radiological surveillance established and maintained complies with applicable international regulations; it is possible to detect deviations of the safety procedures and good practices, in the same day of measurement of effluents and the training of personnel can have a significant effect on reducing these deviations but also it is necessary implementing some simple measures that lead decreasing of generation of these waters. The biggest release fraction to liquid effluents registered belongs to ^{32}P and this implies the necessity of reducing the frequency of the contaminations. The presence of a mixing of beta emitters requires has been determined with a

quick method with good results. In the spite of low frequency of operations, the labelling of compound has a bigger release fraction than the practice in the hot cell due to the concentration by evaporation. The release fraction is in the 77% of the discharges about 1E-05 which is two orders lower than the projected value.

The Cuban Regulatory Authority has set the same limits for clearance and discharges. This is indeed very conservative. Clearance and discharges have not only the same limits but also the same conditions for proof of compliance. The results presented in this paper are therefore useful for clearance processes in other cases. The deriving clearance levels from specific prospective dose assessment for CENTIS is the key step for application of the most appropriate system of protection, utilizing both compliance with quantitative constrains and optimization of protection.

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