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^{210}PB DATATION REVIEW AND GENERALIZATION OF RADIOCHRONOLOGICAL MODEL

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Abstract

The present investigation reviews the different datation models and their connection to the sedimentation process of ^{210}Pb in soils. The objective is to create a generalizable model capable to add different initial conditions of soil formation. We use the Least Square Fitting Method to predict a radiochronological scale profile obtained through gamma spectroscopy. With soil samples of Reserva Alberto Manuel Brenes, we obtained the chronostratigraphic profile with the highest ^{137}Cs peak at 53 ± 9 years, coinciding with the 1963 atmosphere nuclear weapon test. The formation model that best fits the data is the Constant Initial Concentration model.

Resumen

En la presente investigación, se realizó una revisión de los distintos modelos de datación y su conexión con los procesos de sedimentación de ^{210}Pb en los suelos. El objetivo era crear un modelo generalizado capaz de integrar diferentes condiciones iniciales de formación. Se utilizó el método de ajuste de mínimos cuadrados para predecir la escala radiocronológica de un perfil de profundidad, a partir de la concentración de actividad de ^{210}Pb determinada por espectroscopía gamma. Usando muestras de sedimentos obtenidos de la Reserva Biológica Alberto Manuel Brenes, se obtuvo un perfil cronoestratigráfico, con el pico más alto de ^{137}Cs a una edad de 53 ± 9 años, coincidiendo con las pruebas de armas nucleares en la atmósfera de 1963. El modelo que mejor se ajustó a los datos fue el de Concentración Inicial Constante.

Keywords: ^{210}Pb, ^{137}Cs, Radiochronology, Dating models.

Palabras clave: ^{210}Pb, ^{137}Cs, Radiocronología, modelos de datación.

I. INTRODUCTION

Radiochronology is a nuclear analytical technique used to determine the age of a sample by measuring the activities of different radionuclides. For example, the ^{14}C is used to determine the age of materials up to 15000 years old. Another example is the use of ^{238}U to determine the age of the Earth. By comparing the activity of this radionuclide with the amount of ^{208}Pb in meteorite samples, scientist can estimate the age of the Earth. The importance of dating a sample is to have a reference or proxy for past events on Earth (Bernal et al., 2010).

When compared to those samples, the half-live of ^{210}Pb is 22,3 years. This time is very brief in comparison with the half-lives of ^{238}U (4.468 billion of years) or ^{14}C (5700 years). However, this radionuclide can be used to understand the effects of human activity on the environment. It can date events

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that occurred no longer than 100 to 150 years ago, conveniently in the post-industrial revolution age. (Sanchez-Cabeza & Ruiz-Fernández, 2012).

^{210}Pb can be found in any sedimentary material due to its natural origin in the uranium-radium decay series. The decay chain starts with ^{238}U and ends with ^{208}Pb, which is why this radionuclide is present in sedimentary materials because of natural processes. There are two ways that ^{210}Pb is added to sedimentary materials. Firstly, the nuclide remains in the soil due to the decay of the radium present in the soil (referred to as supported ^{210}Pb). On the other hand, another radionuclide in the chain is the ^{222}Rn, a gas which is emitted from the Earth's surface with a mean flux of 1160 $Bq \cdot m^{-}2 \cdot d^{-1}$ (Appleby, 2001). The fallout of ^{222}Rn in the atmosphere generates ^{210}Pb and the lead deposits in soil varves as airborne particles that precipitates through rainfall or snow. This atmospheric lead is known as "excess ^{210}Pb", because is not in secular equilibrium with ^{226}Ra. For this reason, excess ^{210}Pb can be used to date sedimentary materials using a depth profile of mass against age (Appleby & Oldfield, 1978).

