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# **Colombian Neutron Activation Analysis Laboratory (CNAAL): Applications and Development Using the Nuclear Research Reactor IAN-R1**

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Guillermo Parrado, David Alonso, Julián Orozco,  
Mary Peña, Andrés Porras, Martha Guzmán,  
Nelson Acero and Mauricio López

Additional information is available at the end of the chapter

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## **Abstract**

This chapter describes recent advances of the Colombian Neutron Activation Analysis Laboratory (CNAAL) within the framework of expansion of geoscientific and nuclear knowledge in Colombia. Having the necessary historical references as a pillar of the current developments, the authors initially describe technical facilities of the laboratory and then articulate in an integral way of the value chain of this singular scientific and technological installation of Colombia. Its different stages beginning with the preparation of the samples, its irradiation process by different systems, recent novelty of the development of an “automated system of positioning of samples for gamma spectrometry,” analysis of gamma spectra to obtain concentration data of chemical elements, management of the radioactive waste generated, analytical quality control of the data obtained and finalizing in the use of this data to cover selected topics of knowledge in strategic sectors of our country’s development like a sustainable exploitation of mineral and hydrocarbons resources, researches in environmental and forensic sciences, technical developments in nuclear sciences, all aimed at improving the quality of life of Colombian citizens.

**Keywords:** neutron activation analysis, nuclear research reactor, applications, method validation, rare earth elements

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## 1. Introduction

The Colombian Neutron Activation Analysis Laboratory (CNAAL) is a facility used for qualitative and quantitative nondestructive chemical multielemental analysis by activating samples at the Nuclear Research Reactor IAN-R1 and analyzing their decay products using Gamma Spectrometry. Neutron activation analysis (NAA) in Colombia started at the Institute of Nuclear Affairs (IAN) when the nuclear reactor research IAN-R1 first achieved criticality in 1965. This technique has been used for over 30 years mainly for elemental analysis with applications in geology, hydrology, environmental and forensic sciences. In 1998, the Colombian government closed down the Reactor and the country's nuclear development fell behind other Latin American countries who also began their nuclear research applications in the 1960s.

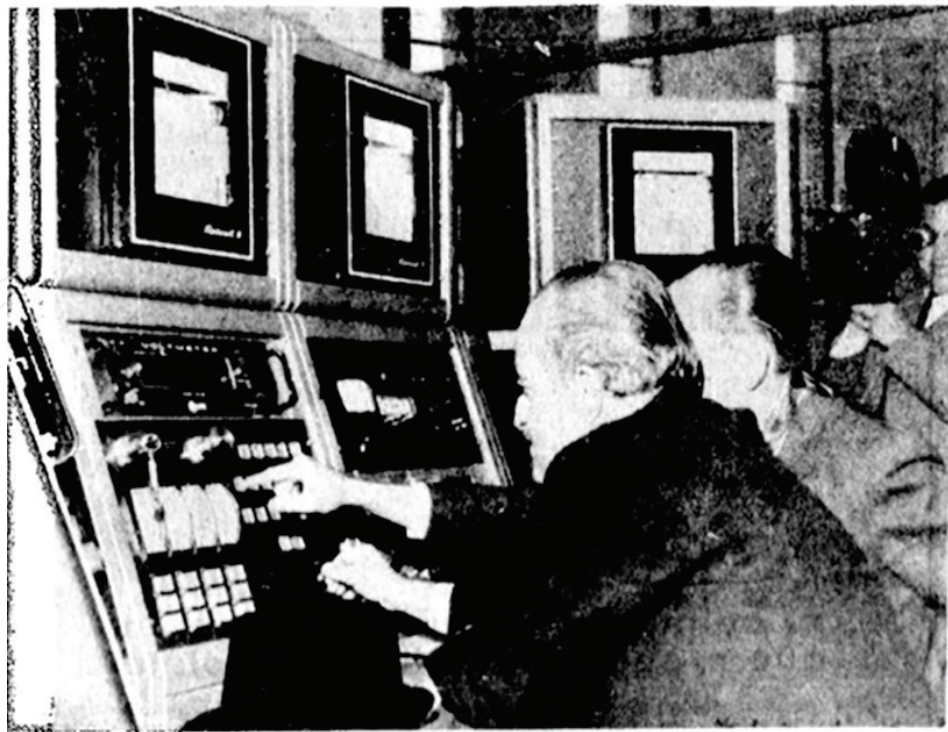
The onset of NAA in Colombia began in the mid-1950s, not long after the United States President Dwight D. Eisenhower gave his atoms for peace speech at the United Nations General Assembly on December 8, 1953. The Atoms for peace program served as shorthand for a number of programs intended to spread the peaceful uses of nuclear physics around the world and demonstrating its usefulness in the fields of medicine and energy generation. In 1955, the Colombian Institute of Nuclear Affairs (ICAN) was created, but it was not until 1957 that the Radioactive Analysis Laboratory was built as part of the ICAN chemistry program [1], being the predecessor of the current Neutron Activation Analysis Laboratory. This laboratory had basic instrumentation for personnel training purposes and was used for radiometric analysis by low-resolution gamma spectrometry using scintillation detectors like NaI(Tl) and single-channel systems.

