



Applicability of the LSC technique for determining ²¹⁰Pb in liquid samples for purposes of occupational dose assessment via *in vitro* bioassay

Sampaio^a C.S., Medeiros^a G.C.O., Mesquita^a S.A., Dantas^a B.M., Sousa^a, W.O.

^a CNEN/IRD/DIDOS, 22783-127, Rio de Janeiro, RJ, Brazil camilla.sampaio@ird.gov.br

ABSTRACT

A new approach for the determination of ²¹⁰Pb by liquid scintillation counting was developed by the LBIOVT. In order to assess the applicability of this technique previously developed for internal monitoring of occupationally exposed workers, some probable scenarios for the intake of ²¹⁰Pb were elaborated and evaluated in this complementary work. The methodology formerly developed provides detection limit values suitable for detecting activities corresponding to committed effective doses below the recording level of 1 mSv in a variety of scenarios. In a monitoring program for occupational exposed workers the methodology could be used in a biannual monitoring interval both for daily urine or faeces collection, either considering scenarios of ²¹⁰Pb intake by inhalation or ingestion, and the detection limits would still be able to detect activities corresponding to a committed effective doses below that the methodology developed is suitable to be used in a monitoring program for occupational exposed workers.

Keywords: in vitro bioassay, ²¹⁰Pb, internal dosimetry.



1. INTRODUCTION

Radioactivity is a natural phenomenon and natural sources of radiation can be found widely distributed in the environment. Radiation and radioactive substances have many beneficial applications, ranging from power generation to uses in medicine, industry and agriculture. The radiation risks to workers and the public that may arise from these applications should be assessed and, if necessary, controlled [1].

²¹⁰Pb is a naturally occurring radionuclide and can be incorporated by humans through food chain or directly by ingestion or inhalation in situations of occupational or accidental exposures. Because of its metabolic and dosimetric characteristics, ²¹⁰Pb becomes an important isotope from the radiation protection point of view [2].

Internal doses cannot be measured directly; they can only be inferred from individual measurements of other quantities, such as measurements of activity in excretion samples. *In vitro* bioassay is an indirect analytical method which identifies and quantifies radionuclides deposited internally in the human body through the analysis of urine and faeces [3]. In that regard, the LBIOVT (*In vitro* Bioassay Laboratory) plays an important role in the DIDOS (Division of Dosimetry) of IRD (Institute of Radioprotection and Dosimetry) in Brazil, from the point of view of occupational radiological protection.

A new approach for the determination of ²¹⁰Pb by liquid scintillation counting (LSC) was developed by the LBIOVT in 2013 [4]. In order to assess the applicability of this technique previously developed for internal monitoring of occupationally exposed workers, some probable scenarios for the intake of ²¹⁰Pb were elaborated and evaluated in this complementary work.

2. MATERIALS AND METHODS

The methodology developed in the previous work consisted of the determination of an efficiency curve as a function of the time elapsed after the separation of ²¹⁰Pb by analyzing two different regions of interest (ROI). One was considering mostly ²¹⁰Pb spectra contributions comprehending ROI [5-350] and the other was considering both ²¹⁰Pb and its daughter ²¹⁰Bi

spectrum contributions comprehending ROI [5-1024]. Both efficiency curves were plotted during a time interval of 60 days. It should be highlighted that the time necessary for ²¹⁰Pb and ²¹⁰Bi to achieve secular equilibrium is 15 days [4].

According to ISO (International Organization for Standardization) the detection limit is the smallest true value of the measurand that is detectable by the measuring method, associated with the statistical test and hypothesis in accordance with the decision threshold, with the probability of the true value being higher than the DL (error of the second kind) is at most equal to 5% ($\beta = 0.05$) [5].

The values of detection limit (DL) considered in this work were the highest ones obtained for the methodology formerly developed, with the sample being analyzed before the secular equilibrium between lead-210 and bismuth-210 is achieved (the highest DL found was at 1 day after the precipitation of ²¹⁰Pb) and the sample being analyzed after the secular equilibrium was reached. The values of detection limits are showed at table 1 [4].