The problem with the datation process is that the flux of ^{210}Pb cannot be considered constant and the same across the Earth's surface. Therefore, a reference age value is necessary. In this case, the ^{137}Cs is used to validate the model. ^{137}Cs is an anthropogenic radionuclide that appeared before atmospheric nuclear tests and nuclear accidents (Jaworowski, 1982). The maximum peak of ^{137}Cs is around 1963 the last year of atmospheric nuclear test. (Sykora & Froehlich, 2009)

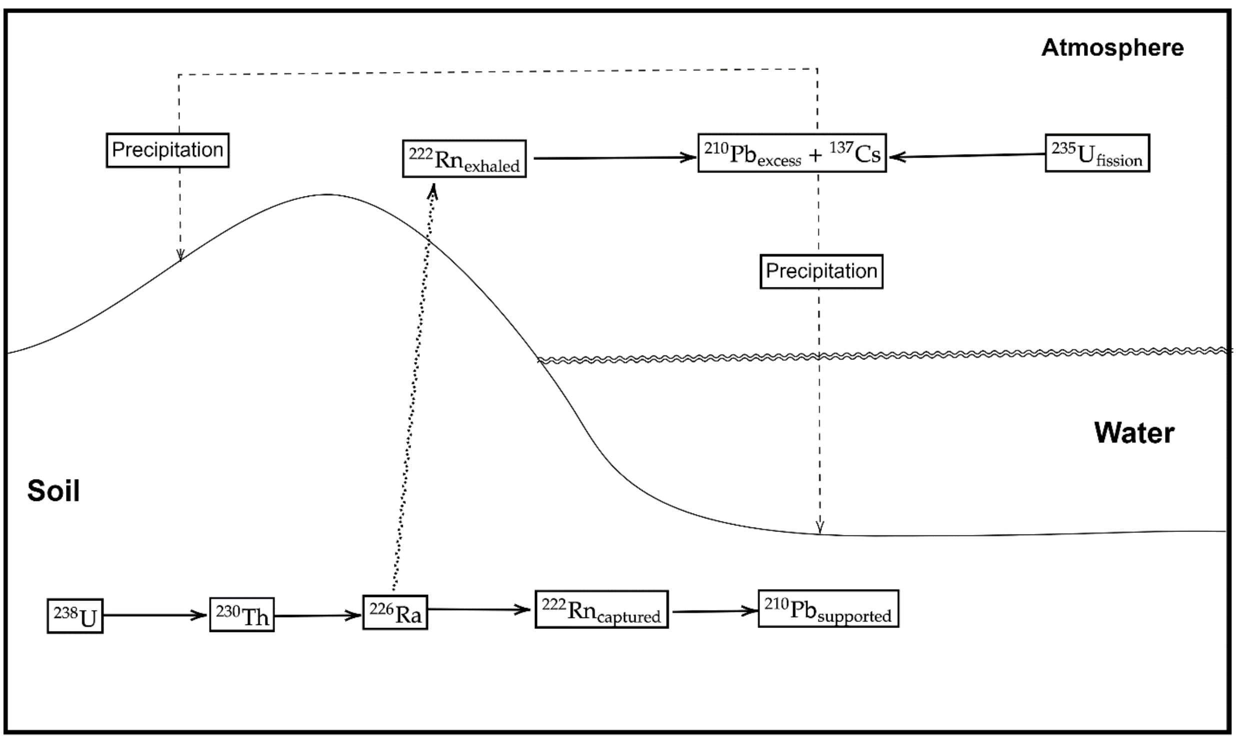

Figure 1. Diagram of addition of ^{210}Pb and ^{137}Cs in sedimentary soils. Figure adapted from (Bernal et al., 2010).

The ^{210}Pb models have the next assumptions (J.-A. Sanchez-Cabeza, Díaz-Asencio, & Ruiz-Fernández, 2012):

- The atmospheric ^{210}Pb has a constant flux to the varves system.
- The atmospheric ^{210}Pb do not go away once it is captured by the sedimentary soils.
- The ^{210}Pb is airborne shorter than captured time in sedimentary soils. Some models are characterized by the initial condition on ^{210}Pb deposition.