Colombia was admitted to the International Atomic Energy Agency (IAEA) in the year 1960 and in 1964 began the construction of the dome building where the nuclear reactor stands today, later that same year the country received nuclear fuel manufactured in the United States as a donation by the American government. On January 20, 1965, the nuclear reactor achieved its first criticality (**Figure 1**) and operated at 10 kW during its first months [2].

In 1965, the first NAA samples were irradiated for elemental analysis [3], and the results were published 2 years later in 1967. Many scientific papers were published during this time, out of which two were of particular interest: the determination of inorganic iodine in samples of urine and the determination of trace amounts of selenium and tellurium in sulfur samples [4, 5]. The year 1971 witnessed the arrival of the first high-resolution Gamma Spectrometry System consisting of a germanium-lithium detector coupled to a multichannel analyzer (**Figure 2**). During this year, research focused on petrochemical analysis with the determination of vanadium in oil samples followed by the irradiation of food samples to determine nitrogen content in cereals.

Due to the boom in radioactive mineral exploration activities, the then Institute of Nuclear Affairs promoted a program for the quantitative evaluation of uranium and thorium in Colombia, for which Neutron Activation Analysis was used for the study of radioactive minerals and elemental determination in geological samples [1, 6]. Given the increased demand for this type of analysis and the interest from different companies on radioactive mineral exploration





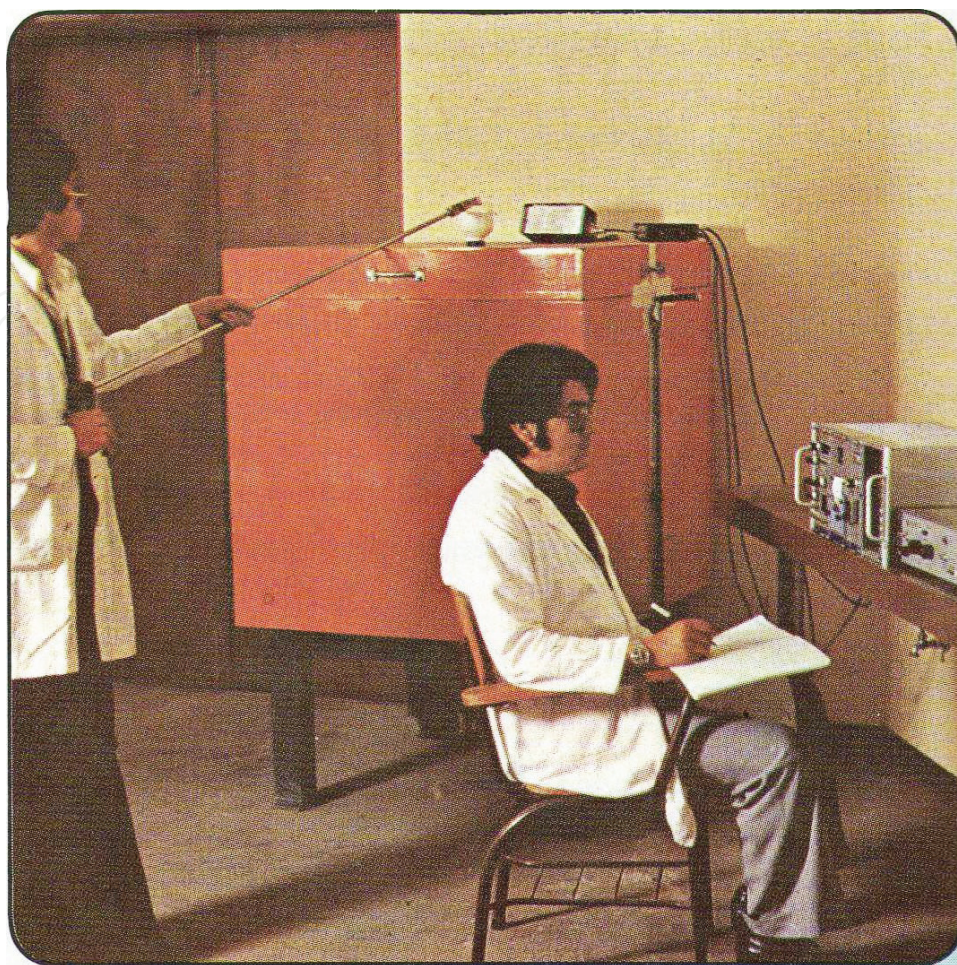
**Figure 1.** Nuclear reactor start-up by Colombian president Guillermo León Valencia (1965).

