

**Determination of Radioisotope Content by Measurement of Waste Package  
Dose Rates– 13394**

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**ABSTRACT**

The objective of this communication is to report the observed correlation between the calculated air kerma rates produced by radioactive waste drums containing untreated ion-exchange resin and activated charcoal slurries with the measured radiation field of each package. Air kerma rates at different distances from the drum surface were calculated with the activity concentrations previously determined by gamma spectrometry of waste samples and the estimated mass, volume and geometry of solid and liquid phases of each waste package. The water content of each waste drum varies widely between different packages. Results will allow determining the total activity of wastes and are intended to complete the previous steps taken to characterize the radioisotope content of wastes packages.

**INTRODUCTION**

The Radioactive Waste Department (GRR) is responsible for the treatment and interim storage of the radioactive wastes generated by the Nuclear and Energy Research Institute (IPEN-CNEN/SP) in São Paulo, Brazil. The main facility of IPEN is a nuclear research reactor – a 5 MW pool-type reactor that operates since January 1958 for radioisotopes production, research in nuclear physics and correlate areas, and irradiation experiments. The GRR has also a training program on radioactive waste management and conducts research and development work, under a post-graduate program with the University of São Paulo.

One of the duties of GRR is to characterize the wastes to comply with the safety and quality requirements established by Brazilian regulations. The characterization of the radioactive wastes includes among other things the development and application of methods to determine the radioisotope content of waste packages.

The determination of the radioisotope content of the wastes is required in two instances: characterization of raw waste and characterization of waste forms. The radioactive inventory of the raw waste is necessary to guide the treatment processes, to establish radiation protection requirements for waste handling, and to prescribe the operational details of the treatment like the number of packages required, storage space, etc. For waste forms, the radioactive inventory report of each waste disposal package is required for transportation and for acceptance of the waste in a repository for final disposal.

The GRR currently develops the protocols for sampling and analysis of spent ion-exchange resin and activated charcoal filter beds from the water polishing system of the reactor. Water is used as moderator, coolant and biological shielding. The fission and activation products, released from the reactor core, as well other impurities present in the coolant are retained in the resin and charcoal filters to keep the water quality within acceptable operational limits [1]. When the sorption capacity is no longer able to keep operation conditions, the filter beds are replaced and discharged as radioactive waste. Seven drums of 200L capacity with ion-exchange resin and 14 with activated charcoal slurries are presently stored in the GRR waiting for treatment.

Previous work has focused on gamma spectrometry of samples drawn from each waste drum [2]. The main gamma emitters present are  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  with minor quantities of other fission and activation products. Their concentrations were expressed as activity per gram of dry sorbent because the sampling protocol could not maintain the proportion of dry sorbent to the water present in the drums and because the water content in each waste drum as received from the waste generator varies widely. The variation in water content was a consequence of pumping the slurries from the filter tanks to the drums, and collecting the water used to flush the tanks and pipes to remove any remains of the sorbents. However, a few drums have a low water content and the sorbent is almost dry. Consequently, the determination of the activity content of each waste package requires the determination of its water and dry sorbent content.

In this communication we report the method used to estimate the water content of the waste drums and the results of the calculated total package activity are described. Furthermore, the measured and the calculated air kerma rates delivered by waste drums are compared as a means of checking the accuracy of the activity determinations.

## **MATERIALS AND METHODS**

To calculate the total activity present in each drum, the previously determined activity concentrations, dry basis, are multiplied by the total mass of the sorbent, which is equal to the total net mass of the package minus the mass of water. This approximation was made because the concentration of activity in the free water is orders of magnitude lower than the concentration in the sorbents.

The water content of a drum is the sum of the mass of water present ‘internally’, the interstitial water and the supernatant water. Internal water is that remaining in the sample when the sorbent is saturated and no free water is visible, and interstitial water is that visible among the sorbent grains. However, the water content of each waste drum cannot be determined directly, it is only possible to measure the height of the solid phase and the height of the liquid phase.

