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Use of nuclear techniques in soil science: A literature review of the Brazilian contribution

Luiz Fernando Pires[(1)*] (iD), **Fábio Augusto Meira Cássaro**[(1)] (iD) **and Vladia Correchel**[(2)] (iD)

[(1)] Universidade Estadual de Ponta Grossa, Departamento de Física, Ponta Grossa, Paraná, Brasil.
[(2)] Universidade Federal de Goiás, Departamento de Solos, Goiânia, Goiás, Brasil.

* Corresponding author:
E-mail: lfpires@uepg.br

ABSTRACT: This review presents the basic research and some applications of the gamma-ray attenuation, neutron gauges, and ^{137}Cs fallout techniques for studying soil physical processes and properties. The selected studies aimed to give the readers a general idea of the use of these nuclear techniques carried out by Brazilian researchers in the past decades. It is expected to assist future researchers by identifying knowledge gaps and opportunities for applying the methods presented here. Around 100 studies were selected for this review. The papers dealing with gamma-ray attenuation are mostly related to the analysis of soil radiation interactions, the measurement of basic soil physical properties, the evaluation of hydraulic conductivity, water retention curve, and soil mechanical analysis. Neutron gauge applications are related to monitoring the water distribution and balance at the field scale, procedures for calibrating the existing gauge for the Brazilian soils, and analyzing the spatial and temporal variability of the soil water content. The ^{137}Cs methodology involves studies about the erosion and sediment deposition in small watersheds and riparian zones, the spatial variability of ^{137}Cs inventories at reference sites, and the measurement of sediment spatial distributions. Future studies with the gamma-ray attenuation methodology should focus on a better comprehension of the photon interaction with the soil and a correct selection of photon energies to investigate contrasting soils. This is mandatory for adopting it as a reliable tool for soil characterization. This review also revealed that the challenges for the future use of the ^{137}Cs fallout technique involve the continuous decrease of the ^{137}Cs activity worldwide, including in Brazil. Advances in detection systems (gamma spectrometers) will be required to overcome this issue. Future studies should focus on the use of correction factors related to the enrichment of fine particles during the transport of sediments to improve the estimates obtained through the conversion models. The use of neutron gauges to detect soil water content at the field scale depends on the adoption of reliable calibration curves. Then, comprehending how soil properties alter this curve and how it affects the water balance is a study of great interest. Motivated by strict regulations on the use of radioactive materials, the adoption of gauges with less activity is becoming a new goal. Thus, the development of more effective systems of neutron detection is crucial.

Keywords: gamma-ray attenuation, ^{137}Cs fallout, neutron gauge, soil physical properties, soil erosion, soil structure.

INTRODUCTION

The discovery of radioactivity was followed by the discovery of X-rays in 1895 by the German physicist Wilhelm Röntgen. In the year of 1896, the French physicist Henri Becquerel, after several studies analyzing the phosphorescence of uranium minerals, drew to the conclusion that the uranium itself caused the emission of light by these minerals. This was the first evidence of the existence of radioactivity (L'annunziata, 2016; Pires, 2021). Few years after the work of Becquerel, the physicists Marie and Pierre Curie carried out several studies with uranium minerals. Marie Curie quickly noticed that the radioactivity emitted by uranium minerals was directly linked to the amount of uranium in them. The analysis of a special material known as uranium ore pitchblende makes the Curies concluded that it contained another radioactive component. Marie and Pierre isolated two new radioactive elements; the first was called Polonium, and the second one was Radium. These discoveries open the gates for a new area of knowledge named radiochemistry (L'annunziata, 2016). Because of the radioactivity discovery, Becquerel and the Curies won the Nobel Prize in Physics in 1903. Some years later, Marie Curie was honored again with a second Nobel Prize in 1911, this time in Chemistry, for her contributions to new advancements in this area of knowledge (L'annunziata, 2016; Pires, 2021).

Some decades after the pioneering studies in radioactivity, experiments using nuclear techniques started in soil science. To mention some of them, gamma-ray attenuation, neutron gauges, and monitoring radioactivity materials deposited in the soil have been employed to complement other conventional techniques in agricultural experimentation (Visvalingam and Tandy, 1972; Ritchie and McHenry, 1990; Pires, 2018). Among the nuclear techniques utilized in soil science, the gamma-ray attenuation method (GAM) has been widely applied in the field as well as in laboratory studies through transmission and scattering experiments. The pioneering studies using GAM were made in the 1950s. The technique was first used for measuring the soil bulk density, for analyzing appropriate beam geometries, for developing detection and electronic instrumentation, and for investigating the best radioactive sources to be employed in environmental studies (Vomocil, 1954; Bernhard and Chasek, 1955).

Another nuclear technique regularly utilized for soil investigation is related to the use of neutrons and their interactions with the soil. The technique is mainly employed for monitoring the soil water content at field conditions. In combination with other conventional methods, it can determine soil water retention curves, soil water storage, soil water balance, etc. (Visvalingam and Tandy, 1972). The development of neutron gauges and the principles related to their use, for measuring volumetric soil water content, were made at the beginning of the 1950s (Belcher et al., 1950; Gardner and Kirkham, 1952). The use of neutron gauges has been an alternative for measuring the soil water content in long-term experiments. It is mainly due to the little disturbance caused in the soil profile by introducing the access tubes, the possibility of continual measurements at different soil depths, and the small-time consumed for each measurement, usually a few minutes (Kirda and Reichardt, 1992).

Radioactivity found in the soil can also be utilized for evaluating the soil erosion and movement of sediments at different natural conditions. Techniques based on the analysis of the proportion of ^{137}Cs (Cesium-137) retained in the soil have been successfully employed to study soil erosion, which is a significant environmental issue (Didoné et al., 2019). The use of the fallout of radioisotopes to monitor soil erosion had started several decades ago, more specifically at the end of the 1950s. Strontium-90 (^{90}Sr) was the first radioisotope indicated to study the movement of soil sediments (Menzel, 1960). One of the first analyses using Cesium-137 was proposed by Yamagata et al. (1963) in studies of particle movement from catchments induced by runoff.

Although many basic studies and applications of nuclear techniques have been carried out in the last decades, as will be highlighted in this paper, many other types of research

can be proposed using the methods included here. The possibility of non-destructive and less-invasive measurements in the field and at the laboratory levels represents one of the main advantages of nuclear techniques. Additionally, new advances in instrumentation and electronics are offering new possibilities for the nuclear methods presented here. For instance, the gamma-ray sources can be combined with computed tomographic systems of second or third generations allowing three-dimensional (3D) analyses of large samples, at millimetric or micrometric scales (Pires et al., 2010). The use of gamma-ray photons for imaging soils is preferable as they are monoenergetic, which, in many cases, can avoid artifacts usually found in X-ray imaging of dense materials (Passoni et al., 2015). The high energy of the gamma-ray photons can also permit the analysis of samples of hundreds of millimeters, which enables obtaining representative measurements of soil physical properties independently of the soil composition. The advent of 4D imaging systems is another possibility for studying water dynamic processes that take part inside the soil matrix (water infiltration and water redistribution).

Neutron gauges have been successfully employed to measure the soil water content across soil profiles in long-term experiments. Nonetheless, in tropical soils, the accuracy of its measurements is highly dependent on its calibration. This fact requires the development of experimental methods to perform the gauge calibration, for example, in soils with contrasting compositions and stratified soils (Bacchi et al., 2002). Neutron imaging, not covered in this review, is another possibility of the utilization of neutron sources in soil science. The high effective cross-section of hydrogen for neutron dispersion makes these nuclear particles appropriate to monitor the soil water retention and movement and quantify the organic matter distribution inside the soil profile.

Cesium-137 isotopes, which were deposited on the soil surface in the first fallouts that happened in the 1950s and 1960s, have been used as a tracer element in studies of soil erosion and deposition in the last decades. Nevertheless, its relatively short half-life (around 30 years) has been limiting its use as a tracer material, mainly in the Southern Hemisphere (Poreba, 2006). Therefore, some efforts are required to enhance efficiency in detecting the gamma-ray photons emitted by these radioisotopes adsorbed by soil particles. The development of new models for estimating erosion rates using the ^{137}Cs is another research field of interest that deserves attention. Also, the use of ^{7}Be as a soil redistribution tracer, not covered in this review, represents another interesting possibility due to its continuous formation at the upper atmosphere as well as its rapid adsorption by soil particles (Pinto et al., 2013).

This review aimed to give an overview of the use of some nuclear techniques in soil science, carried out by Brazilian scientists in the last decades. The main objectives covered here involve: 1) to summarize the main applications of nuclear techniques to evaluate soil physical properties and processes; 2) to discuss possible advantages and disadvantages of the methods presented; and 3) to give future perspectives about the use of the nuclear techniques to analyze the soil environment.

BASIC THEORETICAL BACKGROUND

Process of radiation interaction

Understanding the gamma-ray interaction with the matter is crucial to comprehend the attenuation and detection of these kinds of photons by nuclear detection systems. According to Beer-Lambert's attenuation law, the emerging intensity I (count rate) of a collimated gamma-ray beam, whose incident intensity is I_0 (count rate), is given by (Kaplan, 1963):

$$I = I_0 e^{-\kappa x} \hspace{6cm} \text{Eq. 1}$$

in which κ (cm^{-1}) represents the linear attenuation coefficient, which measures the photon absorption or scattering probability per unit length while interacting within the sample and x (cm) is the absorber length. The term I/I_0 represents the gamma-ray transmission ratio. This research manuscript presents the units as they usually appear in soil science research papers, regardless of the International System of Units.

The analysis of equation 1 shows that the transmission decreases with the increase in the absorber thickness. When the absorber thickness remains unchanged, increases in the photon energy will induce increases in the transmission intensity. However, it is important to highlight that the linear attenuation is dependent on the physical state of the material. Thus, to avoid this dependence, a new parameter can be defined, the mass attenuation coefficient, which is the linear attenuation coefficient divided by the density of the material (Ferraz and Mansell, 1979). In terms of the mass attenuation coefficient, equation 1 can be expressed as follows:

$$I = I_0 e^{-\mu\rho x} \qquad\qquad \text{Eq. 2}$$

in which μ (cm^2 g^{-1}) and ρ (g cm^{-3}) are the mass attenuation coefficient and the density of the absorber, respectively.

The main parameters responsible for the attenuation of gamma-ray photons by the matter are the atomic number of the absorber, its density, and the photon energy. The total mass attenuation coefficient appears as the result of the contribution of the individual attenuation coefficients (Equation 3) related to the materials the beam encounters in its path (Medhat et al., 2014). These individual attenuation coefficients are due to four main physical processes: the photoelectric absorption, coherent (Rayleigh effect) and incoherent scatterings, and pair production.

$$\mu = \mu_{PA} + \mu_{CS} + \mu_{IS} + \mu_{PP} \qquad\qquad \text{Eq. 3}$$

in which the subscripts PA, CS, IS, and PP stand for the contribution of the photoelectric absorption, coherent scattering, incoherent scattering, and pair production to the total mass attenuation coefficient.