ROI	Detection limit (Bq/L)			
	Before secular equilibrium	After secular equilibrium		
5 - 350	0.030	0.025		
5 - 1024	0.039	0.021		

Table 1: Highest detection limit values for two different ROIs, before and after secular equilibrium between ²¹⁰Pb and ²¹⁰Bi is achieved.

To evaluate the methodology, it was considered that the activity measured in the sample (M) corresponded to the detection limit value corrected to the value of a 24-hour collection. The standard amount considered was 1.6 L for a daily urine collection [6] and 2.79 g of ash for a daily faeces collection, both for adult male [7].

The quantity of primary interest for internal dose is the intake "I" (activity of the radionuclide taken into the body). The value of the intake is obtained by dividing the measured body excretion rate "M" by the intake fraction "m (t)" excreted from the body (via urine or faeces) at time "t" after the intake [8]

$$I = M/m(t) \tag{1}$$

Values of intake fraction excreted from the body are derived from biokinetic models suggested by the International Commission on Radiological Protection (ICRP).

The committed effective dose (E) is estimated by multiplying the value of the intake by e(g), the committed effective dose per unit intake for ingestion or inhalation [8]:

$$E = I \cdot e(g) \tag{2}$$

The commitment period (g) is taken to be 50 years for adults.

ICRP 68 presented committed effective dose coefficient values for occupational workers [9]. Values of m(t) in accordance to ICRP 68 can be obtained through the computer program AIDE (Activity and Internal Dose Estimates) [10]. Recently ICRP introduced changes that affected the calculation of effective dose, which implied in a revision of the dose coefficients for internal exposure [11]. Revised values of the committed effective dose coefficients for ²¹⁰Pb can be found at ICRP 137 and the revised values of intake fraction excreted from the body can be calculated through the supplemental material OIR Data Viewer provided by ICRP [11, 12].

We considered intake scenarios of single incorporation of ²¹⁰Pb. Values of e(g) can be found at table 2 [9, 11]. According to ICRP 78 and ICRP 130 we used values for AMAD 5 μ m for scenarios of incorporation by inhalation [8, 13].

			Inhalation (AMAD 5 µm)			Ingestion	
Nuclide	T _{1/2}	Reference	type	Factor	e (Sv/Bq)	Factor	e (Sv/Bq)
²¹⁰ Pb	22.3 y _	ICRP 68	F	$f_1 = 0.2$	1.1E-06	$f_1 = 0.2$	6.8E-07
	22.3 y -	ICRP 137	F	$f_{A} = 0.2$	7.0E-07	$f_{A} = 0.2$	3.2E-07

Table 2: Committed effective dose coefficients (e) for inhalation and ingestion of radionuclide for

 f_1 : Gatrointestinal absorption fractor

f_A: alimentary tract transfer factor

For workers it is necessary to consider a monitoring program. ICRP 68 and ICRP 78 recommends that, for ²¹⁰Pb, the maximum interval for a monitoring program should be 180 days [8, 9]. Therefore, we evaluated monitoring intervals of 7, 15, 30, 60, 90, 120 and 180 days. It should be highlighted that, in case of occupational exposure, it is considered that the intake occurred in the middle of the interval of the monitoring program, so $m(t_{1/2})$ was used in the calculation (i.e, in an

interval of 30 days we use the m(t) value of day 15) [8]. Furthermore, the detection limits for the monitoring intervals should be one that, when integrated it in a year, do not surpass the recording level of 1 mSv [3].

3. RESULTS AND DISCUSSION

For occupational monitoring of workers it is necessary to implement a routine program, since the total dose over one year period must be recorded if it is equal to or greater than 1 mSv. The simulation results for ²¹⁰Pb inhalation and ingestion by workers according to ICRP 68 are shown in figures 1, 2, 3 and 4 and the simulation results according to ICRP 137 are shown in figures 5, 6, 7 and 8. ICRP 68 recommends, in this case, a maximum monitoring interval of 180 days [9].