The drums were weighted and the levels of sorbent and water measured by visual inspection and observing the marks left by the water in a stick inserted in the drums. Figure 1 shows schematically the height of water and height of sorbent observed in the 21 drums.

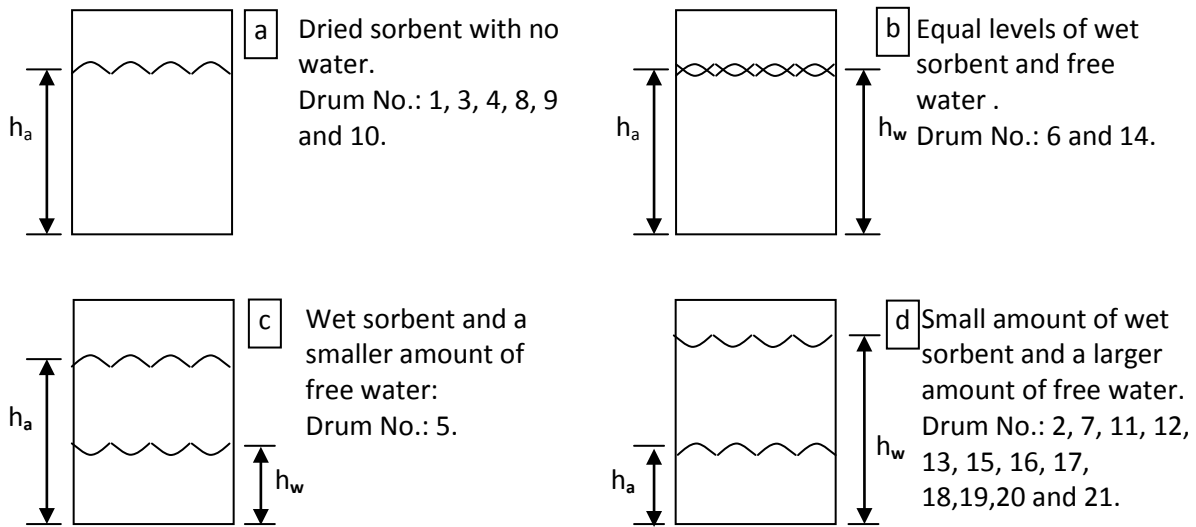


Figure 1 – Water content of ion-exchange resin or activated charcoal waste drums.

In the case of drums with the configuration **a** of Fig. 1, the sorbent mass is assumed to be equal to the net mass of the drum and the total activity is the product of activity concentration by the net mass of the drum.

To calculate the total activity in the case of drums with the configurations **b** or **c** of Fig. 1, the sorbent mass is obtained by assuming a linear relationship between the ratios of phase heights and phase masses, with masses summing up to the net mass of the drum.

Finally, in the case of a drum with the configuration **d** of Fig. 1, the sorbent mass is obtained similarly as in the previous case, but subtracting from the net mass the supernatant water, which can be obtained by simple geometry considerations.

The relationship between the phase heights and masses was estimated using non-radioactive samples of the sorbents assuming that the relationship between the heights and masses observed in fresh samples is reproduced in the actual waste.

The relationship between the mass ratios and height ratios are derived as follows:

$$\begin{cases} \mathbf{m}_t = \mathbf{m}_a + \mathbf{m}_w \\ \frac{\mathbf{m}_w}{\mathbf{m}_a} = \mathbf{k}_1 \cdot \frac{\mathbf{h}_w}{\mathbf{h}_a} + \mathbf{k}_2 \end{cases} \Rightarrow \mathbf{m}_a = \frac{\mathbf{m}_t}{\mathbf{k}_1 \cdot \frac{\mathbf{h}_w}{\mathbf{h}_a} + \mathbf{k}_2 + 1}$$

where

$\mathbf{m}_t$  is the net mass of a drum,

$\mathbf{m}_a$  and  $\mathbf{m}_w$  are the masses of sorbent and of water,

$\mathbf{h}_a$  and  $\mathbf{h}_w$  are the heights of sorbent and of water, measured in the waste drums, and

$\mathbf{k}_1$  and  $\mathbf{k}_2$  are the correlation constants obtained empirically in tested samples.