in the Colombian territory, a similar nuclear technique was implemented at the time which helped improve the precision of analysis: delayed neutron counting (DNC) (**Figure 3**). Delayed neutrons are emitted after nuclear fission events by one of the fission products some-time after the fission process [7].



**Figure 2.** Neutron activation analysis instrumentation, Ge-li detection system (1971).





**Figure 3.** U/Th analysis by DNC (1974).

As of 1973 and thanks to the support offered by the French Government, NAA was implemented as an alternative to conventional analytical methods for the determination of small quantities of precious metals such as gold, silver, platinum and palladium, among others [8]. For 12 years, from 1975 to 1986, the NAA technique reached its stage of the greatest development and was used as standard for elemental analysis. Thanks to the support given by the IAEA, the country received funds to upgrade its NAA laboratory due to the potential growth of users demanding this type of analysis (**Figure 4**). During this period, the use of NAA in Forensic Sciences was also introduced.

From 1986 to 1989, work focused on improving procedures and methodologies in the application of NAA for mineral resource exploration and studies of sediments and water pollution [9–11]. From 1987 to 1990, there was a drastic decrease in workload (**Figure 5**) due to an upgrade at the nuclear reactor.

Once the reactor was up and running, the laboratory continued its routine analysis, providing services to internal projects as well as to external clients. NAA was used mainly for mining companies and special forensic studies (**Figure 6**) [9]. In 1992, the laboratory was moved to a new space, which was built as an annex to the Reactor's Building with the sole purpose of installing a pneumatic transfer system that would allow for the measurement of short-lived radionuclides (with average half-lives of the order of minutes and seconds).