Figure 1 illustrates the total mass attenuation coefficient variation with photon energy (from 10^{-3} to 10^5 MeV) for a representative sandy soil and the contribution of the partial attenuation coefficients (PA, CS, IS, PP). The mass attenuation coefficient for each of the three main partial effects can also be written in terms of their cross-sections (Equation 4):

$$\mu_{(IS,PA,PP)} = \frac{N_A}{A}\, \sigma_{(IS,PA,PP)} \qquad\qquad \text{Eq. 4}$$

in which N_A (mol^{-1}) is the Avogadro's number, A (g mol^{-1}) and Z are the atomic weight and the atomic number of the absorber, and σ (cm^2) is the cross-section. It is important to mention that the cross-section represents a measurement of the probability of the gamma-ray interaction with a single atom.

Photoelectric absorption is important for low photon energies (<50 keV) and depends greatly on the chemical composition of the soil. The incoherent scattering predominates for medium photon energies (50 keV to 1 MeV), influenced mainly by the physical density of the absorber (Akkurt et al., 2005).

The total cross-section for the photoelectric absorption (σ$_{PA}$) is given by equation 5.

$$\sigma_{PA} \propto Z^{4-5} E^{-3.5} \qquad\qquad \text{Eq. 5}$$

in which Z and E (eV) are the atomic number and photon energy, respectively (Kaplan, 1963; Jackson and Hawkes, 1981).
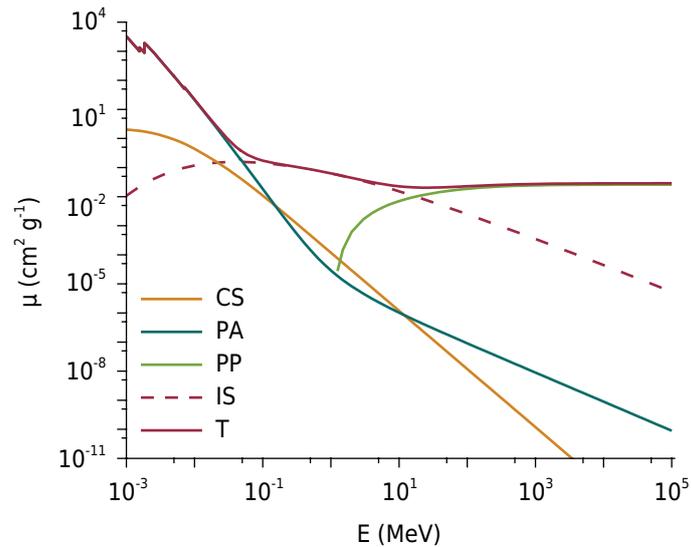
Figure 1. Total (T) mass attenuation coefficient (μ) variation with photon energy (E) for a representative sandy soil and the contributions of the photoelectric absorption (PA), coherent (CS) and incoherent (IS) scatterings, and pair production (PP).

In contrast to photoelectric absorption, the process of incoherent scattering suffers a great influence on the electron density of the absorber. The linear attenuation coefficient for this process is defined according to equation 6.

$$\kappa_{IC} = n_e \sigma_{IC} \qquad \text{Eq. 6}$$

in which n_e (electrons per cm^3) is the electron density and σ_{IC} (cm^2) is the total cross-section for the process of incoherent scattering derived from the Klein-Nishina equation (Cruvinel and Balogun, 2006).

Vinegar and Wellington (1987) presented a generalized equation (Equation 7), which demonstrates the dependence of the linear attenuation coefficient with the density and the atomic number of the absorber. This equation is expressed by:

$$\kappa = \rho \left[z + w \left(\frac{Z^{3.8}}{E^{3.2}} \right) \right] \qquad \text{Eq. 7}$$

in which ρ (g cm^{-3}) is the density of the absorber and z and w are two energy-dependent constants.

Hainsworth and Aylmore (1983) demonstrated the existence of a linear relationship between the linear attenuation coefficient and soil physical properties (Equation 8):

$$\kappa = (SV_s)\kappa_s + (SV_w)\kappa_w \qquad \text{Eq. 8}$$

in which SV_s (cm^3 cm^{-3}) and SV_w (cm^3 cm^{-3}) are the relative solid volume and the volumetric water content, respectively; and κ_s (cm^{-1}) and κ_w (cm^{-1}) are the linear attenuation coefficient of the solid and water fractions (Rogasik et al., 1999). The air fraction is not considered due to its negligible attenuation properties.

Process of neutron interaction

Neutron gauges have as neutron sources a combination of an alpha particle emitter, like, for instance, Americium (Am), and another element like, for example, Beryllium. Beryllium acts as a target for the alpha particles emitted by the Americium. In this interaction, depicted in equation 9, fast neutrons plus Carbon atoms are produced:

$$_2^4\alpha + {}_4^9Be = {}_0^1n + {}_6^{12}C \qquad \text{Eq. 9}$$

in which α, Be, n, and C represent the alpha particle, the Beryllium atom, the neutron particle, and the Carbon atom, respectively.

Americium radioisotope emits alpha particles and gamma-rays, which makes it necessary to shield the gauge to protect the user from this harmful radiation. Neutron gauges are manufactured to allow measurements at the soil surface (surface neutron gauges) or different depths by inserting aluminum access tubes through the soil profile. The choice of aluminum is due to the low interaction of the neutrons with this particular material.

These fast neutrons are moderated, mainly by the water present in the soil profile, which allows their subsequent detection in a slow neutron detector system. Detectors sensitive to slow neutrons such as Boron tri-Fluoride (BF_3) or ^{3}He are usually employed in neutron gauges. Following the detection of the slow neutrons, an electronic counting system is required to count the number of neutrons that reach the detector. The components of the electronics consist basically of an amplifier, high-voltage unit, counter, timer, set of batteries, and microprocessor.

Fast neutrons emitted by the Am-Be combined source will interact with the soil particles and soil water content around the gauge. There are different phenomena through which the neutron can interact (scattering or absorption) with the components of the soil, but due to the average energy of its emission, around 2 MeV, neutron scattering by elastic and non-elastic collisions is the main mechanism of interaction.

Total microscopic cross-section (σ_T) for the occurrence of the interaction of the neutrons (Equation 10) with the components of the soil is given by:

$$\sigma_T = \sigma_{abs} + \sigma_{sca} \hspace{4cm} \text{Eq. 10}$$

in which σ_{abs} (cm^2) and σ_{sca} (cm^2) represent the cross-sections for absorption and scattering, respectively.

Collision of fast neutrons with atoms having masses close to its mass, like hydrogen, reduces the neutron energy (kinetic energy) after a certain number of collisions. For hydrogen, around 18 collisions (on average) are necessary to reduce the neutron energy from 2 MeV to 0.025 eV. It is important to mention that only slow neutrons are relevant for the neutron gauge measurements. On the region of thermalized neutrons (0.01< E <0.5 eV), the absorption cross-section increases with the inverse of the neutron speed (Equation 11) while the scattering cross-section is almost constant (Equation 12):

$$\sigma_{abs} \propto \left(\frac{1}{v}\right) \hspace{4cm} \text{Eq. 11}$$

$$\sigma_{sca} \propto constant \hspace{4cm} \text{Eq. 12}$$

in which v ($cm\ s^{-1}$) is the neutron speed.

For neutron imaging techniques (not covered in this paper), the transmission of the neutrons through the absorber is employed for obtaining information about the soil properties. However, different from the Am-Be neutron sources, employed in neutron gauges, neutron imaging techniques use neutrons emitted by Nuclear Research Reactors. The transmission of collimated neutron beams also obeys an exponential law (Equation 13):

$$\varphi = \varphi_0\ e^{-\Sigma_T(v)x} \hspace{4cm} \text{Eq. 13}$$

in which φ (number $s^{-1}\ cm^{-2}$) and φ_0 (number $s^{-1}\ cm^{-2}$) are the transmitted and incident neutron fluxes, x (cm) is the absorber length, and $\sum_T(v)$ (cm^{-1}) is the total cross-section for the neutron interaction in its path through the material, which represents the probability of the neutron suffers any type of interaction per unit length of material.

The fallout of the ^{137}Cs

Cesium-137 is a radioactive material that is not found naturally in the environment. However, some analyses show that ^{137}Cs is one of the radioactive elements present in the soil profile. Atmospheric nuclear weapon tests, made in the past (the 1950s and 1960s), and emissions from nuclear reactors spread out great amounts of ^{137}Cs to the high atmosphere, which was further deposited (the fallout of the ^{137}Cs) on the Earth's surface by atmospheric processes throughout the time. The peak redistribution of the ^{137}Cs fallout in the Southern Hemisphere happened in 1963. Thus, in Brazil, the erosion rates or sediment deposition, calculated by the ^{137}Cs fallout redistribution analysis technique, allow a study of these processes retrospectively to the last 58 years of land use.

The choice of appropriate areas as reference is one of the most important aspects related to the ^{137}Cs technique. Before applying the technique to analyze the redistribution of sediments across a study area, it is first necessary to evaluate the vertical distribution of the ^{137}Cs inventories. This has to be made in flat areas, not subject to erosion or sediment deposition processes, located close to the studied site, and remained unchanged since 1963. In these areas, the vertical distribution of ^{137}Cs activities must be concentrated in the first centimeters of the soil profile showing an exponential reduction with depth (Figure 2).

With the average value of the total inventory of ^{137}Cs at the reference site, the percentage of loss (negative value) or gain (positive value) of the ^{137}Cs inventory, for each measured point, is calculated according to equation 14:

$$\Delta A_{Cs} = \left(\frac{A_{Cs} - A_{Cs\text{-}ref}}{A_{Cs\text{-}ref}} \right) \times 100 \qquad \text{Eq. 14}$$

in which ΔA_{Cs} (%) is the percentage of losses or gains of the ^{137}Cs inventory at the sampling point, and A_{Cs} (Bq m^{-2}) and $A_{Cs\text{-}ref}$ (Bq m^{-2}) are the measured ^{137}Cs inventory at the sampling point and the average value of the total inventory of ^{137}Cs at the reference site, respectively.

The conversion of the ΔA_{Cs} to the erosion or deposition rates is based on the proportional model (Equation 15):

$$E_{asl} = \left(\frac{\Delta A_{Cs}\, \rho_b\, z}{TF} \right) 10 \qquad \text{Eq. 15}$$

in which E_{asl} (t ha^{-1} yr^{-1}) represents the mean annual soil loss, ρ_b (kg m^{-3}) is the soil bulk density, z (m) is the depth of the analyzed soil, and T (yr) and F are the time elapsed since initiation of ^{137}Cs accumulation and a particle size correction factor, respectively. The multiplier 10, present in equation 15, is a factor used for unit adjustments.