These equations also hold for the configuration **d** above, except that the net mass  $m_t$  of the drum is replaced by the value calculated by:

$$m_t \leftarrow [m_t - r^2 \cdot \pi \cdot (h_w - h_a) \cdot \rho_w]$$

where

$r$  is the internal radius of the drum, and

$\rho_w$  is the density of the supernatant water, assumed to be  $1 \text{ g.cm}^{-3}$ .

The values of  $k_1$  and  $k_2$  were obtained as the average of a series of experimental determinations of the mass ratio and height ratio of the sorbents and water.

Approximately 0.5 kg of fresh samples of ion-exchange resin or activated charcoal were left to absorb water until saturate, were filtered to remove free water, weighted to determine the mass of absorbed water, and were then packed in flat bottom acrylic column, as shown in Fig. 2. Water was added until a clear water level was visible, filling the interstitial space between the sorbent grains. The mass and the level height of added water were recorded. Two more steps of water filling of the packed column had the mass and height recorded until the free water level exceeded the sorbent column height.

Water was drained and the sorbents were dried in oven at  $90^\circ \text{C}$  for 24 h, weighted, left to absorb water, repacked in the acrylic columns and the previously described steps were repeated.

Results were plotted as water-sorbent mass ratios against height ratios, and the best-fit straight line yielded the values of  $k_1$  and  $k_2$ , which were used to estimate the mass ratios of water and sorbent based on the net weights and heights of water and sorbent observed in the waste drums.

Sorbent mass, dry basis, in each drum could then be calculated and the total gamma emission estimated by multiplying the sorbent mass by the previously determined concentrations of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ .

The radiation field at various relative distances from drum surface was calculated by using initially the method described by Rockwell [3] and later with Microshield®, a commercially available computational program for gamma ray shielding and dose assessment.

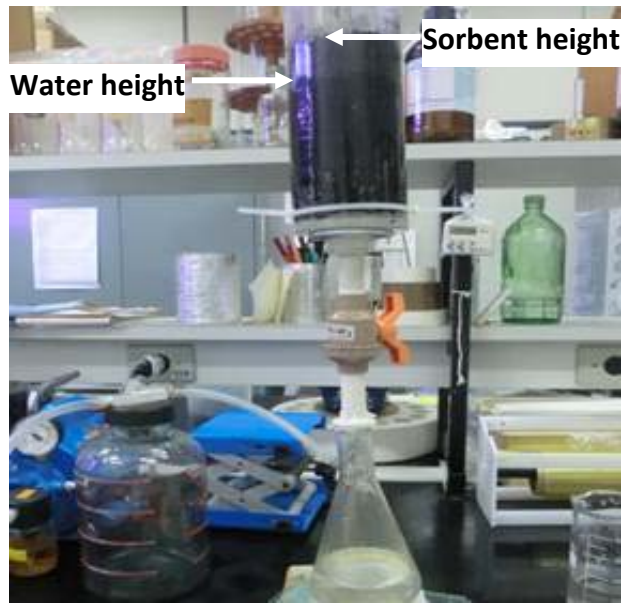


Figure 2 – Experimental setup for determination of mass-height ratios

The air kerma rate at a point P, as shown in Fig. 3, in the median plane of the sorbent volume of height **h** inside the waste drum, can be calculated by [3]:

$$\dot{\mathbf{D}} = \Phi \cdot \mathbf{G}$$

where

$\dot{\mathbf{D}}$  is the air kerma rate at point P, in Gy.h<sup>-1</sup>,

$\Phi$  is the photon flux at point P, in cm<sup>-2</sup>.s<sup>-1</sup>, and

$\mathbf{G}$  is the air kerma rate factor per unit of photon flux, in Gy. h<sup>-1</sup>.cm<sup>2</sup>.s