The possibility of monitoring through urine samples, referring to a single inhalation of ²¹⁰Pb for workers according to ICRP 68, is represented in Figure 1.

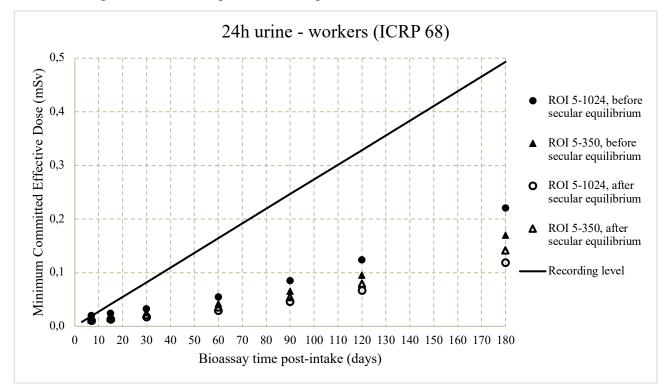


Figure 1: *Minimum detectable committed effective dose variation as a function of time after intake* (scenario: single inhalation with $AMAD = 5 \mu m$, type F, $f_1 = 0.2$, worker, 24h urine collection).

The method developed could be used to control occupational exposure of workers at intervals from 15 days to 180 days ($\beta = 0.05$), for both ROIs, regardless of using the efficiency curve before or after reaching secular equilibrium. If the monitoring interval is 7 days, only the ROI [5-1024] with sample being analyzed before secular equilibrium is achieved should not be used ($\beta = 0.05$).

Figure 2 shows the possibility of monitoring occupational exposure by inhalation of ²¹⁰Pb to workers through daily faeces collection, according to ICRP 68.

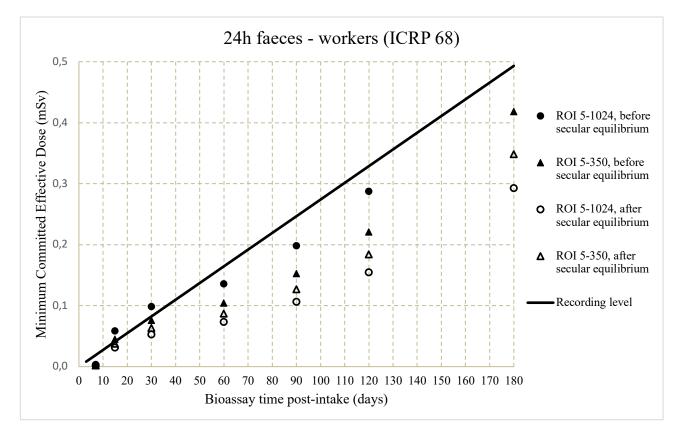


Figure 2: *Minimum detectable committed effective dose variation as a function of time after intake* (scenario: single inhalation $AMAD = 5 \mu m$, type F, $f_1 = 0.2$, worker, 24h faeces collection).

Any proposed methodology could be used to control occupational exposure to workers at frequencies of 60, 90 and 120 days ($\beta = 0.05$).

If the methodology chosen uses ROI [5-1024], with sample being analyzed before the secular equilibrium is achieved, the monitoring intervals that could be used are 60, 90 and 120 days ($\beta = 0.05$). Such procedure allows detecting activities corresponding to committed effective dose

values below the recording level of 1 mSv per year. However, for the same ROI, but with analysis being held after secular equilibrium is reached, all the monitoring intervals should be used ($\beta = 0.05$).

In the case of using ROI [5-350], all monitoring intervals could be used if the sample is analyzed after the secular equilibrium to be achieved ($\beta = 0.05$). The only monitoring interval that should be avoided for ROI [5-350] is 15 days in case the sample is analyzed before secular equilibrium is reached ($\beta = 0.05$).