METHODS

Literature searching for this paper was made using the engines as: Scielo, ScienceDirect, Google Scholar, and Web of Science. The following search terms were utilized: "gamma-ray attenuation AND soil bulk density", "gamma-ray attenuation AND porosity", "gamma-ray attenuation AND soil structure", "gamma-ray attenuation AND hydraulic conductivity", "gamma-ray attenuation AND water retention curve", "gamma-ray attenuation AND representative measurements", "gamma-ray attenuation AND attenuation coefficient", "gamma-ray attenuation AND radiation interaction parameters", "neutron gauge AND calibration", "neutron gauge AND management systems", "neutron gauge AND hydraulic conductivity", "neutron gauge AND water retention curve", "neutron gauge AND water balance", "neutron gauge AND time series", "Cs-137 fallout AND soil superficial erosion", "soil spatial variability AND Cs-137 inventories", "Cs-137 inventories AND soil reference sites", and "Cs-137 inventory AND soil redistribution".
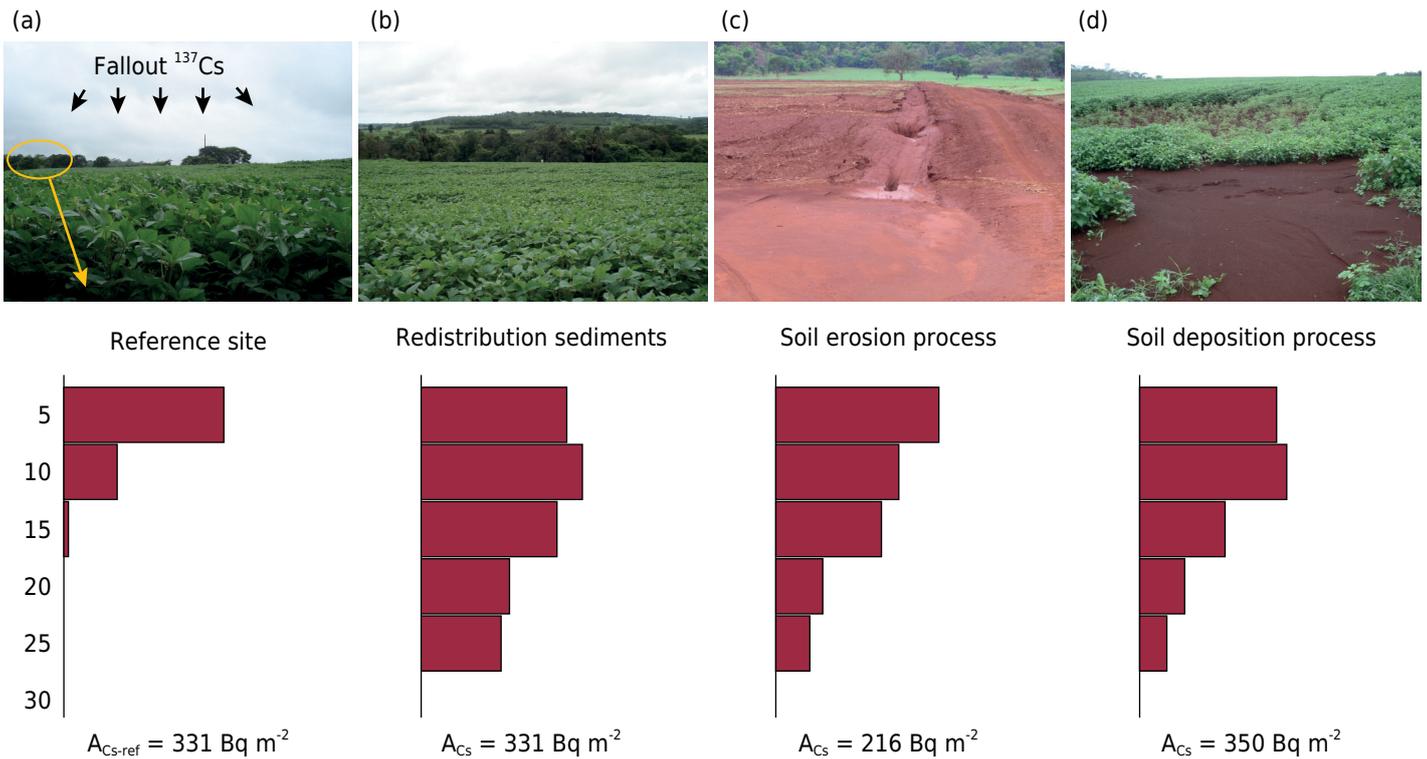
Figure 2. Distribution of ^{137}Cs activities in the soil profile and ^{137}Cs inventories from: samples collected in a reference area (A$_{Cs-ref}$) (a); in cultivated area with sediment redistribution process (b); in an area with loss process (c); and in an area with sediment deposition process (d).

This literature review does not cover all the papers published by the Brazilian scientists in the three main subjects selected for this study (gamma-ray attenuation, neutron gauge, and Cs-137 fallout). The papers were selected to give a general idea of the main applications of the three nuclear techniques in the area of soil science. Studies published in Portuguese and English were chosen. All the papers were peer-reviewed, and full texts are available through different databases. All the studies selected strictly comprise gamma-ray attenuation, neutron gauge, and the fallout of the Cs-137 in soil science. Around 100 selected pieces of research, dealing mainly with basic studies of soil attenuation properties and the application of the three nuclear techniques were included in this review.

OVERVIEW OF SOME APPLICATIONS

Gamma-ray attenuation method

In Brazil, the thesis of Professor Klaus Reichardt, presented in 1965 at the University of São Paulo (Reichardt, 1965), can be considered the pioneering study in the use of GAM for measurements of soil bulk density and water content (Table 1). Another important study was made by Professor Epaminondas Ferraz, in his associate professor thesis presented in 1974, measuring simultaneously soil bulk density and water content combining two radioactive sources (Ferraz, 1974).

Gamma-ray attenuation has been applied in both field and laboratory studies (Table 1) since the first experiments carried out in the 1950s. The choice of the radioactivity sources to be employed in GAM is mainly related to the detection systems usually utilized for the measurements. Due to their limited resolutions, solid scintillator detectors, such as NaI(Tl), require radioactive sources with primary energy peaks in regions with less or no interference of other radiations (Ferraz and Mansell, 1979; Pires et al.,

Table 1. Overview of some studies dealing with the application of the gamma-ray attenuation technique in soil science carried out by Brazilian scientists

Type of study	Objectives	Methods	Reference
Laboratory	Determination of the soil bulk density and soil water content for three contrasting soil types	Experimental measurement of the soil mass attenuation coefficient, soil bulk density, and soil water content using collimated radiation beams (radioactive source of ^{134}Cs)	Reichardt (1965)
Laboratory/Field	General review of gamma-ray attenuation methods for determining soil water content and bulk density	Experimental measurement of the soil bulk density and soil water content using single- and dual-energy setups (radioactive sources of ^{241}Am and ^{137}Cs)	Ferraz and Mansell (1979)
Laboratory	Proposal for a new method of soil mechanical analysis and continuous measurements of soil particle concentration	Experimental measurement of the soil particle size distribution and their fractions in soil suspensions under sedimentation using collimated radiation beams (radioactive source of ^{241}Am)	Vaz et al. (1992)
Laboratory/Simulation	Detailed analysis of the mass attenuation coefficient dependence with energy and soil composition	Experimental measurements of the mass attenuation coefficient using collimated radiation beams (radioactive sources of ^{241}Am, ^{152}Eu, ^{133}Ba, ^{137}Cs, ^{54}Mn, and ^{60}Co) and calculations made using the XCOM	Appoloni and Rios (1994)
Laboratory	Proposal for a new method of soil water retention curve evaluation using gamma-ray attenuation	Presentation of an experimental methodology for the use of the gamma-ray attenuation as an auxiliary technique for the evaluation of the water retention curve	Bacchi et al. (1998)
Laboratory	Measurement of soil pore size distributions and the proposal of calibration equations for monitoring changes in soil pore space	Experimental measurements of the total, intra-aggregate, and inter-aggregate porosities of dry soil clods using collimated radiation beams (radioactive source of ^{241}Am)	Oliveira et al. (1998)
Laboratory	Measurement of the soil water content and determination of the soil hydraulic conductivity	Experimental measurement of the soil water content, for different depths and times, in soil columns during water vertical infiltration experiments using collimated radiation beams (radioactive source of ^{241}Am)	Moreira et al. (2001)
Laboratory	Analysis of soil surface sealing induced by the application of sewage sludge at the topsoil	Experimental analysis of density increments of thin soil layers using gamma-ray computed tomography and gamma-ray attenuation (radioactive source of ^{137}Cs)	Pires et al. (2002)
Laboratory	Definition of optimal thicknesses for measurements of soil mass attenuation coefficients and their influence in determinations of soil physical properties	Experimental measurements of the mass attenuation coefficient of soil samples with different thicknesses using collimated radiation beams (radioactive sources of ^{241}Am and ^{137}Cs) and of the soil bulk density through gamma-ray computed tomography (radioactive source of ^{241}Am)	Borges et al. (2014a)
Laboratory/ Simulation	Evaluation of the influence of different methods to determine the mass attenuation coefficient	Experimental measurement of the mass attenuation coefficient using collimated radiation beams (radioactive sources of ^{241}Am) and computer-based-simulations (XCOM, GEANT4, Fluka, and MCNP)	Pires and Medhat (2016)
Laboratory/ Simulation	Analyses of radiation interaction parameters based on information of X-ray diffraction data and Rietveld Method	Experimental measurement of the soil elemental composition through X-ray diffraction and X-ray fluorescence and simulation of the mass attenuation coefficient through XCOM	Pires et al. (2019)

2003). The costs involved in the acquisition of the nuclear system, the half-life of the radioisotopes, and their activities are other aspects that need to be considered. Long half-life radioisotopes are preferred to avoid the need for corrections for decay during the experiments (Table 1). Total and specific activities are important due to the errors related to the random nature of radioactive disintegration (Ferraz and Mansell, 1979; Pires et al., 2002).

Before any application of GAM for measuring soil physical properties, it is important to obtain representative gamma-ray attenuation coefficients and analyze the influence of the soil chemical composition in their values. Cesareo et al. (1994) and Appoloni and Rios (1994) presented studies measuring the mass attenuation coefficient variation for different photon energies due to different soil chemical compositions. Through these studies, the authors were able to show the relationship between soil properties like bulk density, the atomic number of the components and electron densities, effective atomic number, and the attenuation coefficient. It was demonstrated that soils with different compositions attenuate the radiation differently depending on the photon energy (Figure 3). The detailed measurements of the mass attenuation coefficient presented in these two papers showed the importance of reliable measures of attenuation parameters to study soil-water interactions.