The photon flux is given by:

$$\Phi = \frac{\mathbf{B} \cdot \mathbf{S}_v \cdot \mathbf{R}_0^2}{2(\mathbf{a} + \mathbf{Z})} \cdot \mathbf{F}(\theta, \mathbf{b})$$

where

$\mathbf{B}$  is the build up factor

$\mathbf{S}_v$  is activity concentration, in Bq.cm<sup>-3</sup>,

$\mathbf{R}_0$  is the drum radius, in cm

$\mathbf{a}$  is distance between the drum surface and the point P, in cm

$\mathbf{Z}$  is called the effective self-attenuation distance, in cm

$F(\theta, \mathbf{b})$  is a function of the angle  $\theta$  and  $\mathbf{b}$ , with  $\mathbf{b}$  given by

$$\mathbf{b} = \mu_s \cdot Z$$

$\mu$  is the linear attenuation coefficient of the source material, in  $\text{cm}^{-1}$ ,

For  $\theta_1 = \theta_2$ , P at the median plane,

$$\theta = \arctg\left(\frac{h}{2a}\right)$$

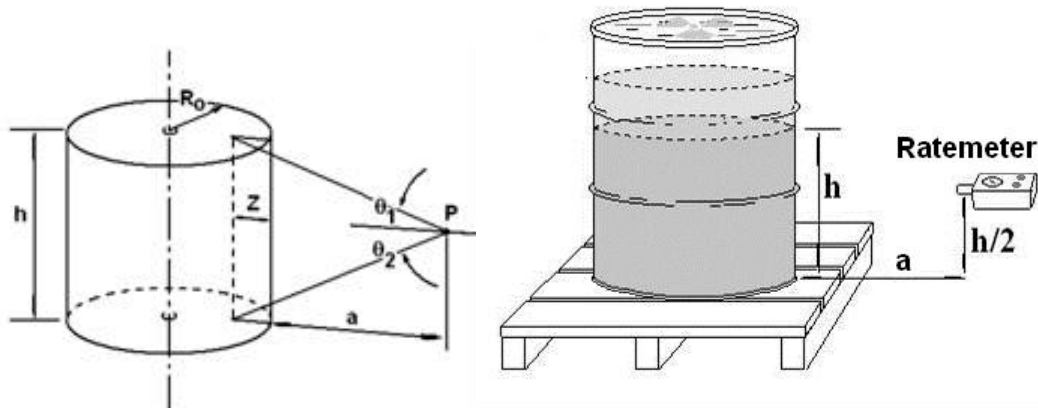


Figure 3 – Source–detector geometry for the air kerma rate calculations and measurements.

## RESULTS

Table 1 shows the net mass of the waste drums and the heights of the liquid and solid phases. Water level reported as zero in the table refers to drums of resin or charcoal with a low moisture content.

Figures 4 and 5 show the graphs with which the values of the coefficients  $k_1$  and  $k_2$  were determined to fit the experimental data on the water content of ion-exchange resin and activated charcoal. The concentrations of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  activity shown in table 2 are the average of the determinations performed in dry or dried waste samples drawn from each drum; the mass of sorbent and water, and the total activity present in each drum are also reported.

The activity concentrations were determined in dried samples of the sorbents, so these data are expressed in becquerels per gram, dry basis. In all cases, a few grams of the wastes were taken as a precaution to keep low the radiation exposures during handling, preparation and analysis of samples.

Finally, in table 3 the calculated and measured air kerma rates are reported at 1 meter from the lateral surface of the drum. As initially in the development of the work, the air kerma rates were calculated manually, following the method described by Rockwell [1956], we choose to present these results side by side with results obtained with the Microshield package.