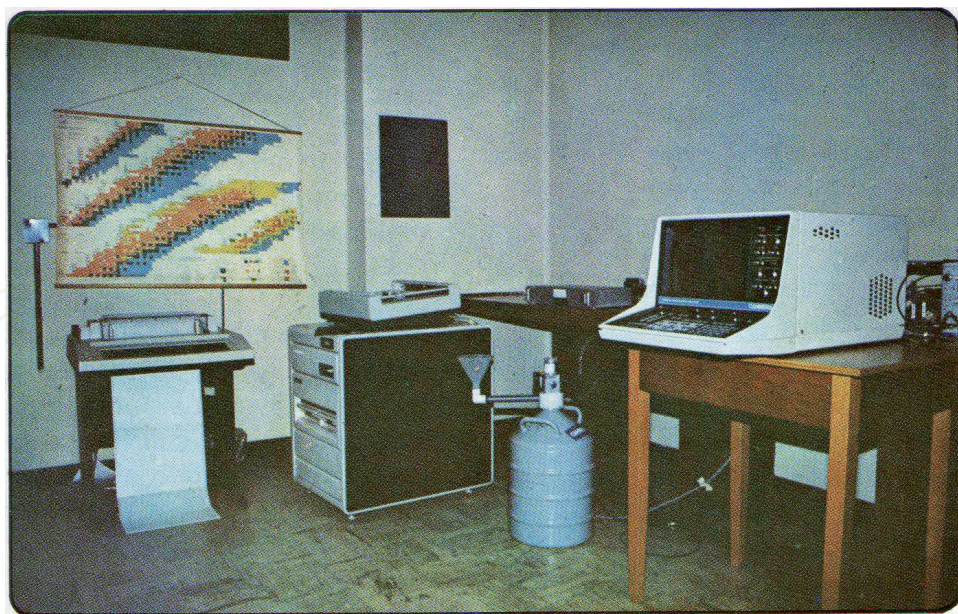


Figure 4. NAA instrumentation (1976).

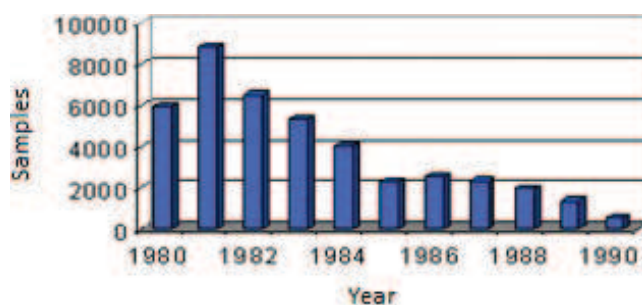


Figure 5. Irradiated samples from 1980 to 1990 [12].

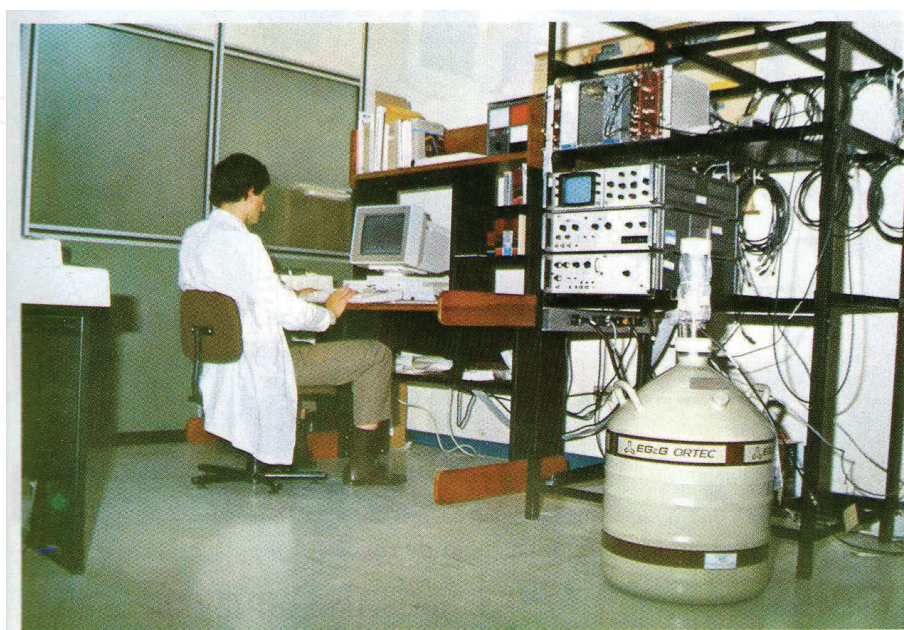


Figure 6. NAA laboratory (1989).



In September 1994, the nuclear reactor went into an extended shutdown period due to modernization of its instrumentation and control systems as well as conversion from highly-enriched uranium (HEU) fuel to low-enriched uranium (LEU) TRIGA fuel. Process took place from 1995 to 1997.

In spite of the liquidation in 1997 of the Institute of Nuclear Sciences and Alternative Energies (Formerly Institute of Nuclear Affairs), the nuclear reactor still operated with its new TRIGA fuel and was utilized for the analysis of forensic samples needed by the Police Department for the determination of barium, antimony and copper. During the first quarter of 1998, forensic samples were analyzed for the determination of mercury in hair, but due to the closure of the institute, the Reactor was shut down on March 31 that same year [3]. This series of events halted the operational experience that the NAA Laboratory had built for about 32 years (Figure 7).