Measurements of soil physical properties using GAM can be influenced by an inadequate selection of the experimental setup geometry and the gamma-ray energy absorption buildup effect (Kurudirek et al., 2011). Frequently, for cases of "bad" geometries, it is necessary to correct the equations to better evaluate the mass attenuation coefficient. Kurudirek et al. (2011) found that the photon energy influences the buildup factor with maximum values in the incoherent scattering dominance and minimum values
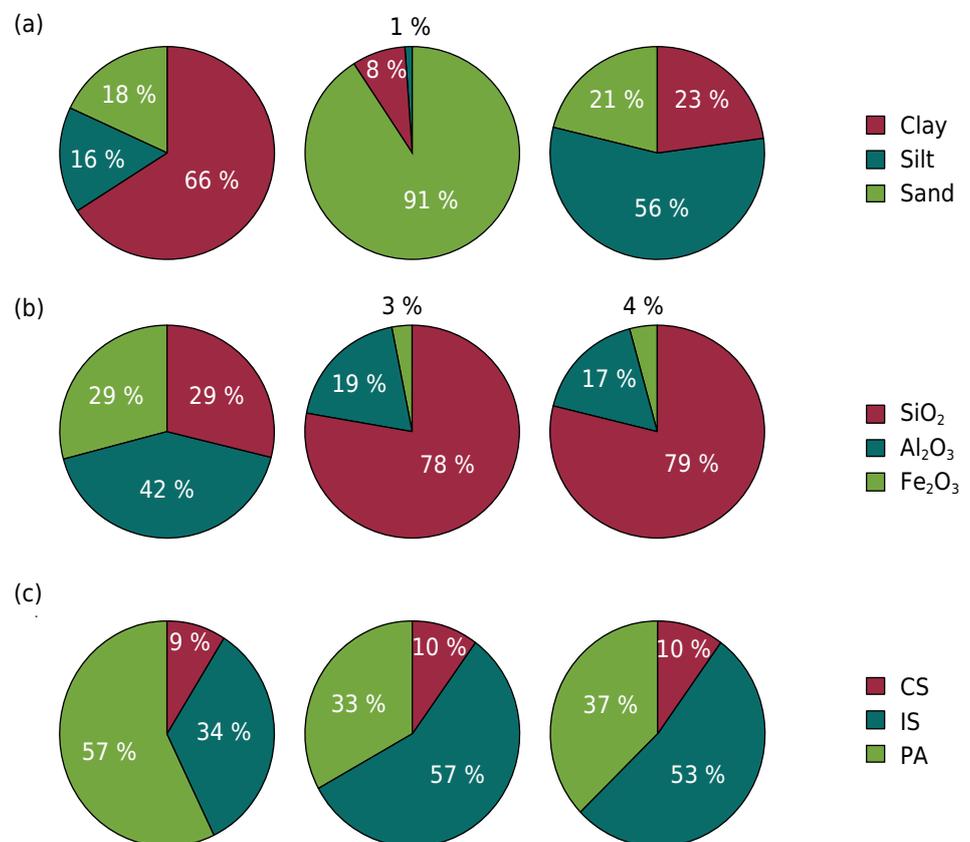

Figure 3. Different soil types based on their textural classes (clay, sand, silt loam) (a). Major oxide contents of the three contrasting soils (b). Contribution of the main radiation interaction processes (CS: coherent scattering; IS: incoherent scattering; PA: photoelectric absorption) as influenced by the differences in the soil texture and elemental compositions (c).

where photoelectric absorption (low photon energies) and pair production (high photon energies) are dominant. The buildup correction is needed to avoid the measurement of not consistent values of soil properties by GAM.

Medhat et al. (2014) analyzed several soil types to understand the role of their compositions in radiation attenuation. They observed close relations between the effective atomic number and the electron density. Theoretical estimates and experimental measurements of the mass attenuation coefficient exhibited a good agreement between them. Elements of high atomic number such as Fe_2O_3 and TiO_2 greatly influenced the attenuation of low photon energies (Camargo et al., 2021). Following the same idea, Medhat and Pires (2016) published another study with a detailed analysis of the photon atomic cross-section (σ_a) of soils of different types. The development of a tool to classify soils with distinct textures based on the photon atomic cross-section (Equation 16) was proposed (Figure 4). Preliminary results demonstrated that the photon atomic cross-section seems to be a potential parameter for studying soil texture based on the photon interaction.

$$\sigma_a = \sigma_m \left(\frac{1}{\sum_i n_i} \right)$$
Eq. 16

in which σ_m (cm^2 per molecule) is the molecular cross-section and n_i is the number of formula units in the molecule.

Soil minerals and amendments can influence the radiation interaction parameters leading to variabilities in measurements of soil physical parameters based on GAM. To evaluate the importance of soil mineralogy in the attenuation of radiation, Pires et al. (2016) analyzed five hard setting soils and found that not only the chemical composition influences the values of the mass attenuation coefficient but also the soil mineralogy. One of the results observed by the authors was that higher amounts of goethite increase the mass attenuation coefficient (Prandel et al., 2021).

Concerning the role of soil amendments, Ferreira et al. (2018) investigated the effects of surface liming on soil chemical attributes and radiation attenuation properties. Besides the influence of the soil amendments in the mass attenuation coefficient, the authors also demonstrated how changes in it would promote alterations in the soil bulk density and porosity. The results indicated that lime increased the radiation attenuation parameters at the topsoil, especially for photons of lower energies, with slight influences on the soil bulk density and porosity.

The optimum thickness (x_{op}) (Equation 17) of soil samples is also important for reliable measurements of soil physical properties by GAM. Factors such as the energy of the photon, the soil chemical composition, and the water content influence the optimum thickness (Ferraz and Mansell, 1979). The best measurements for photons of higher
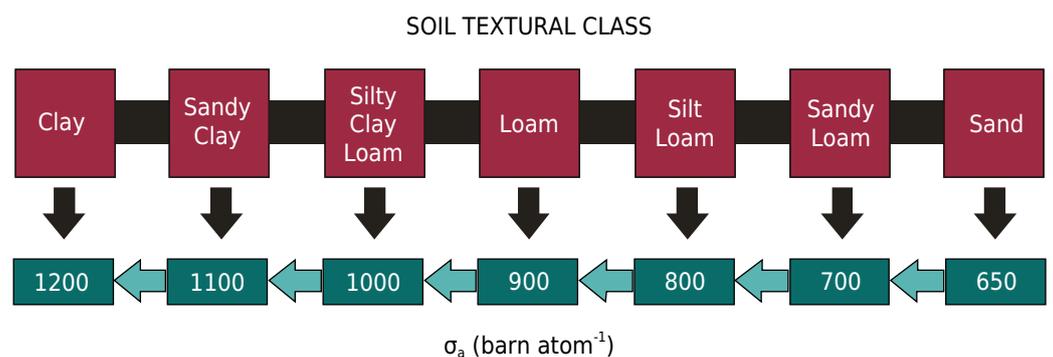
SOIL TEXTURAL CLASS

σ_a (barn atom^{-1})

Figure 4. Soil textural classes are classified according to the photon atomic cross-section (σ_a). . Adapted from Medhat and Pires (2016).

energies are expected for larger samples, while for photons of lower energies, smaller samples give the best results. Borges et al. (2014a) and Costa et al. (2014) presented two papers investigating the role of the sample size in measurements of the mass attenuation coefficient. In both papers, the authors reported the independence of the soil texture in the measures involving GAM. They also demonstrated that the best results for intermediate photon energies (e.g., ^{137}Cs) were obtained for samples larger than 0.10 m, while for small photon energies (e.g., ^{241}Am) samples smaller than 0.05 m provided the best results (Equation 17).

$$x_{op} = \left(\frac{2}{\mu_s \rho_b + \mu_w \rho_w \theta_r} \right)$$ Eq. 17

in which μ_s (cm^2 g^{-1}) and μ_w (cm^2 g^{-1}) are the soil and water mass attenuation coefficients, ρ_b (g cm^{-3}) and ρ_w (g cm^{-3}) are the soil bulk and water densities, and θ_r (cm^3 cm^{-3}) is the residual volumetric water content.

Besides the basic studies related to measurements of the radiation interaction with the soil, there are several applications of GAM in soil science. Important properties employed for evaluating soil quality have been measured throughout the last decades. Oliveira et al. (1998) presented a detailed analysis of total porosity (Equation 18), macroporosity (Equation 19), and microporosity (Equation 20) variation with soil depth determined by GAM. The technique allowed them to analyze changes in the intra-aggregate and inter-aggregate pores with millimetric resolutions, which cannot be accomplished by the traditional methods of investigation. Detailed studies of porosity were also presented by Appoloni and Pottker (2004), Pires et al. (2006), Pires and Pereira (2014), and Pires and Medhat (2016). All these papers give an idea of the potential of GAM for fast and non-destructive measurements of porosity as well as pore size distribution.

$$T_p = \frac{\kappa_p - \kappa_s}{\kappa_p}$$ Eq. 18

$$M_p = \frac{\kappa_{agg} - \kappa_s}{\kappa_p}$$ Eq. 19

$$m_p = \frac{\kappa_p - \kappa_{agg}}{\kappa_p}$$ Eq. 20

in which κ_p (cm^{-1}) is the linear attenuation coefficient for the soil particles, κ_s (cm^{-1}) is the linear attenuation coefficient for the whole soil, and κ_{agg} (cm^{-1}) is the linear attenuation coefficient for the intra-aggregate pore space. T_p (cm^3 cm^{-3}), M_p (cm^3 cm^{-3}), and m_p (cm^3 cm^{-3}) stand for total porosity, macroporosity, and microporosity determined by GAM.

Another important property utilized to evaluate soil quality is its bulk density (Equation 21). Pires et al. (2009a) made numerous measurements of the soil bulk density for different sampling depths and places at the field using different techniques and GAM. This study was also used for analyzing the spatial variability of the soil bulk density. The authors observed good agreement between GAM and the traditional method employing the volumetric ring; and that both methods demonstrated the same trend of spatial variability. In another study, Costa et al. (2014) investigated the influence of collimator sizes and sample thickness in measurements of the soil bulk density, demonstrating that both factors can affect these determinations. Recently, Pires et al. (2020) presented a methodology for measuring the soil bulk density of disturbed samples for educational purposes using GAM. The methodology described by the authors can be easily implemented in soil science courses at different levels of education.

$$\rho_b = \frac{-1}{x\mu_s} \left[ln \left(\frac{I}{I_0} \right) + x\mu_w \, \theta \rho_w \right]$$ Eq. 21

in which x (cm) is the sample thickness, μ_s (cm^2 g^{-1}) and μ_w (cm^2 g^{-1}) are the soil and water mass attenuation coefficients, respectively, I_0 (count rate) and I (count rate) the incident and transmitted gamma-ray photons, and ρ_w (g cm^{-3}) and θ (cm^3 cm^{-3}) are the water density and water content.

Soil mechanical analysis has also been made with the use of GAM (Figure 5). Vaz et al. (1992) presented a new method to evaluate the particle size fractions of soils with contrasting characteristics (Equation 22). The authors concluded that the new method does not affect the sedimentation process; it is fast, can measure all particle size fractions, and permits detailed analysis of the silt and sand fractions. Oliveira et al. (1994) utilized this nuclear method to evaluate the influence of soil management in the clay fraction. The method allowed a detailed analysis of the clay fraction distribution, which could not be accomplished by the traditional methods usually employed for mechanical analysis. Further improvements in the nuclear technique related to new methods, instrumentation etc., were later made by Elias et al. (1999), Vaz et al. (1999), and Naime et al. (2001).

$$C = \frac{ln\left(\dfrac{I_r}{I}\right)}{D[\mu_p - (\mu_w\rho_w/\rho_p)]}$$

Eq. 22

in which C (g L^{-1}) is the suspension concentration, I_r (count rate) represents the emerging beam transmitted by the dispersant solution (water and chemical dispersing agent), I (count rate) the emerging beam transmitted by the dispersant solution plus the dispersed soil particles, μ_p (cm^2 g^{-1}) and μ_w (cm^2 g^{-1}) are the soil particle and water mass attenuation coefficients, D (cm) is the container internal thickness, and ρ_p (g cm^{-3}) and ρ_w (g cm^{-3}) are the soil particle and water densities, respectively.