Table 1 – Net mass and heights of sorbent and water in waste drums.

<b>Drum No.</b>	<b>Content</b>	<b>Water height (cm)</b>	<b>Sorbent height (cm)</b>	<b>Drum net mass (kg)</b>
1	Resin	0	51	123
2	Charcoal	56	50	172
3	Resin	0	42	94
4	Charcoal	0	21	46
5	Charcoal	8	53	136
6	Charcoal	58	59	188
7	Charcoal	66	59	197
8	Charcoal	0	52	118
9	Charcoal	0	50	115
10	Resin	0	28	53
11	Resin	48	33	133
12	Resin	67	26	192
13	Charcoal	73	36	204
14	Resin	56	56	161
15	Charcoal	72	40	206
16	Resin	58	49	157
17	Charcoal	54	16	150
18	Charcoal	48	19	138
19	Charcoal	70	37	212
20	Charcoal	37	~ 0	92
21	Charcoal	43	11	126

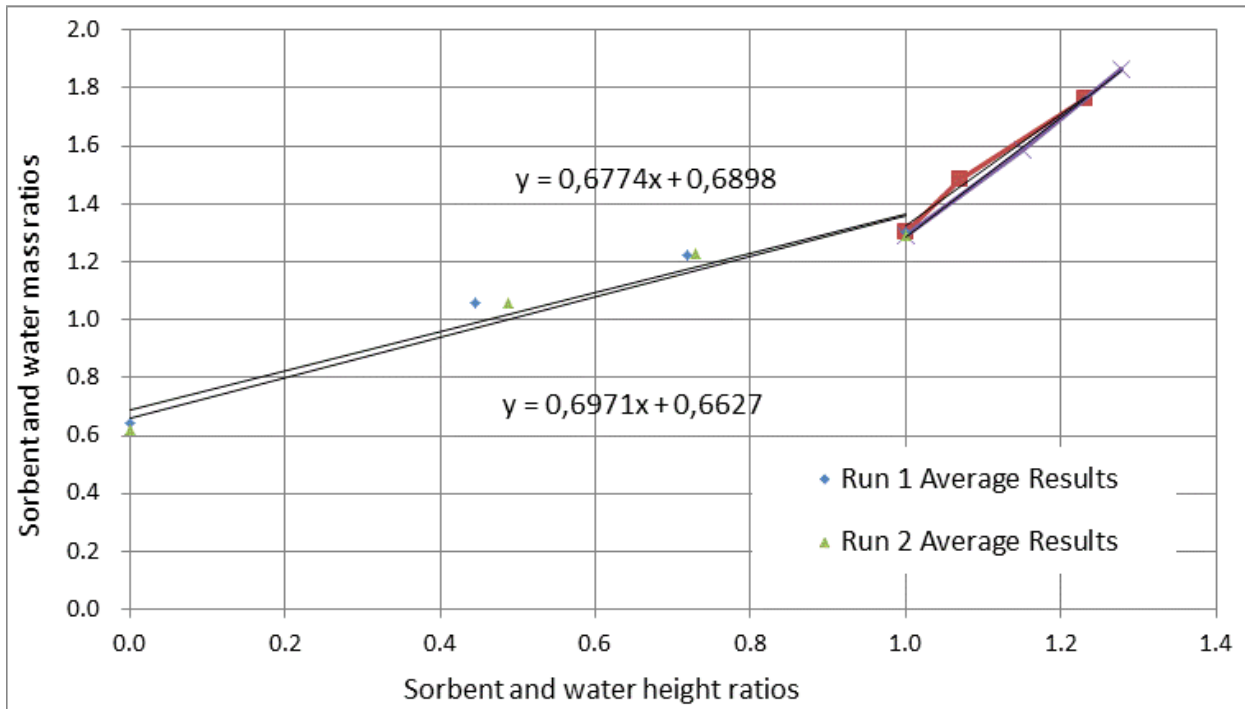


Figure 4- Relationship between reasons of heights and water-absorbing resin mass.