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Those assumptions generate the next sedimentary profile to the core of sedimentary soil (J. A. Sanchez-Cabeza & Ruiz-Fernández, 2012).

$$\frac{\mathrm{d}z}{\mathrm{d}t} = \frac{r(t)}{\rho(z)} = s(z). \quad (1)$$

Where, z is the position of the sample from the surface, depth, $r(t)$ is the mass accumulation rate per unit of surface (MAR) [$\mathrm{g \cdot y^{-1} \cdot cm^{-2}}$], $\rho(z)$ is the density of dry sediment [g·], and $s(z)$ is the sediment accumulation rate (SAR) [$\mathrm{cm \cdot y^{-1}}$]. Similarly, the depth mass or accumulative mass m [$\mathrm{g \cdot m^{-2}}$] could be describe by:

$$\frac{\mathrm{d}m}{\mathrm{d}t} = r(m). \quad (2)$$

The activity equation of ^{210}Pb can be described by the disequilibrium of excess ^{210}Pb, and the secular equilibrium of supported ^{210}Pb:

$$\mathrm{A}_{^{210}\mathrm{Pb}}(\mathrm{t}) = \frac{\lambda_{^{226}\mathrm{Ra}}}{\lambda_{^{210}\mathrm{Pb}}} \mathrm{A}_{^{226}\mathrm{Ra}}(0) + \mathrm{A}_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(0) \mathrm{e}^{-\lambda_{^{210}\mathrm{Pb}} t}. \quad (3)$$

The activity of $^{210}\mathrm{Pb}_{\mathrm{excess}}$ can obtain subtracting the activity of the ^{226}Ra form the ^{210}Pb, this equation is the main equation of the model.

$$A_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(t) = A_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(0) e^{-\lambda_{^{210}\mathrm{Pb}} t}. \quad (4)$$

It is possible to generate the profile dating process using the condition described in the equations (1) and (2) into (4). The main problem with datation of samples is the unknown initial activity of the sample. This problem has not a simple solution. For that reason, many models exist considering plenty initial constrains around the initial activity, the flux of ^{210}Pb from the atmosphere, the accumulation of mass and sedimentation rates, the mix of ^{210}Pb into the different sedimentary varves, etc.

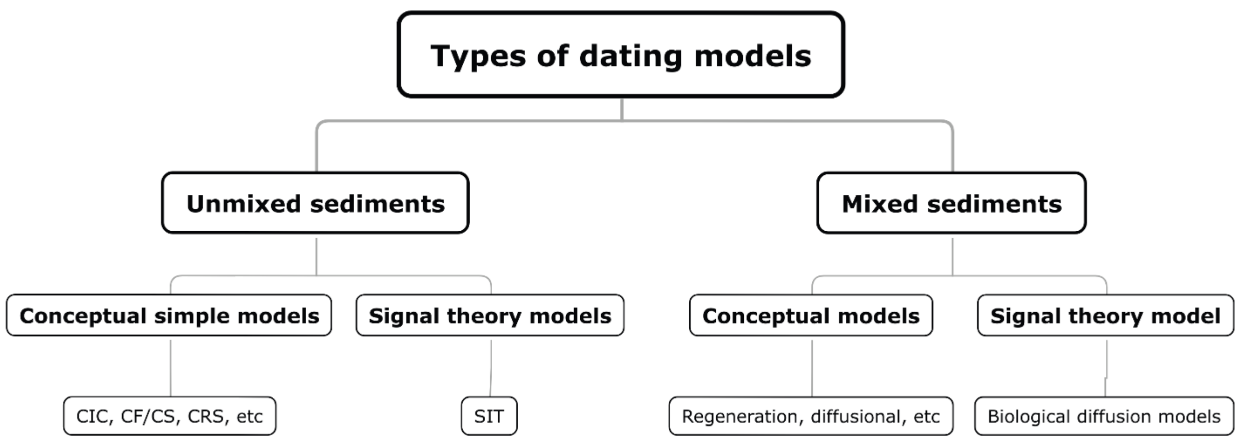

Figure 2. Type of dating models based on the initial condition. Figure adapted from (Carroll & Lerche, 2003).

Figure 2 shows a classification scheme for dating models that considers the movement of ^{210}Pb between the sedimentary layers and the fact that ^{210}Pb is captured by the soil. It can be noticed in the diagram that there are two types of models: conceptual and signal models. The conceptual models only considered theoretical conditions, such as ^{210}Pb flux, mass accumulation rate and diffusion of ^{210}Pb throw the soil. On the other hand, the signal models incorporate information about sedimentation conditions and include Fourier terms in the chronological profile to account for the internal effects in sedimentary cores that are not considered in the conceptual models.