An interesting study for determining the hydraulic conductivity and diffusivity through columns of reconstructed soils was presented by Appoloni et al. (1990). Photons with an energy of 1.17 MeV (^{60}Co) were utilized to monitor the water content inside soil columns. Horizontal and vertical movements of the columns allowed detailed analysis of the water distribution with increments of 2 cm. The measurements made by these authors demonstrated the potential of GAM to study water dynamics in disturbed samples. Later in the 2000s, Moreira et al. (2001) estimated the hydraulic conductivity of undisturbed soil samples using a ^{241}Am radioactive source. Soil water content (Equation 23) was measured through GAM using the following equation (Santos et al., 1996):

$$\theta(z,t) = \frac{1}{x\rho_w\,\mu_w}\left(ln\,\frac{I_{0s}}{I_s} - x\mu_s\rho_b\right)$$

Eq. 23

in which μ_s (cm^2 g^{-1}) and μ_w (cm^2 g^{-1}) are the soil and water mass attenuation coefficients, x (cm), ρ_w (g cm^{-3}), and ρ_b (g cm^{-3}) are the soil thickness and the water and bulk densities, I_{0s} (count rate) and I_s (count rate) are the emergent radiation intensity from the empty soil column and the emergent radiation intensity from the filled soil column, respectively.

Gamma-ray attenuation method has also been utilized as a tool to evaluate the water retention curve. Bacchi et al. (1998) proposed the utilization of the gamma beam attenuation technique as an auxiliary tool to evaluate the soil water content retained at different matric potentials. Some years later, Pires et al. (2005a,b) implemented the nuclear method to measure the soil water retention curve at the matric potential interval of 0 to -0.1 MPa. In this study, an acrylic pressure chamber was specially designed for the measurements (Pires and Bacchi, 2006). Two contrasting soils were analyzed, and a ^{241}Am gamma-ray source was utilized in the measurements of the soil water content (Equation 23). As main conclusions, the authors verified a higher accuracy in the determination of the thermodynamic equilibrium, which reduced the total time required
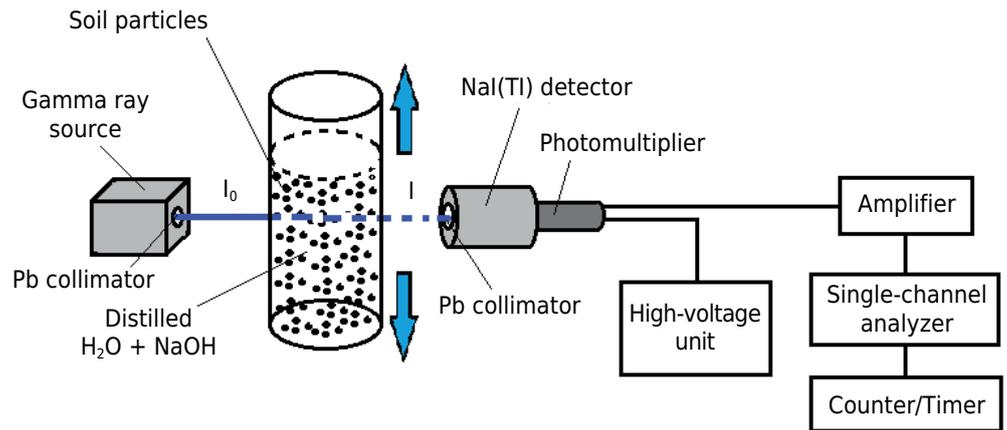
Figure 5. Schematic diagram of the gamma-ray attenuation system employed for soil mechanical analysis.

to measure the water retention curve, and continuous monitoring (each one minute) of the water content variation following pressure application in the soil samples, which cannot be accomplished by the traditional methods employed for similar measurements.

Many of the methods presented before are based on measurements of the soil physical properties in samples of regular shapes, e.g., cylinders, monoliths, etc. For these samples, it is relatively easy to measure the sample thickness using calipers. When the soil samples have an irregular shape, an alternative method of GAM can be utilized. This method is known as the two-media method, which requires combinations of measurements of the radiation transmitted through two different media with known attenuation coefficients. Cunha e Silva et al. (2000) presented a detailed description of the method and its validation for the measurement of the mass attenuation coefficient. Kuramoto and Appoloni (2002) made a study of the best combination of media to lead to the lower uncertainty in the measurements of the attenuation coefficient. They showed that the dependence of the results of the mass attenuation coefficient is closely related to the two media selected. Elias (2003) proposed the air as one of the two media. Through theoretical analysis, this author demonstrated that the equations utilized in the method could be simplified as well as the experimental uncertainties. It is important to highlight that the two-media alternative method can be an interesting tool for the determination of the mass attenuation coefficient when samples of regular shape cannot be sampled.

Soil neutron gauge technique and usage

Many studies in soil hydrology have as objective to better understand and describe hydrological processes inside the soil profile. In this regard, aspects as infiltration, redistribution, drainage, evaporation, and evapotranspiration, which are primarily related to these processes, are individually analyzed and subsequently combined (or integrated) for giving one condition to, most of the time, feed analytical or soil-related calculation models (Bacchi et al., 2002; Quesada et al., 2004; Antonino et al., 2005; Cruz et al., 2005; Silva et al., 2006) (Table 2).

Special attention is given to understanding or modeling these processes and characterizes water balance or storage changes at the field level, which requires a detailed characterization of the three main components that assemble the porous soil system: the solid, liquid, and gaseous phases (Bacchi et al., 2002). The water balance has main components that consider water inputs (rain, irrigation, and upward water fluxes from the water table) and outputs (evaporation, evapotranspiration, drainage for deeper zones, redistribution, and runoff) (Bacchi et al., 2002; Quesada et al., 2004; Antonino et al., 2005; Cruz et al., 2005; Silva et al., 2006; Quesada et al., 2008; Moreira et al., 2014).

The main components used for calculating the water balance are depicted in figure 6 and presented in equation 24 (Bacchi et al., 2002):

$$P + I + RO + Q_L - (E_V + T) = \Delta S_L \hspace{4cm} \text{Eq. 24}$$

in which P (mm h^{-1}) is the precipitation, I (mm h^{-1}) is the irrigation, RO (mm h^{-1}) is the runoff, Q_L (mm h^{-1}) is the water draining from the soil at depth L (cm), E_V (mm h^{-1}) is the evaporation, T (mm h^{-1}) is the transpiration, and ΔS_L (mm h^{-1}) is the change in soil-water storage in the investigated soil layer. It has to be mentioned that all the quantities presented in equation 24 are collected over a predetermined time interval selected by the gauge operator.

Neutron gauges are primarily used for water balance studies as the measurements they provide facilitate the calculation of soil water storage changes ΔS_L, for various depths in soil profiles at the field level (Table 2). The gauge measurements are correlated with the

Table 2. Overview of some studies dealing with the application of the neutron depth and surface gauge technique in soil science carried out by Brazilian scientists

| Type of study	Objectives	Methods	Reference
Field	Determination of the water soil content in the field	Experimental measurement of the soil water content at the field level using depth neutron gauges, tensiometers, and resistant blocks	Kirda and Reichardt (1992)
Field	Determination of the soil water content in the field, near the soil surface	Experimental measurement for evaluating the sphere of influence of the neutrons produced by depth neutron gauges near the surface	Falleiros et al. (1993)
Field	Calibration procedures for improving the determination of soil water content in the field	Experimental procedures for improving the calibration of depth neutron gauges for soil water content measurements	Reichardt et al. (1997)
Field	Determination of hydraulic conductivity in the field	Experimental procedures for determining the soil hydraulic conductivity using depth neutron gauge and tensiometers	Timm et al. (2000)
Laboratory/Field	Training course for gamma and neutron techniques usage and description of the methods	IAEA training course series: gamma and neutron techniques for determining soil physical properties and soil water content	Bacchi et al. (2002)
Laboratory/Field	Use of surface neutron gamma gauges for determining soil surface water content and bulk density	Experimental results for determining soil water contents at the soil surface and detection of denser layers at the soil profile	Tominaga et al. (2002)
Theoretical/ Field	Theoretical prevision of the calibration curve for a wide range of soil water contents	Theoretical calibration curve compared to the obtained in field conditions for the same soil	Crispino et al. (2004)
Laboratory/Field	Use of surface neutron gauges for determining soil bulk density	Experimental measurements of the soil bulk density using surface neutron gauges in the field and the comparison of the results with traditional non-radioactive methods	Timm et al. (2005)
Field	Use of surface neutron gauges for determining the water balance in coffee crops	Experimental results investigating the water balance component variabilities	Silva et al. (2006)
Field	Use of cosmic-ray neutrons for soil water content measurements at the landscape scale	Experiments using Cosmic-Ray Neutron Sensing for Hydrological Applications at the landscape level	Franz et al. (2020)
Field	Development of detection technology	Experimental developments of nuclear density gauges that use extremely low-activity radioisotope sources	Dep et al. (2021)
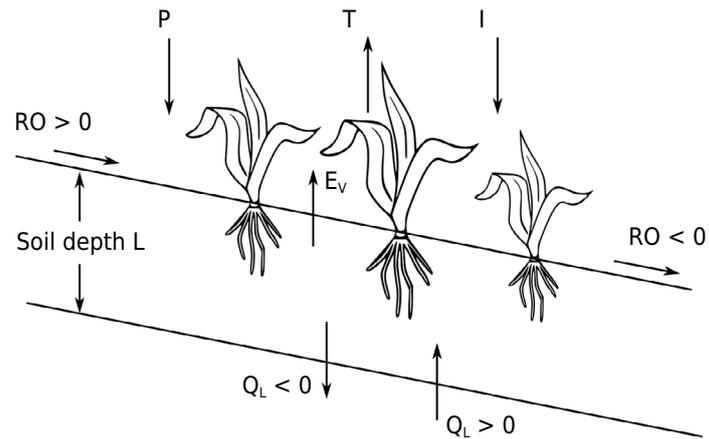
Figure 6. Main components of the water balance. Adapted from Bacchi et al. (2002).
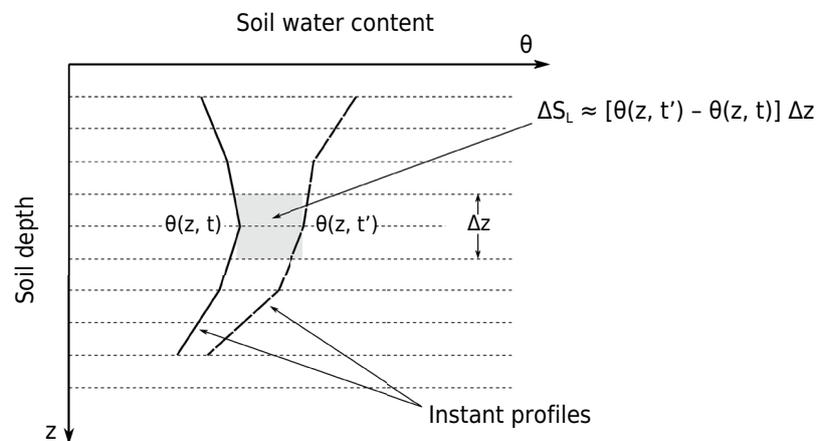

Figure 7. Instant profiles and water storage variation approximation in a depth z from the soil surface (gray rectangle).

soil water content in the soil profile. The evaporation and evapotranspiration in the soil profile can be calculated using equations related to the storage variation (Equation 24). The matric potential, usually recorded by using tensiometers and piezometers, permits calculating the water flux (Q_L) through the soil profile (Bacchi et al., 2002; Quesada et al., 2004; Antonino et al., 2005; Cruz et al., 2005; Silva et al., 2006; Souza et al., 2011; Borges et al., 2014b; Moreira et al., 2014). The method is considered non-destructive, and recurrent measurements may be made as necessary.