It was not until 2005 that the Colombian Government decided to restart its Nuclear Program and began training personnel at the Reactor and associated nuclear laboratories. In May 2006 with support from the IAEA, the NAA technique was finally resumed, and tests were performed for the two Gamma Spectrometry Systems available at the time. One system equipped with a NaI(Tl) scintillation detector and the other with a Canberra 7229P HPGe semiconductor detector.

However, the NAA laboratory formally resumed activities in 2009, when the authorization for radioactive material handling was granted by the National Regulatory Authority.

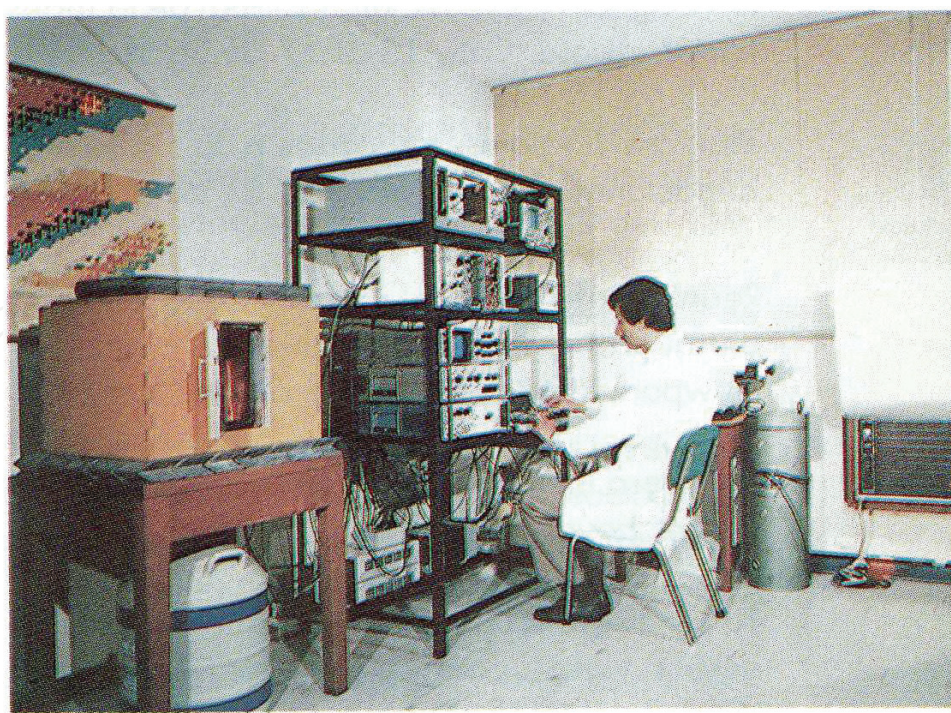


Figure 7. Gamma spectrometry system (1997).



Several expert missions were received for training of new personnel. The first objective of the Laboratory in this new stage was to provide the service of multielement analysis of geological samples, for which the relative method (comparator method in the literature) is used through certified reference materials (CRM) [13, 14]. **Figure 8** shows the state of the laboratory in 2009.

During the years of 2013 and 2015, the gamma spectrometry systems were modernized with High-Purity Germanium (HPGe) Canberra® detectors (**Figure 9**), an automated positioning system was also installed in the laboratory, making it unique in Latin America. IAEA experts have been continuously assisting on the validation for the test method and neutron flux characterization of the core. Today, thanks to the support given by the IAEA, the nuclear reactor and NAA laboratory have some of the most modern installations in Latin America with strong future prospectives in various fields of science.



**Figure 8.** NAA laboratory (2009).



**Figure 9.** NAA laboratory (2017).

## 2. Laboratory description

Neutron activation analysis is a multielemental chemical analytical technique based on neutrons generated by the nuclear reactor to create radioactive isotopes from stable isotopes in a sample material. The technique relies on excitation by neutrons so that the treated sample emits gamma-rays, and this radiation is then analyzed enabling the user to detect, identify and measure the presence of radioactivity in natural or man-made sources. The main use for the laboratory is to complement the conventional analytical techniques adopted in the institute, especially for those elements whose routine determination may require costly procedures with high environmental impact due to their nature and complexity. This type of analysis is used for the elaboration of a national geochemical map, which is essential for mineral exploration in the territory.