In this paper, a review on the conceptual unmixed models is developed. The purpose of this review to provide evidence for a generalized initial condition model, considering the MAR and SAR. The following table summarizes the models used in the research and their conditions.

Table 1. Description of the conceptual unmixed sediment models used for a generalized initial condition model.

Model	Accumulation type	Initial conditions	Equation
Constant Initial Concentration (CIC)	Sediment accumulation rate	^{210}Pb initial concentration activity	$C_{CIC} = k_{CIC} e^{-\lambda_{^{210}Pb} t}$
Constant sedimentation rate (CRS)	Sediment accumulation rate	^{210}Pb flux to the sedimentary soil	$I_{CRS} = \frac{k_{CRS}}{\lambda_{^{210}Pb}} e^{-\lambda_{^{210}Pb} t}$
Constant flux – Constant sedimentation (CF/CS)	Mass accumulation rate	^{210}Pb flux to the sedimentary soil Mass accumulation rate	$C_{CF/CS} = \frac{k_{CF/CS}}{r} e^{-\lambda_{^{210}Pb} \frac{m}{r}}$

$I_{CRS} = \int_{\infty}^{z} \rho(z') A_{^{210}Pb_{excess}}(z) dz'$ is the ^{210}Pb inventory.
k_{CIC} is the ^{210}Pb initial concentration [Bq·kg^{-1}].
k_{CRS} is the ^{210}Pb flux [Bq·y^{-1}·cm^{-2}], the flux relates with the concentration activity and mass accumulation rate by $f = r \cdot C_{^{210}Pb}$.
$k_{CF/CS}$ is the ^{210}Pb flux [Bq·y^{-1}·cm^{-2}].

The purpose of the research is to date a sedimentary soil core and compare the results of the different models applied in the sample core extracted from Alberto Manuel Brenes Biological Reserve.

II. MATERIALS AND METHODS

First, the sedimentary samples were extracted at Alberto Manuel Brenes Biological Reserve (RBAMB), in San Ramón, Costa Rica. The samples were recollected at (-84,5987, 10,2216). The sedimentary core was extracted in the following way: first, the interaction surface between the soil-atmosphere was removed, and then layers were taken every 3 cm to make a trench hole. The sedimentary samples were then stored with a code containing the extraction date, the location, and the depth of extraction.

The activity of the sedimentary samples was measured with gamma spectroscopy technique using a coaxial negative configuration of hyperpure germanium detector (HPGe). The geometry of the sedimentary samples was 500 mL and 200 mL Marinelli geometry.

The activity data was analyzed with the initial conditions of the models described in the table 1. The analysis of the models was carried out using the least squared method for a linear fitting with the activity concentration of the excess ^{210}Pb with the depth, and the inventory of ^{210}Pb with the depth, and using the concentration activity of ^{137}Cs to compare the ages brought by CIC and CRS model.

III. RESULTS AND DISCUSSION

The proposed model is based on the MAR and SAR conditions, with the idea of finding a generalization that separates the initial condition constraints from the chronological information. In the introduction, certain conditions were mentioned, and the goal is to find a transformation that can use these initial conditions to establish a correlation. First, supposed the following transformation for the excess lead into SAR condition.

$$\xi_{^{210}Pb_{excess}}(z) = \int_{z}^{\infty} w_i(z') C_{^{210}Pb_{excess}}(z')\, dz' = \xi_{^{210}Pb_{excess}}(0) e^{-\sigma(z)}. \quad (5)$$

Where, $\xi_{^{210}Pb_{excess}}(z)$ is the transformation proposed, $w_i(z)$ is a weight transformation kernel, $\xi_{^{210}Pb_{excess}}(0)$ is the initial constrain, $\sigma(z) = \lambda_{^{210}Pb_{excess}} t$ is the chronological condition. $\sigma(z)$ has the following conditions. First, $z = 0 \Rightarrow \sigma(z) = 0$, it means the chronology at surface must be null. In the hand, $z \rightarrow z_{max} \Rightarrow \sigma(z) \rightarrow$