Soil water storage changes are defined as follows in equation 25:

$$\Delta S_L = \int_{z_2}^{z_1} \theta(z,t) \, dz \approx [\sum_z \theta(z,t') - \theta(z,t)]\Delta z \qquad \text{Eq. 25}$$

in which z_1 (cm) and z_2 (cm) define the soil layer analyzed (z_1-z_2), $\theta(z,t)$ (cm³ cm⁻³) is the soil water content at a particular depth z from the soil surface (z = 0), t (day) and t' (day) (t > t') are different time instants during the measurements, Δz (cm) is a soil layer around z, and ΔS_L (cm) is the water storage variation.

Based on the instant soil moisture profile defined by $\theta(z,t)$ (Figure 7), the method exhibits two consecutive profiles and the amounts used for calculating the storage variation. An approximation of the water storage variation (Equation 25) is obtained by summing, using numerical methods, the contributions $\Delta S_L = [\theta(z,t') - \theta(z,t)]\Delta z$ (gray rectangle in Figure 7) from the soil surface to any investigated chosen depth. The soil water content measurements are obtained using surface or depth neutron gauges.

The neutron source of the gauges (Figure 8) is lowered in the soil profile using aluminum access tubes. Fast neutrons, energies up to 14 MeV, produced in the neutron probe source (Equation 10 and Figure 8), interact with the soil particles and water through three interaction processes: neutron absorption by nuclei, neutron scattering through collisions, and neutron disintegration. The working principle of the neutron probe is primarily based on neutron scattering by elastic and non-elastic collisions. Through collisions, fast neutrons (energies higher than 2 MeV) lose energy (are moderated) and become slow or thermal neutrons (energies lower than 0.025 eV). Less energy is lost by fast neutrons when they interact with heavier target nuclei. Hydrogen is the target atom that most efficiently reduces neutron energy. Water, for its hydrogen content, is a good neutron moderator. Therefore, the presence of slow neutrons around the fast-neutron source of the probe is increased as the wetter is the soil. When the neutron source and detector are lowered into the access tube, a spherical "cloud" of slow neutrons develops quickly around them (sphere of influence in Figure 8). The number of slow neutrons per unit volume is proportional to the water content of the soil within the cloud. It is observed that the count rate (counts per minute – cpm or counts per second – cps) indicated by the instrument is proportional to the soil-water content. It permits the calibration of the neutron gauge using samples of known soil water content measured, for instance, using the gravimetric method (Bacchi et al., 2002). The procedure of the gauge calibration is schematically shown in figure 9.
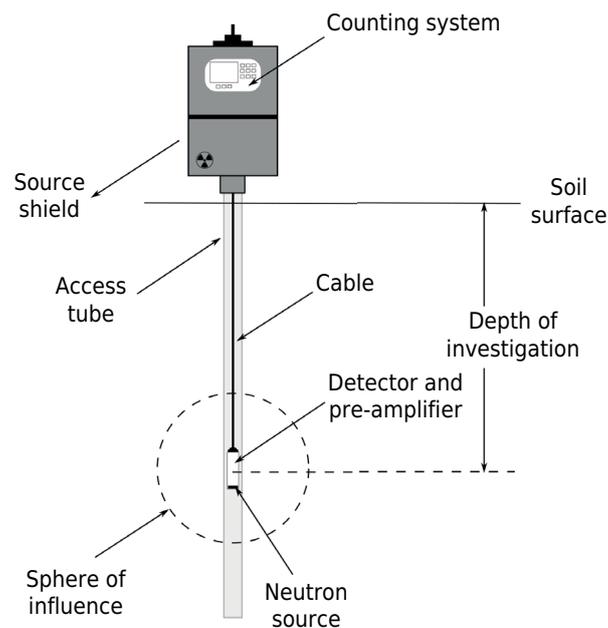

Figure 8. Schematic representation of the depth neutron gauge, the access tube, the sphere of influence, and the depth of investigation.
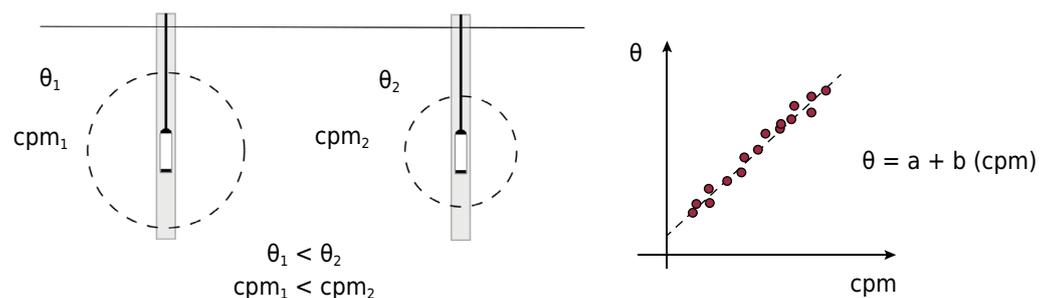

Figure 9. Schematic representation of the gauge calibration process. cpm: registered count per minute; θ: soil water content; a and b are the intercept and the slope of the linear regression model, respectively.

Laboratory calibration involves the use of packed soil samples with distinct and known levels of water content (Table 2). Usually, large amounts of soil are used in this procedure and the soil packing should be done carefully to ensure homogeneity of the soil water content and bulk density, which consists in a difficult and laborious task. Field calibration comprises the installation of access tubes, registration of the neutron counts with the neutron gauge, and prompt collection of soil samples around the access tube for measuring the water content by traditional methods. This procedure is repeated in different positions of the field to give one a set of pairs (θ, cpm) within a soil profile. In general, the soil water content and neutron counts in the detector are linearly related (Bacchi et al., 2002).

Soil water content and bulk density close to the surface of the soil profile are measured using a surface neutron-gamma gauge, which is schematically represented in figure 10. In this kind of gauges, the fast-neutron and gamma-ray sources, and the detectors (Geiger-Mueller and ^{3}He) are installed on fixed positions at the base of the gauge, which rests right at the top of the soil surface. The principle of obtaining soil water content using the fast neutrons produced at the surface gauge is the same as discussed early, the radius of the sphere of influence, besides being soil water content dependent, is approximately 15 to 20 cm. The gamma-ray source, located at the tip of a moveable stainless-steel rod, is introduced in the soil profile through a previously augered hole (direct geometry). In the direct geometry (left in Figure 10), the gamma-radiation that reaches the detector serves to determine the soil bulk density using the Beer-Lambert law of attenuation (Equation 21) (Bacchi et al., 2002).

The gamma-ray detector measures the photons that return after interacting with the soil components in the backscatter geometry. The intensity of the back-scattered photons is related to the density of the medium according to equation 26 (Bacchi et al., 2002; Timm et al., 2005):

$$\rho_b = B \, ln \left(\frac{A}{cpm - C} \right)$$ Eq. 26

in which A (count rate), B (g cm^{-3}), and C (count rate) are calibration parameters, which are determined experimentally using materials of known density; cpm is the backscattered counting rate per minute; and ρ_b (g cm^{-3}) is the soil bulk density.

Measuring the water distribution and consequently determining water storage in the soil profile is a task that relies on a consistent gauge calibration and similarity of the gauge response with any traditional method of determining the soil water content (Table 2). This calibration relates the observed counting to the water content at the time and position of the measurement. Several studies were conducted on this matter, and it is known
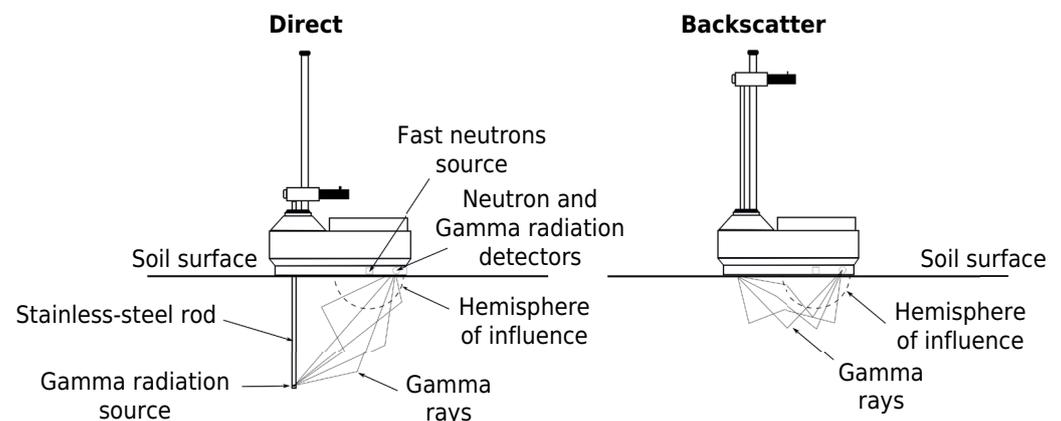

Figure 10. Surface neutron-gamma gauge operating in the direct geometry (left) and backscatter geometry (right).

that sometimes a particular or a dedicated calibration equation is needed (Bacchi et al., 2002; Antonino et al., 2005; Silva et al., 2006; Carvalho and Libardi, 2010; Borges et al., 2018). Also, studies designed for comparing neutron gauge measurements with traditional methods as, for instance, non-radioactive methods (soil-core sampling, tensiometers, piezometers, etc.) are presented in Kirda and Reichardt (1992), Souza et al. (2011), and Bacchi et al. (2002).

Studies using neutron gauges for the soil water balance, despite being, according to some authors, laborious and costly, are long-established (Falleiros et al., 1993; Bacchi et al., 2002; Quesada et al., 2004; Antonino et al., 2005; Cruz et al., 2005; Grego et al., 2006; Quesada et al., 2008; Souza et al., 2011; Borges et al., 2014b; Moreira et al., 2014). These studies are based on the water balance in the presence or absence of different crops in field conditions. Some of the studies indicate the need for dedicated calibration curves for determining the soil water content using the neutron gauges (Reichardt et al., 1997; Bacchi et al., 2002; Timm et al., 2005; Silva et al., 2006; Grego et al., 2006; Carvalho and Libardi, 2010; Souza et al., 2011). Also, it is mentioned that at the field level, the use of data obtained at meteorological stations distant from the experiment plot can produce an inadequate measurement of the rainfall (P) and consequently bias the water storage variation calculations (Silva et al., 2006).