Figure 3 shows the variation in the minimum detectable committed effective dose for occupational monitoring, using daily urine samples, referring to a single intake of ²¹⁰Pb by ingestion for workers, according to ICRP 68.

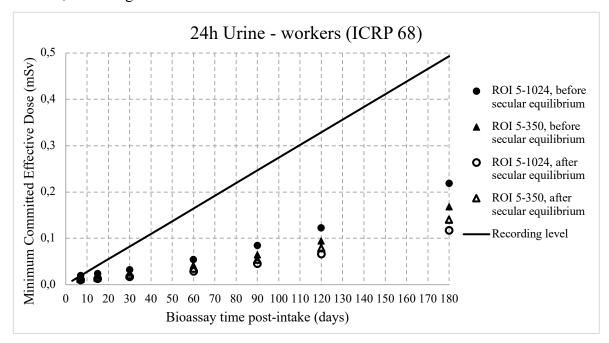


Figure 3: *Minimum detectable committed effective dose variation as a function of time after intake* (scenario: single ingestion with $f_1 = 0.2$, worker, 24h urine collection).

The method developed can be used to control occupational exposure of workers at intervals from 15 days to 180 days, for both ROIs, regardless of using the efficiency curve before or after reaching secular equilibrium ($\beta = 0.05$). If the monitoring interval is 7 days, only the ROI [5-1024] with sample being analyzed before secular equilibrium is achieved should not be used ($\beta = 0.05$). Figure 4 shows the possibility of monitoring occupational exposure to workers through daily faeces collection, according to ICRP 68.

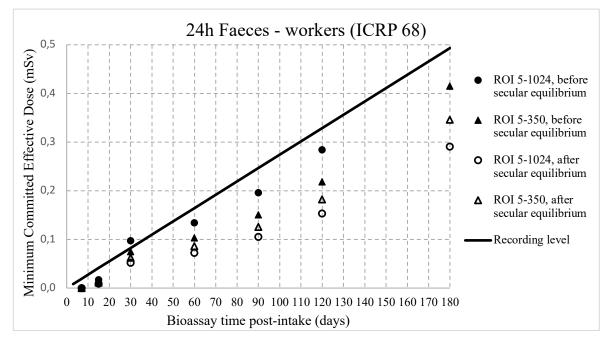


Figure 4: *Minimum detectable committed effective dose variation as a function of time after intake* (scenario: single ingestion with $f_1 = 0.2$, worker, 24h faeces collection).

Any proposed methodology could be used to control occupational exposure to workers at frequencies of 7, 15, 60, 90 and 120 days ($\beta = 0.05$).

If the methodology chosen is uses ROI [5-1024], with sample being analyzed before the secular equilibrium is achieved, the monitoring intervals that could be used are 7, 15, 60, 90 and 120 days ($\beta = 0.05$). Such procedure allows detecting activities corresponding to committed effective dose values below the recording level of 1 mSv per year. However, for the same ROI, but with analysis being held after secular equilibrium is reached, all monitoring intervals evaluated could be used ($\beta = 0.05$).

In the case of using ROI [5-350], all monitoring intervals could be used if the sample is analyzed before secular equilibrium is reached and if the sample is analyzed after the secular equilibrium to be achieved ($\beta = 0.05$).

Figures 5 and 6 show the possibility of monitoring occupational exposure to workers in accordance to ICRP 137, in case of a single intake of ²¹⁰Pb by inhalation.

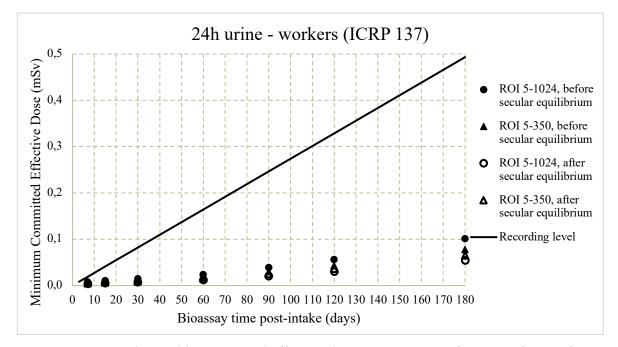


Figure 5: *Minimum detectable committed effective dose variation as a function of time after intake* (scenario: single inhalation with $AMAD = 5 \mu m$, type F, $f_A = 0.2$, worker, 24h urine collection).