In 2009, not long after restarting the IAN-R1 Research Reactor, the laboratory was re-established at the Colombian Geological Survey, serving the country once again as a key player in the determination of elemental composition in geological matrices. Samples are irradiated under appropriate safety conditions following national regulations, which are lined up to the International Atomic Energy Agency (IAEA) and International Commission on Radiation Protection (ICRP) guides and scientific publications.

From 2016 to 2017, the delayed neutron counting technique was re-established for the determination of uranium and thorium in resource exploration projects due to the sensitivity of the technique, which is lower than  $1 \mu\text{g/kg}$  and can be used to analyze materials with high uranium content (including  $\text{U}_3\text{O}_8$ ) and enrichment of  $^{235}\text{U}$  [15].

The scientific staff are qualified and trained with several years of experience and extensive operational knowledge in the management of radioactive material, radiation protection, qualitative and quantitative chemical analysis, waste management, isotope applications and nuclear energy applications.

A brief description of the rooms that make up the CNAAL is given in the following sections.

### 2.1. Sample preparation room

This room has the necessary infrastructure and equipment for the preparation and adaptation of samples. A Niton XL3t GOLDD portable X-ray fluorescence analyzer, which is used for the preliminary characterization of the samples, a homogenizer mill, three analytical balances, a tablet press and a bag sealer. Reference materials and standards are properly stored under controlled conditions in this room.

### 2.2. Neutron activation analysis room

This room is where the samples are sent into and out of the core if the pneumatic transfer system is to be used. The systems console and Port No. 1 are located here. There is a gas and vapor extraction cabin with a 1.5 cm thick shielded port used to receive irradiated material from the reactor, and there is also a leaded glass which protects the staff from radiation exposure from the samples (**Figures 10 and 11**). Verification sources used for radiation monitors and calibration of gamma spectrometers are also stored in this room.



**Figure 10.** Gas extraction cabin.

### **2.3. Gamma spectrometry rooms 1 and 2**

The Gamma Spectrometry Systems used in the detection and quantification of the gamma-rays emitted by the activated samples after irradiation are located in two different rooms. A HPGe Canberra GC-1020 detector, a Canberra 2002CSL pre-amplifier and the InSpector 2000 multichannel module are located in the first room.



**Figure 11.** Shielded port for sample transfer.





**Figure 12.** Gamma spectrometry – Room 2.

The second room (**Figure 12**) has four HPGe detectors. Two Canberra GC-3018 detectors with 30% efficiency and energy resolution of 1.8 keV at 1.33 MeV at full-width at half-maximum (FWHM), and two GC-7020 units with 70% efficiency and energy resolution of 2.0 keV at 1.33 MeV (FWHM); each detector comes with its respective shielding, a LYNX® digital signal analyzer and is controlled by the Canberra's Genie2000 v.3.3 software. There is also an automated positioning system that uses a robotic arm to automatically place the samples in each of the four HPGe detectors and reads the gamma spectra during the time it is programmed; this system was designed for radiological protection purposes.

#### 2.4. Delayed neutron counting room

The room assigned for delayed neutron counting consists of a console and Port No. 2 of the pneumatic transfer system, one ton of paraffin shield which sits on top of 20 cm thick concrete blocks, a geometric arrangement of eight proportional  $\text{BF}_3$  counters and its associated electronic instrumentation for neutron counting and determination of uranium and thorium in geological, environmental and forensic matrices. There is a central hole in the paraffin shield where samples are placed for reading, and there is also a manual mechanism for sample extraction once readings are done (**Figure 13**).



**Figure 13.** Delayed neutron instrumentation.

## 2.5. Decay room

The radioactive material decay room is a space with lead and concrete shields needed to store activated samples for decay. It has two cylindrical lead containers with 6 cm thick walls for the storage of radioactive waste and two compartments made out of 15 cm thick concrete blocks for the storage of samples according to their half-life (**Figure 14**). Activated samples are temporarily stored in this room until the exemption levels are reached [16].

## 2.6. Pneumatic transfer system

The pneumatic transfer system allows for the rapid exchange of samples between the neutron activation room, the delayed neutron counting room and the nuclear reactor. Its master control is located in the Reactor's console room and without authorization from the reactor's personnel, it is not possible to send samples for irradiation, and this results in a redundancy in the safety of the sample positioning system.