Some studies investigate the influence of soil water content on neutron thermalization processes (Bacchi et al., 2002). It is, for instance, the case of the study by Crispino et al. (2004), where it was found that thermal neutron flux is linearly related to the water content in a homogeneous soil, for a wide range of soil water contents. Also, this study has proposed a method based on the soil chemical composition and soil bulk density for obtaining a theoretical calibration curve. Their results showed a good agreement between the theoretical and the experimental curves for a homogeneous soil at field conditions.

Other studies have as objective to delineate the needs of certain crops for water in different stages of plant development (Cruz et al., 2005; Silva et al., 2006; Souza et al., 2011). In certain cases of low water availability, irrigation may complement crop needs, and in cases of excess, drainage procedures may eliminate the surplus (Bacchi et al., 2002).

Depth neutron gauges are also used to outline studies for verifying how agricultural conservation practices can enhance the amount of water retained by the soil profile (Borges et al., 2014b). This study observed that plant management and mulching combination enhanced the soil moisture, increased the soil infiltration, provided better use of rainwater, and contributed to crop development by reducing water loss through runoff. Antonino et al. (2005), using depth neutron gauges, investigate the water balance in recession agriculture, consisting of cropping on slight slopes at the margins of small reservoirs, while the water level progressively decreases. The study was conducted in the semi-arid zone of the Brazilian Northeast region, where, due to scarcity of water, the knowledge of the water dynamics in this kind of management system is fundamental for establishing a basis for sustainable agriculture in this area.

In Falleiros et al. (1993), there is a study verifying the use of a neutron gauge for determining the soil water content close to the soil surface, as traditionally, these kinds of gauges are used for measurements at deeper layers. According to their results, special attention has to be given in this application, as for shallow layers the neutron sphere of influence (around 0.15 to 0.20 m in the soil) is not entirely encompassed by the soil, and the number of neutrons detected by the gauge detector can be completely biased due to neutron losses. To avoid this kind of problem, neutron surface gauges were designed for measuring soil water content close to the soil surface, as described before. In these gauges, neutron emitters and detectors are placed nearby at the bottom of the gauge, which rests just at the soil surface (Figure 10). Then, the sphere of influence is restricted to the topsoil layer, making the gauge measurement related to this portion of the soil profile (Tominaga et al., 2002).

Finally, there are also studies for determining the non-saturated hydraulic conductivity of the soil at field conditions (Bacchi et al., 2002; Timm et al., 2005; Carvalho and Libardi, 2010). In this kind of study, the internal soil drainage is registered using tensiometers and neutron gauge measurements. The hydraulic conductivity is determined assuming that water redistributes only in response to the gravitational field, and it is an exponential function of soil water content. It is called The Simplified Field Method, which is based on the instantaneous soil moisture profile (Libardi et al., 1980).

^{137}Cs fallout technique

The ^{137}Cs fallout technique has been applied in the last decades in different Brazilian regions (Table 3), with a total of 794 ^{137}Cs inventories identified in the papers analyzed. In these papers, around 88 % of the total inventories were selected in areas under agricultural, forest, or mineral exploration. The remaining 12 % are located in areas

Table 3. Overview of some studies dealing with the application of the ^{137}Cs fallout redistribution technique in soil science carried out by Brazilian scientists

Type of study	Objectives	Methods	Reference
Laboratory/Field	Temporal analysis of ^{137}Cs, ^{232}Th, ^{226}Ra, and ^{40}K activities from soils of the state of Paraná, Southern of Brazil	Analyses using gamma spectrometry	Schuch et al. (1994)
Laboratory/Field	Analysis of the spatial distribution of sediment redistribution rates in watersheds cultivated with pasture and sugarcane using the ^{137}Cs technique, USLE, and WEPP models	Analyses using gamma spectrometry, Universal Soil Losses Equation (USLE), and Water Erosion Prediction Project (WEPP)	Bacchi et al. (2003)
Laboratory/Field	Study of the organic nutrient pools declining in Semi-Arid tropical soils under subsistence farming, using the ^{137}Cs technique to estimate the soil erosion and land degradation effects	Analyses using gamma spectrometry	Fraga and Salcedo (2004)
Laboratory/Field	Comparison of soil erosion rates obtained by the ^{137}Cs technique through direct measurements in runoff plots	Analyses using gamma spectrometry and runoff plots	Correchel et al. (2005)
Laboratory/Field	Evaluation of random and systematic spatial variabilities of ^{137}Cs inventories at reference sites in South-Central region of Brazil	Analyses using gamma spectrometry	Correchel et al. (2006)
Laboratory/Field	The use of ^{137}Cs and soil carbon isotopic ratios (δ^{13}C) techniques to investigate the sediment deposition and erosion rates on a slope transect cultivated with sugarcane followed by a secondary riparian forest zone	Analyses using gamma spectrometry and soil carbon isotopic ratio (δ^{13}C)	Pires et al. (2009b)
Laboratory/Field	Study of the effectiveness of riparian zones on sediment retention in Brazilian sugarcane fields by the ^{137}Cs technique and WEPP model	Analyses using gamma spectrometry and Water Erosion Prediction Project (WEPP)	Bacchi et al. (2011)
Laboratory/Field	Analysis of the efficiency of riparian forests on the retention of sediments from annual crop lands using the ^{137}Cs technique	Analyses using gamma spectrometry	Santos and Sparovek (2011)
Laboratory/Field	Quantification of the impact of the no-tillage system on soil redistribution in a cultivated catchment from the Southern region of Brazil employing ^{137}Cs inventory measurements	Analyses using gamma spectrometry	Didoné et al. (2019)

under native forest or areas that did not experience any anthropic action. The studies were carried out in 14 Brazilian states, as shown in figure 11a.

Values for ^{137}Cs total inventories range from 0 Bq m^{-2}, found by Correchel et al. (2006) in some samples collected in runoff plots in Campinas (SP), to 1620 ± 463 Bq m^{-2} obtained by Handl et al. (2008) in Gramado (RS). These limits were established by analyzing soil profiles from latitudes varying from 00° 49' N (Handl et al., 2008) to 52° 12' S (Minella et al., 2014). The influence of the latitude on ^{137}Cs stocks has been pointed out as a relevant factor in the deposition of the fallout in soil profiles in Brazil (Schuch et al., 1994; Fraga and Salcedo, 2004; Handl et al., 2008; Andrello et al., 2009; Antunes et al., 2010; Montes et al., 2019).

It is known that the ^{137}Cs fallout deposition varies with latitude; nonetheless, variations in the soil ^{137}Cs stocks have also been observed within the same latitude, as shown through the analysis of coefficients of variation of inventories analyzed in reference areas (Correchel et al., 2005; Andrello et al., 2009; Antunes et al., 2010). Other factors, such as soil type, clay content, presence of Fe and Al oxides, organic materials, and climatic conditions, also result in ^{137}Cs inventory variability (Schuch et al., 1994; Handl et al., 2008; Didoné et al., 2019).

The low values of ^{137}Cs inventories, found in the first studies carried out in Brazil, raised some doubts in the possibility of application of the ^{137}Cs fallout technique in sediment redistribution studies for evaluating erosive processes in Brazilian agricultural areas. This fact motivated the study by Correchel et al. (2006), who found a good correlation (r = 0.76) between erosion rates, estimated by the ^{137}Cs technique and direct measurements in runoff plots, for a Typic Hapludox (Table 3).

The ^{137}Cs fallout technique has been used in recent decades mainly to estimate erosion and deposition rates caused by anthropic action at different scales. At the scale of experimental plots or transects, many studies have been made in areas cultivated with grain crops submitted to different systems of soil use and management (Fraga and Salcedo, 2004; Arthur et al., 2007; Santos and Sparovek, 2011), sugarcane (Sparovek et al., 2000;
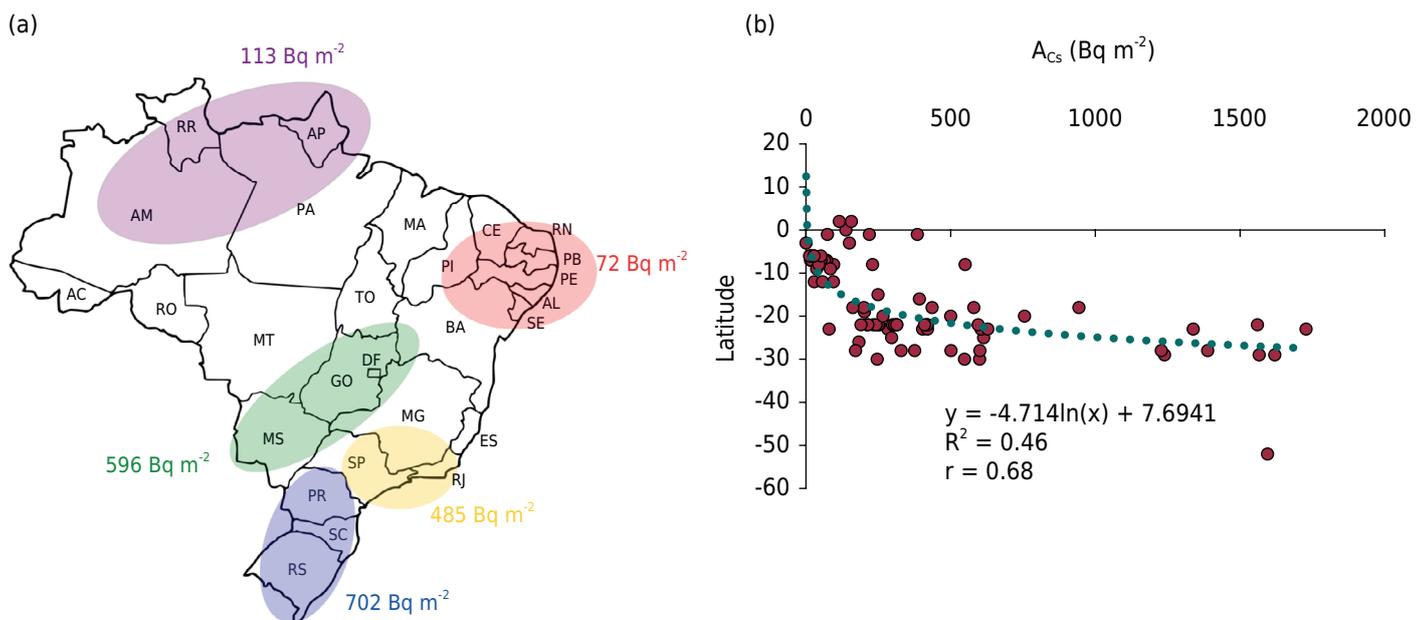

Figure 11. Average values of ^{137}Cs inventories of soils from the following Brazilian regions: North (purple), Northeast (red), Center-West (green), Southeast (yellow), and South (blue) (a) and ^{137}Cs inventory average values distribution for different latitudes (b) published in the papers of Schuch et al. (1994), Bacchi et al. (2000), Guimarães et al. (2003), Fraga and Salcedo (2004), Correchel et al. (2005), Arthur et al. (2007), Handl et al. (2008), Andrello et al. (2009), Pires et al. (2009b), Antunes et al. (2010), Santos and Sparovek (2011), Medeiros et al. (2014), Minella et al. (2014), Ribeiro et al. (2016), Didoné et al. (2019), Montes et al. (2019), and Ivanoff et al. (2020). N = 81.