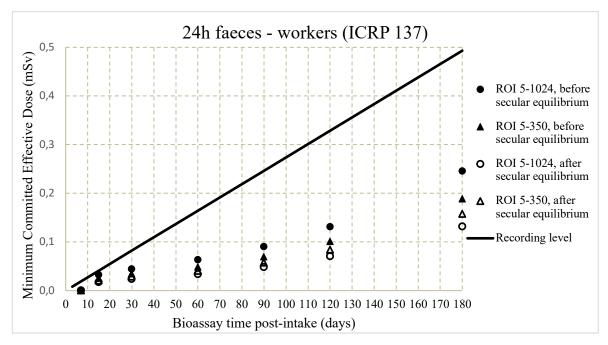


Figure 6: *Minimum detectable committed effective dose variation as a function of time after intake* (scenario: single inhalation $AMAD = 5 \mu m$, type F, $f_A = 0.2$, worker, 24h faeces collection).

Considering the revised parameters in accordance to ICRP 137, the proposed methodology could be used to control occupational exposure to workers in case of a single intake of ²¹⁰Pb by inhalation for daily collection of urine or faeces and for all monitoring intervals, regardless of use of [ROI 5-350] or [ROI 5-1024] (β = 0.05).

Figures 7 and 8 show the possibility of monitoring occupational exposure to workers in accordance to ICRP 137, in case of single intake of ²¹⁰Pb by ingestion.

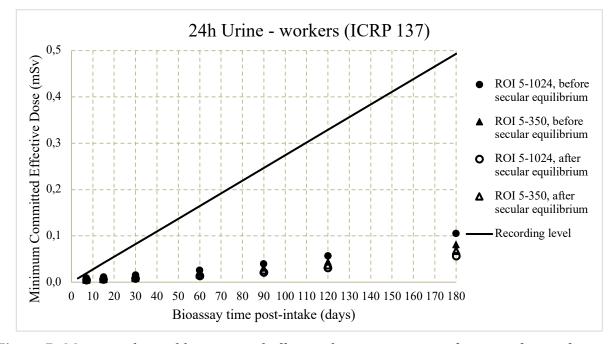


Figure 7: *Minimum detectable committed effective dose variation as a function of time after intake* (scenario: single ingestion with $f_A = 0.2$, worker, 24h urine collection).

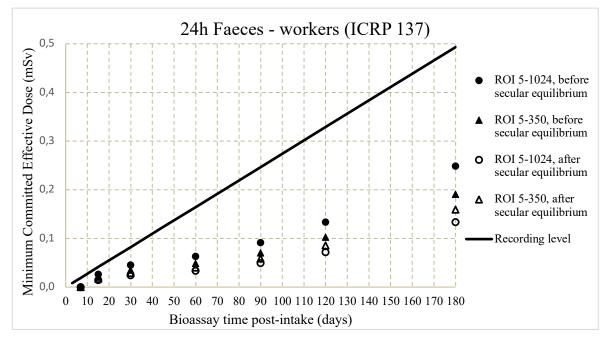


Figure 8: *Minimum detectable committed effective dose variation as a function of time after intake* (scenario: single ingestion with $f_A = 0.2$, worker, 24h faeces collection).

In the same way, considering the revised parameters in accordance to ICRP 137, the proposed methodology could be used to control occupational exposure to workers in case of a single intake of ²¹⁰Pb by ingestion for daily collection of urine or faeces and for all monitoring intervals, regardless of use of [ROI 5-350] or [ROI 5-1024] ($\beta = 0.05$).