150y, this z_{max} is the maximum depth where excess ^{210}Pb activity could be measured. z_{max} could be considered ∞. The kernel is unknown, for that reason, it is necessary to define it. First, the $\sigma(z)$, described for the SAR condition, is:

$$\sigma(z) = \lambda_{^{210}\mathrm{Pb}} \int_0^z \frac{\rho(z)}{r(z)} \mathrm{d}z. \tag{6}$$

Considering the follow differential equation,

$$w_i(z) C_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(z) = -\,\xi_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(0) e^{-\sigma(z)} \frac{d\sigma}{dz} \tag{7}$$

Using (6) in (7), the weight kernel must be:

$$w_i(z) = -\,\xi_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(0) \lambda_{^{210}\mathrm{Pb}} \frac{\rho(z)}{f(z)}. \tag{8}$$

The weight kernel has a problem, it is the initial constrain, but it can be solved be iterative analysis or considered a weighted transformation. It means:

$$\bar{\xi}_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(z) = \int_z^\infty W_i(z') C_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(z')\, \mathrm{d}z' = e^{-\sigma(z)}. \tag{9}$$

In this case, the $W_i(z)$ is defined:

$$W_i(z) = -\lambda_{^{210}\mathrm{Pb}} \frac{\rho(z)}{f(z)}. \tag{10}$$

This solution is completely chronological.

For MAR condition, it is possible to develop the same treatment. However, It is necessary to consider that the depth must be change by accumulative mass, as it is not a merely change of variable. First, the chronology for MAR is:

$$\nu(m) = \lambda_{^{210}\mathrm{Pb}} \int_0^m \frac{1}{r(m')} dm' . \tag{11}$$

With this mass accumulative chronology, it is possible to define the transformation and the weight kernel. Those are:

$$\mu_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(m) = \int_m^\infty p_i(m') C_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(m')\, \mathrm{d}m' = \mu_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(0) e^{-\nu(m)}. \tag{12}$$

The weight kernel for accumulative mass must be:

$$p_i(z) = -\,\mu_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(0) \frac{\lambda_{^{210}\mathrm{Pb}}}{f(m)}. \tag{13}$$

And the weighted transformation is defined by:

$$\bar{\mu}_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(m) = \int_\infty^m \frac{\lambda_{^{210}\mathrm{Pb}}}{f(m')} C_{^{210}\mathrm{Pb}_{\mathrm{excess}}}(m')\, \mathrm{d}m' = e^{-\nu(m)}. \tag{14}$$

The proposed model can reproduce any conceptual model with some consideration for the weight kernel function. The proof is summarized in the following table.

Table 2. Summary of the generalization weight kernel for the conceptual unmixed models.

Model	Accumulation type	Weight kernel function
Constant Initial Concentration (CIC)	SAR	$W_i(z) = \frac{\lambda_{^{210}Pb}\rho(z)}{k_{CIC} r(z)}$
Constant sedimentation rate (CRS)	SAR	$W_i(z) = \frac{\lambda_{^{210}Pb}\rho(z)}{k_{CRS}}$
Constant flux – Constant sedimentation (CF/CS)	MAR	$P_i(z) = \frac{\lambda_{^{210}Pb}}{k_{CF/CS}}$

The data obtained by gamma spectroscopy of RBAMB sedimentary soil samples, was analyzed using SAR models, considering the following condition in the chronological equation (6).

$$\frac{\rho(z)}{r(z)} = M \tag{15}$$

Where M is constant and the slope of the linearized transform equation. For the CIC model, the linear equation is:

$$ln\left(C_{^{210}Pb_{excess}}(z)\right) = -\frac{\rho}{r} z + ln\left(C_{^{210}Pb_{excess}}(0)\right) \tag{16}$$

Similarly, the equation for CRS model is:

$$ln\left(I_{^{210}Pb_{excess}}(z)\right) = -\frac{\rho}{r} z + ln\left(I_{^{210}Pb_{excess}}(0)\right) \tag{17}$$

This is linear for the inventory, but it may not be linear for the concentration of activity, as in the CIC model. The linear regression analysis provided the following data in table and figure 3. This analysis represents preliminary results for a master's thesis.

Table 3. Comparison from the CIC and CRS models analysis on smoothed data obtained at Alberto Manuel Brenes Biological Reserve.