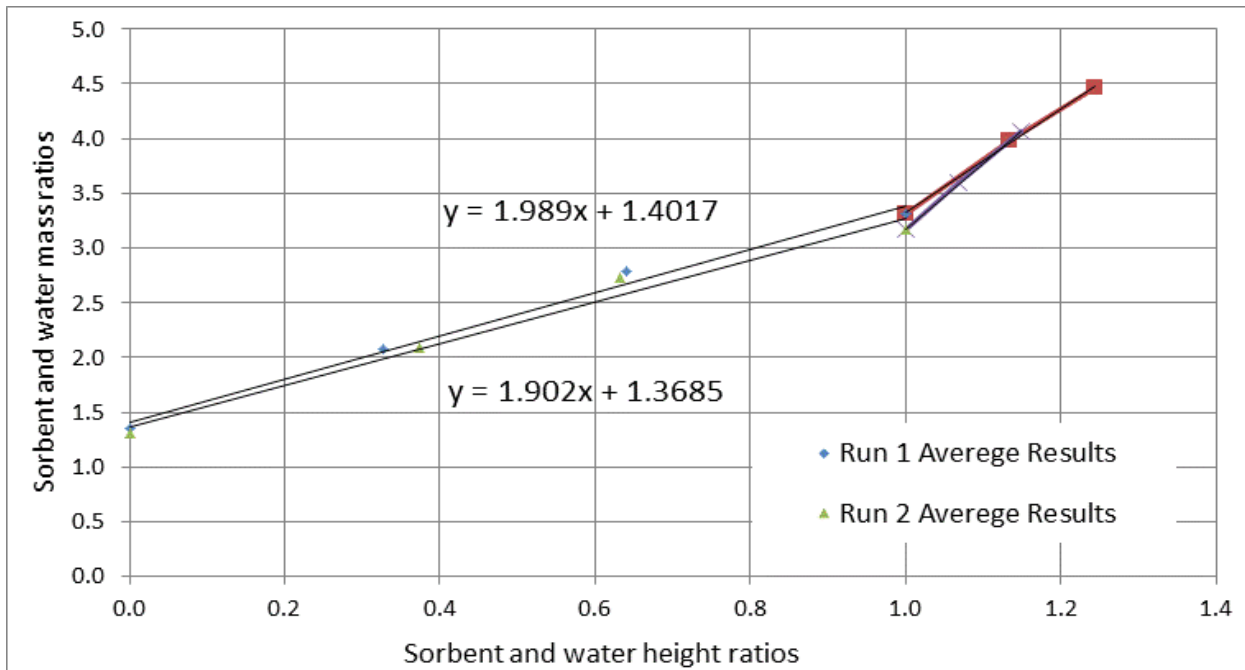


Figure – 5 Relationship between reasons of heights and water-absorbing mass of charcoal.



Table 2 – Mass, activity concentration and total activity of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ 