Bacchi et al., 2003; Pires et al., 2009b; Bacchi et al., 2011), pasture (Bacchi et al., 2003; Fraga and Salcedo, 2004), forest (Andrello et al., 2009), and used for mineral extraction (Minella et al., 2014). In hydrographic micro basins, the ^{137}Cs fallout technique were used in studies carried out in Piracicaba (SP) by Bacchi et al. (2003) and Guimarães et al. (2003), in Aiuapaba (CE) by Medeiros et al. (2014), in the Mato Frio stream region (RS) by Minella et al. (2014), in Arvorezinha (RS) by Didoné et al. (2019), and in the Pampa region (RS) by Valente et al. (2020).

The ^{137}Cs fallout technique was also employed by Pires et al. (2009b), Bacchi et al. (2011), and Santos and Sparovek (2011) to assess the efficiency of the width of riparian forests to retain sediments produced by anthropic actions. Riparian forests are considered permanent preservation areas, characterized by the presence of native species with their width-restricted as a function of the river widths in the region (Brazilian Environmental Legislation, No. 4,771/65). The technique has also been applied in other types of permanent preservation areas, such as lakes, estuaries, and steep slopes aiming to analyze erosion and sedimentation processes (Saito et al., 2001; Figueira et al., 2006; Neves et al., 2014; Carvalho et al., 2016), to map the radioactivity distribution (Ribeiro et al., 2017), and to monitor the environmental pollution (Cunha et al., 1999).

Adequate results in sediment redistribution studies (Table 3), have turned the ^{137}Cs fallout technique effective for the validation of empirical and physical models such as the Universal Soil Losses Equation (USLE) and the Water Erosion Prediction Project (WEPP) (Sparovek et al., 2000; Bacchi et al., 2003, 2011), the Nutrient Monitoring Model (NUTMON), used to assess the balance of nutrients in the coffee crop (Araújo et al., 2009), and the Model of Water Availability in Semiarid Environments with Sediment Dynamics Component (WASA SED) (Medeiros et al., 2014).

The proportional model is the most used model for converting the percentage of ΔA_{Cs} (Equation 14) into sediment redistribution rates (Correchel et al., 2006; Pires et al., 2009b; Bacchi et al., 2011; Santos and Sparovek, 2011; Medeiros et al., 2014). Nevertheless, the mass balance model has also been used in Brazil for the same purpose (Minella et al., 2014; Didoné et al., 2019). In coastal environment sedimentation studies, the chemical-mathematical model of diffusion and convection of vertical flows (Equation 27) has been used to understand the temporal evolution of ^{137}Cs activities in sediment columns (Neves et al., 2014):

$$\frac{\partial A}{\partial t} = -\omega \, \frac{\partial A}{\partial z} + D \, \frac{\partial^2 A}{\partial z^2} - \lambda A \qquad \text{Eq. 27}$$

in which A (mBq cm^{-3}) is the activity per unit of volume, t (yr) is the time, ω (cm yr^{-1}) is the local sedimentation rate, z (cm) is the depth, D (cm^2 yr^{-1}) is the vertical diffusion coefficient of ^{137}Cs in the sediment, and λ (yr^{-1}) is the ^{137}Cs decay constant (2.30×10^{-2} yr^{-1}).

It is important to mention that the ^{137}Cs has also been utilized as a tracer of sediment movement when associated with other techniques, such as the soil carbon isotope ratio (δ^{13}C); adopted by Pires et al. (2009b) to evaluate the efficiency of the riparian forest width in sediment retention. Ivanoff et al. (2020) also employed the ^{137}Cs to validate sedimentation rates estimated by ^{210}Pb in the Lagoa dos Patos (RS).

FUTURE PERSPECTIVES

Gamma-ray attenuation method

As highlighted in this review, the gamma-ray attenuation technique has been utilized to study fundamental aspects concerning the radiation interaction with the soil and for determining soil physical properties of interest.

Understanding how radiation interacts with the soil is fundamental for making reliable measurements of soil physical properties using the method. Techniques of image analyses, such as computed tomography, are based on tomographic units derived from the interaction of the radiation with the soil. An exact comprehension of the processes responsible for the photon interaction is crucial for better discrimination of soil components, e.g., particles, minerals, organic matter, water content, etc. Thus, future studies should focus on soils with different mineralogical compositions, organic matter contents, and bulk and particle densities.

Many studies have demonstrated that photons of different energies are attenuated differently by the soil. Thus, more studies concerning the adequate photon energies to be employed in the investigation of soil physical properties should be considered. It is fundamental to produce images of good quality in computed tomography. This means that studies on the radiation interaction of photons with contrasting energies are of great interest to this field of knowledge. They can help to understand how photons are attenuated by soil and how to improve scan procedures.

The use of the radiation interaction parameters to classify the soil is another branch of study to be explored in the future. Few studies on this subject were published and understanding the role of different soil properties in the attenuation of the radiation is still necessary. Studies concerning selecting the best radiation interaction parameters to discriminate soils should be proposed to fill this gap. Consequently, to perform this type of investigation, choosing the best energy interval and the best nuclear parameters is necessary to characterize the soils.

The radiation interaction parameters can evaluate the effect of changes caused in the soil structure or soil chemical composition. Based on that, its use to detect the effect of management systems in the soil structure or changes in chemical composition can be an interesting matter of investigation. The utilization of the soil as shielding material is another possibility of great interest. Many geological repositories have been utilized around the world to store nuclear wastes. Analyzing the efficiency of these repositories is fundamental, and detailed studies of the interaction of the radiation with the soil used for this purpose are essentials.

There are many applications of the gamma-ray attenuation method in soil science. However, the technique continues attractive to evaluate mainly the soil water content and bulk density when high spatial resolution (millimeters) measurements are required. There is also the possibility of adopting this technique to evaluate swelling soils, with the advantage of being a non-destructive technique for simultaneously measuring the soil bulk density and water content, especially during dynamics processes. Many times it cannot be accomplished by using the methods traditionally employed in soil science.

Soil neutron gauge technique and usage

Neutron usage for soil water estimation is well established. Portable neutron gauges have been used for decades to establish water balance in different soils, crops, soil crop management, and irrigation procedures. From the material produced for this review, it is clear that improving the procedure for calibrating the gauges is essential, as the method is considerably dependent on it for gathering reliable information. Another aspect presented in just one of the investigated studies is the adequacy of the data provided by weather stations in calculating water balances. At the field level, biased results can arise from an inadequate measurement of the rainfall (P). It is caused by the use of data obtained at meteorological stations apart from the experimental plot. As dedicated weather stations are getting more and more accessible, it can be a solution for this kind of problem.

Besides being used for studies at field scale, sometimes, even at this level of examination, it can not reveal the intrinsic soil variability at upper levels of investigation. More recently

(ten years from now), some studies are using Cosmic Ray Neutron Sensor (CRNS) technology and monitoring networks for adequate characterization of landscape average soil water content (SWC) (Franz et al., 2020; Dimitrova-Petrova et al., 2020; Cooper et al., 2021). From this point of view, it can be considered a complementary technique or even represent an extension, to a broad scale of investigation, of the analysis obtained using neutron gauges.

Regulatory agencies worldwide consider nuclear density gauges as controlled devices as they employ radioisotope sources as emitter materials. The need for licensing, special storage, special transportation procedures, gauge operator training, and personal dosimetry, has represented a significant drawback for gauge users. An alternative to overcome these restrictions is to dedicate more attention to the improvement of detection technology. It permitted the development of nuclear density gauges that use extremely low-activity radioisotope sources. As a result, in this configuration, these devices were recently declared exempt from licensing and other nuclear regulatory requirements, for instance, in the United States (Dep et al., 2021).

Recent advances for characterizing the dynamics of water absorption in materials with high complex hydro-mechanical behavior have firstly combined neutron and X-ray tomography techniques. In this respect, multimodal pairwise datasets are registered into a common coordinate system, generating a 5D vector-valued field (position, neutron, and X-ray reconstructed values). In the approach, material information is derived from the X-ray data, while changes in water content are provided from the neutrons (Stavropoulou et al., 2020).

^{137}Cs fallout technique

The studies analyzed in this review show that one of the challenges of the future use of the ^{137}Cs technique is the continuous decrease of the ^{137}Cs activity (half-life of around 30 years). It makes its detection difficult in samples with low ^{137}Cs concentrations, as is usually the case for Brazilian soils. New advances in the instrumentation and detection systems (gamma spectrometers) could perhaps improve the conditions for the analysis of samples with low ^{137}Cs concentrations; however, it seems that the technique has the possibility of offering better results when associated with other methods such as, for example, the ^{210}Pb or the stable isotopic technique of δ^{13}C, as already mentioned.

The use of correction factors related to the enrichment of fine particles that arise during sediments' transport should be better investigated by soil science scientists in future studies. It should be made to improve the estimates obtained by conversion models of ^{137}Cs activities in rates of sediment redistribution.

The published papers do not provide detailed information about the quantity and type of samples collected, the depth and the procedures carried out for sampling; or any other basic information about the studied areas, such as geographic coordinates, altitude, annual average rainfall amount, soil type, particle size composition, carbon amount, etc. Considering that this information is crucial for soil scientists, especially those interested in conducting further studies on vertical migration and adsorption capacity of ^{137}Cs by Brazilian soils, it is strongly recommended to include this information in their future research.

This review also shows there is little or no information available in some Brazilian regions on the distribution of inventories of ^{137}Cs. Thus, a national database of ^{137}Cs inventories is necessary to complement the information already available. This makes it possible to identify priority regions for data collection, future research and financial support, and make these data accessible to researchers and public agents. This database can help monitor regions most susceptible to erosion processes due to deforestation, other anthropic actions, and sedimentation processes, which can cause the siltation or contamination of Brazilian water resources.

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AUTHOR CONTRIBUTIONS

Conceptualization: (iD) Luiz Fernando Pires (equal).

Methodology: (iD) Luiz Fernando Pires (equal), (iD) Fábio Augusto Meira Cássaro (equal) and (iD) Vladia Correchel (equal).

Formal analysis: (iD) Luiz Fernando Pires (equal) and (iD) Vladia Correchel (equal).

Writing – original draft: (iD) Luiz Fernando Pires (equal), (iD) Fábio Augusto Meira Cássaro (equal) and (iD) Vladia Correchel (equal).

Funding acquisition: (iD) Luiz Fernando Pires (lead).

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