This system consists of two compressors: flow diverters, two controls to send and receive samples (**Figure 15**) and a high-density polyethylene duct. The samples enter directly into one of the two aluminum terminals located in the core (positions D3 and C4), where the highest neutron flux can be found. It has the advantage to transfer activated samples to different areas speeds of 15 m/s, being an important aspect in the radiological protection of the personnel. This system is equipped with "air cushion" braking mechanisms to avoid violent crashes against the system ports.

The systems control unit includes a digital counter which can be set up for times between 30 s and 4 h. This unit controls the automatic valves to open and cut the air pressure at the right time. Since it is a complex pneumatic system with two receiving stations and two in-core terminal positions, there are diverters operated remotely from the control unit, which allows for the selection of irradiation positions and terminal stations where samples are received.

Due to the production of Ar41 during activation, terminal ports in the laboratory rooms are located inside extraction cabins with filters for radionuclides, preventing the inhalation of the radioactive gas by the operators.



**Figure 14.** Concrete shielding in decay room.



Figure 15. Pneumatic transfer system controls.

3. Sample preparation/irradiation

The Colombian Geological Survey serves the country by providing reliable scientific data through research into basic and applied subsoil geo-sciences; evaluating and monitoring threats of geological origin; exploring and monitoring petroleum resources, minerals and groundwater; the ability to study the elemental composition of samples such as rocks, soils, sediments, minerals, water and gases is a major asset. Collected samples are often taken to different laboratories for a variety of analyses if required by the research being conducted. Several analytical techniques ranging from Gravimetric Analysis and Atomic Absorption Spectrometry, to X-ray Fluorescence, Inductively Coupled Plasma Mass Spectrometry (ICP-MS), and Neutron Activation Analysis among others are available at the Colombian Geological Survey.

Geological materials analyzed by NAA need to be previously dried at room temperature, crushed, pulverized and sieved to a particle size of 150 μm (100 mesh, **Figure 16**). Once the sample is received (~ 50 g), moisture content needs to be determined in order to make future corrections referencing the dried sample. Samples are homogenized, weighed ( $0.250 \pm 0.001$  g), pressed and encapsulated in plastic hermetically sealed polyethylene vials.



Figure 16. Sample preparation process: (a) drying (b) grinding and (c) screening.



Samples for long-lived element activation (days to years) are placed at the periphery of the core and vials with samples are arranged in racks as shown in the following diagram (Figure 17). These racks are placed in vacuum-sealed Ziploc bags before irradiation in the G3-G4 positions (Figure 18).

The following elements can be determined after a 4-h irradiation operating at 30 kW: Sm, Lu, U, La, Nd, Eu, Hf, Ce, Yb, As, Sb, Ba, Br, Cd, Gd, Ga, Ho, Mo, W, Th, Cr, Cs, Sc, Ir, Ni, Se, Ag, Ta, Tb, Tm, Rb, Fe, Co, Zn, Zr. The neutron flux is measured by 5 mg Al + 0.1% Au rectangular foils as previously shown in Figure 17. Measurement required to obtain the correction factor  $f\phi$ . Samples for short-lived element activation (seconds to a few hours) are irradiated inside the core at positions D3 or C4 (Figure 18). These samples are encapsulated in cylindrical pressure-sealed polyethylene containers, packed in pairs into rabbits (polyethylene vials) and transferred into the core by the pneumatic transfer system.

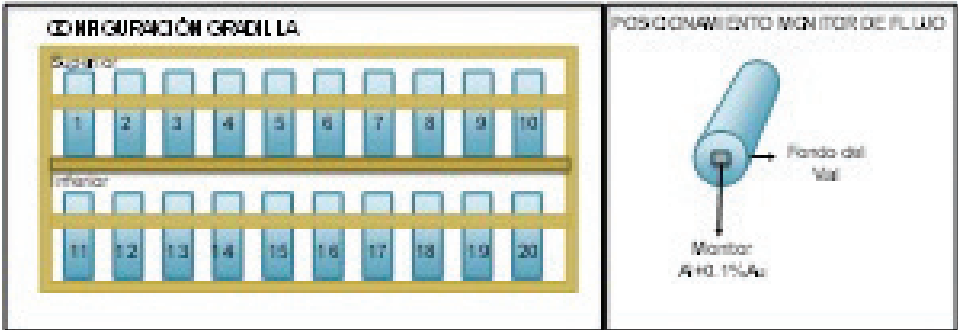


Figure 17. Rack sample configuration (left) neutron flux monitors attached to vials (right).