4. CONCLUSION

The methodology developed by LBIOVT provides detection limit values suitable for detecting activities corresponding to committed effective doses below the recording level of 1 mSv in a variety of intake scenarios.

Considering the parameters values in accordance with ICRP 68, in a monitoring program for occupational exposed workers (adults, standard men), the methodology developed could be used in a biannual monitoring interval of 24 h urine collection and the detection limits would still be able to detect activities corresponding to a committed effective doses below the recording level, both for inhalation and ingestion intake scenario. In the case of 24h faeces collection, the methodology

developed should still be used in a biannual monitoring program, but only if the methodology of choice would not consider mostly contributions of ²¹⁰Pb spectra [ROI 5-350] and the sample being analyzing before the secular equilibrium between ²¹⁰Pb and ²¹⁰Bi is achieved, either for inhalation or ingestion intake scenario.

However, when considering the revised parameters in accordance to ICRP 137, the methodology developed could be used in a biannual monitoring interval of 24 h urine or faeces collection and the detection limits would still be able to detect activities corresponding to a committed effective doses below the recording level, both for inhalation and ingestion intake scenario, regardless of using [ROI 5-350] or [ROI 5-1024].

It is concluded that the methodology developed is suitable to be used in a monitoring program for occupational exposed workers.

REFERENCES

- [1] IAEA International Atomic Energy Agency. Radiation Protection of the Public and the Environment, IAEA Safety Standards Series No. GSG-8, IAEA, Vienna (2018).
- [2] LAURIA, D. C., CARVALHO, L. L., CONTI, C. C. Comparison of different methods for ²¹⁰Pb determination in environmental samples. Adv. Liq. Scintillation Spectrom. LSC 2005, 211–216, 2006.
- [3] IAEA International Atomic Energy Agency. Occupational Radiation Protection, IAEA Safety Standards Series No. GSG-7, Vienna (2018).
- [4] SAMPAIO, C. S., MEDEIROS, G. C. O, MESQUITA, S. A., DANTAS, B. M., SOUSA, W. O. A new approach for the determination of ²¹⁰Pb by liquid scintillation counting. Applied Radiation and Isotopes 156 (2020) 108972.
- [5] ISO International Organization for Standardization. Radiation Protection Perfomance criteria for radiobioassay. ISO 28218:2010 (2010).
- [6] ICRP International Commission on Radiation Units and Measurements. Basic Anatomical and Physiological Data for Use in Radiological Protection Reference Values. ICRP Publication 89 (2002).
- [7] JULIÃO, L. M. Q. C., MELO, D. R., SOUSA, W. O., SANTOS, M. S., FERNANDES, P. C. P. Uncertainty on faecal analysis on dose assessment. Radiation Protection Dosimetry 127 (2007) 1-4 pp. 421-424
- [8] ICRP International Commission on Radiation Units and Measurements. Individual Monitoring for Internal Exposure of Workers (preface and glossary missing). ICRP Publication 78 (1997).
- [9] ICRP International Commission on Radiation Units and Measurements. Dose Coefficients for Intakes of Radionuclides by Workers. ICRP Publication 68 (1994).
- [10] BERTELLI, L., MELO, D, R., LIPSZTEIN, J., CRUZ-SUAREZ, R. AIDE: Internal Dosimetry Software Radiation Protection Dosimetry, vol. 130, n. 3, pp. 358-367, 2008.
- [11] ICRP International Commission on Radiation Units and Measurements. Occupational Intakes of Radionuclides: Part 3. ICRP Publication 137 (2017).

- [12] ICRP International Commission on Radiation Units and Measurements. OIR Data Viewer for P134, P137 and P141 v4010419. Supplemental material of ICRP Publication 137 (2019). Available at: https://journals.sagepub.com/doi/suppl/10.1177/ANIB_46_3-4>. Last accessed: 25 Oct. 2021.
- [13] ICRP International Commission on Radiation Units and Measurements. Occupational Intakes of Radionuclides: Part 1. ICRP Publication 130 (2015).