Depth z_i^c (cm)	Analyzed data							
	Concentration of activity			Mass accumulation rate		Dating		
	Data	CIC	CRS	CIC	CRS	CIC	CRS	Δt
	(Bq·kg^{-1})			(kg·m^{-2}·y^{-1})		y		
3	189,92	227,50	346,88	1,59	1,25	1999	1994	5
6	153,13	132,53	160,28	1,73	1,35	1982	1972	10
9	95,92	77,21	72,86	1,90	1,49	1964	1950	14
12	53,09	44,98	35,07	1,98	1,55	1947	1928	19
15	19,90	26,20	17,55	1,99	1,56	1930	1905	35
18	12,23	15,26	9,00	1,94	1,52	1912	1883	29

The data is represented in the following graph.

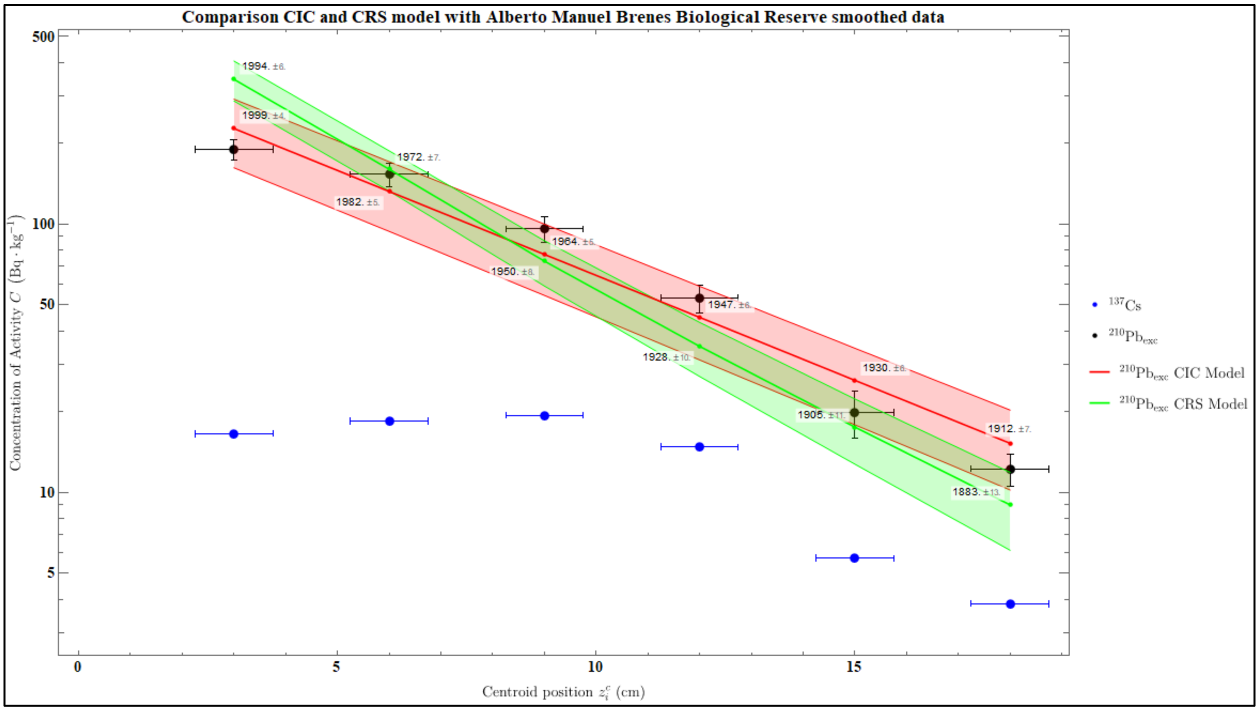

Figure 3. Comparison from the CIC and CRS models analysis on smoothed data obtained at Alberto Manuel Brenes Biological Reserve.

Using the references of ^{137}Cs, it is possible to determine that the CIC model fits better with this profile. It is important to note that this result is only valid for this profile. Other profiles may give different results, but under similar conditions, it is probable that another profile fits better with the CIC model.

A conclusion of this work is that the dating of the sediment fits better with the CIC model. Another observation is the capacity of the proposed model to simulate the behavior of other similar models and provide a simple and unique analysis for dating another sedimentary soil.

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