<b>Drum No.</b>	<b>Mass of water (kg)</b>	<b>Mass of sorbent (kg)</b>	<b><math>^{137}\text{Cs}</math> (Bq.g<sup>-1</sup> dry)</b>	<b><math>^{60}\text{Co}</math> (Bq.g<sup>-1</sup> dry)</b>	<b>Total Activity <math>^{137}\text{Cs}</math> (Bq)</b>	<b>Total Activity <math>^{60}\text{Co}</math> (Bq)</b>
1	0	73	2.0E+01	3.9E+02	1.5E+03	2.9E+07
2	121	36	5.0E+01	2.3E+03	1.8E+06	8.3E+07
3	38	56	1.4E+01	2.9E+02	8.0E+05	1.6E+07
4	27	19	6.4E+01	1.8E+03	1.2E+06	3.5E+07
5	85	51	9.8E+01	2.4E+03	5.0E+03	1.2E+05
6	144	44	4.4E+01	1.7E+03	1.9E+06	7.4E+07
7	138	42	1.2E+02	3.5E+03	4.8E+06	1.5E+08
8	69	49	8.3E+01	3.3E+03	4.1E+06	1.6E+08
9	67	48	8.1E+01	2.7E+03	3.9E+06	1.3E+08
10	21	32	1.1E+03	5.2E+04	3.6E+07	1.7E+09
11	55	41	1.7E+02	3.1E+04	6.9E+06	1.3E+09
12	47	34	1.7E+02	2.1E+04	5.7E+06	7.4E+08
13	87	26	6.1E+01	2.0E+04	1.6E+06	5.2E+08
14	92	69	1.1E+02	1.9E+04	7.4E+06	1.3E+09
15	106	22	1.3E+02	1.1E+04	2.9E+06	2.4E+08
16	77	57	5.1E+02	2.7E+04	2.9E+07	1.6E+09
17	43	13	1.1E+02	2.4E+04	1.4E+06	3.1E+08
18	52	16	1.6E+02	1.8E+04	2.6E+06	2.9E+08
19	101	30	8.7E+01	2.0E+04	2.6E+06	5.9E+08
20	54	38	0.0E+00	5.6E+01	0.0E+00	2.1E+06
21	36	11	7.3E-01	5.2E+01	7.9E+03	5.6E+05

Table 3 – Measured and calculated air kerma rates.

N° Drum	Measured Air Kerma Rate ( $\mu\text{Gy/h}$ )	Calculated Air Kerma Rate ( $\mu\text{Gy/h}$ )	
		Manual	MicroShield
1	3.2E+00	6.6E+00	8.0E+00
2	1.9E+01	9.8E+00	1.4E+01
3	3.4E+00	4.8E+00	3.9E+00
4	1.0E+01	3.4E+01	8.2E+00
6	2.6E+01	7.5E+00	1.4E+01
7	3.7E+01	1.5E+01	2.7E+01
8	2.7E+01	1.5E+01	3.6E+01
9	3.1E+01	1.6E+01	2.0E+01
10	1.4E+02	3.5E+02	3.5E+02
11	1.6E+02	2.3E+02	1.0E+02
12	1.3E+02	1.6E+02	1.4E+02
13	2.0E+02	8.4E+01	1.0E+02
14	1.6E+02	1.4E+02	2.7E+02
15	6.0E+01	3.6E+01	4.7E+01
16	2.0E+02	1.9E+02	2.8E+02
17	6.0E+01	1.1E+02	8.0E+01
18	3.3E+01	9.0E+01	7.0E+01
19	1.3E+02	9.2E+01	1.2E+02
20	1.5E+01	9.8E-04	7.0E-01
21	5.0E+01	3.0E-01	2.5E-01

## DISCUSSION AND CONCLUSIONS

The large discrepancies observed between calculated and measured results of air kerma rate are consequence of imprecise and or inaccurate input data to the models and, probably, a flaw in the experimental method. Figure 5 shows a sample of these discrepancies in the form of a graph of the calculated and measured air kerma rates for three waste drums.

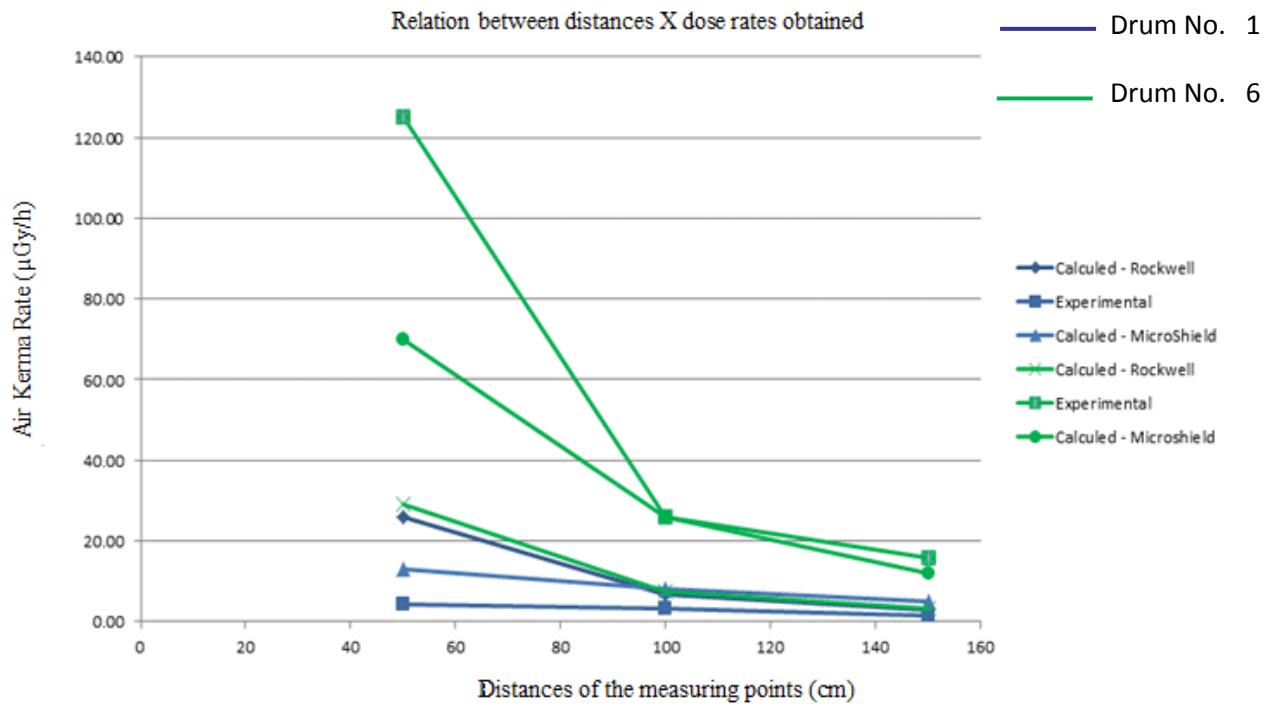


Figure – 6 Correlation between the air kerma rate obtained.

Some intermediate parameters used in the calculations has a great impact on the uncertainty of results because they are obtained by interpolation in graphs of low precision and appear as exponents in the exponential functions that govern the response of the radiation sources to the kerma rates. For the same reason, the inaccuracy of input data like variations in density, thickness and elemental composition of source and shielding materials, result in large variations in the results.

One source of uncertainty lies in the determination of mass and height ratios of sorbent and water. What seemed initially a trivial laboratory experiment was actually a difficult task because it took a time until meaningful results were obtained. A great number of repetitions in the measurement of mass and height ratios systematically failed in reproduce the previous result. The measurement of ‘internal’ water yielded large deviations in the results.

Effects of capillarity prevented to observe the water level in the sorbent packed column. Radiography and neutrongraphy of columns with preset ratios of water and sorbent didn't help to see what the actual water levels were. These experiments will be reported elsewhere, when the results of magnetic resonance of the packed columns are available.

Finally, there is evidence that the methodology must be reviewed. The assumption that the actual ratios are not significantly different from those observed with fresh samples in laboratory is under suspicion. It seems now necessary to use samples of the actual sorbents, with amounts large enough to allow clearly distinguishing the heights of the solid and the liquid phases what will require special provisions in the laboratory because of the exposures and the risks of contamination.

Further characterization work that is already in course is the determination by detailed radiochemical analyses of difficult to measure radionuclides in the waste samples and the correlation of their concentrations with the concentration of selected gamma emitters easily measurable by gamma scanning of the waste packages.

## **ACKNOWLEDGEMENT**

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