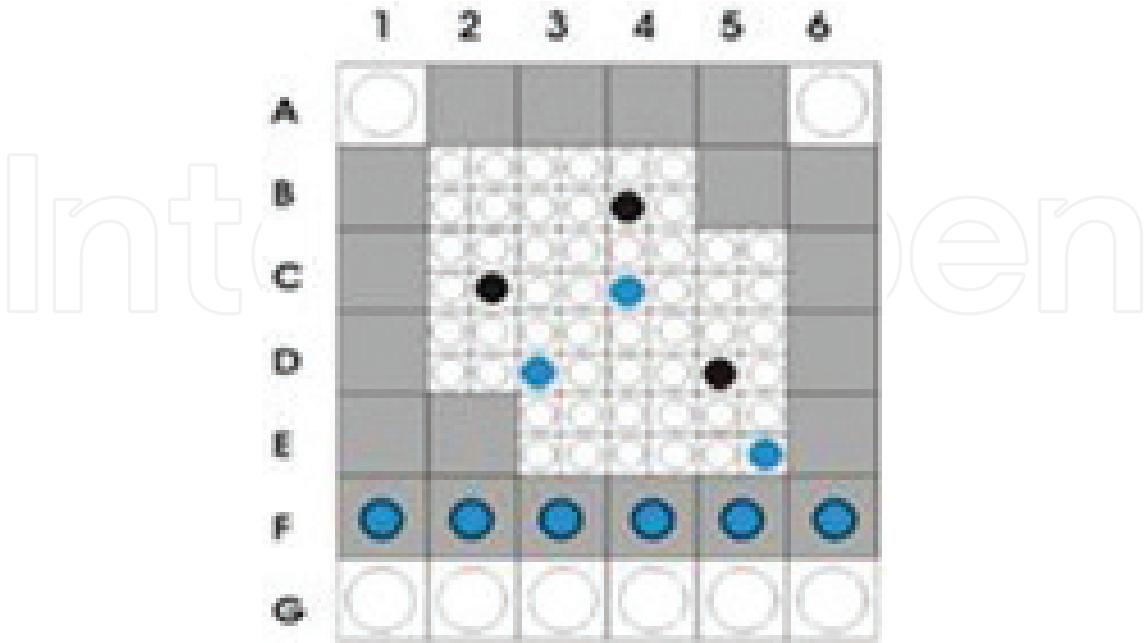


Figure 18. Reactor core schematic (IAN-R1).

Sample irradiation in these rabbits ranges from 40 s at 5 kW for the analysis of uranium and thorium using delayed neutron counting techniques and up to 5 min at 30 kW for the analysis of short-lived elements (Al, Ca, Mg, Ti, V, Dy, Mn, K and Na).

The IAN-R1 nuclear reactor was built by the Lockheed Western Export Company and was commissioned in 1965 as a graphite-reflected pool-type research reactor, cooled by natural convection with light water. The current core consists of 50 fuel elements made of  $\text{U-ZrH}_{1.6}$  enriched up to 19.75%. The reactor is licensed by the Ministry of Mines and Energy (Nuclear Regulatory Body) to operate at the maximum steady-state power of 30 kW, and it is located inside a cylindrical tank made of carbon steel  $6 \times 10^{-3}$  m thick, 5.25 m tall and 2 m in diameter with capacity to store up to 16 m<sup>3</sup> of water.

The nuclear reactor’s instrumentation and control systems were fully upgraded during 2012 and 2013 (**Figure 19**) by National Institute of Nuclear Research (ININ, México), in 2016 a new automated pneumatic transfer system (**Figure 20**) was installed, replacing the original system installed in 1997. This system opened up two irradiation positions inside the core, remotely



Figure 19. Nuclear research reactor IAN-R1.

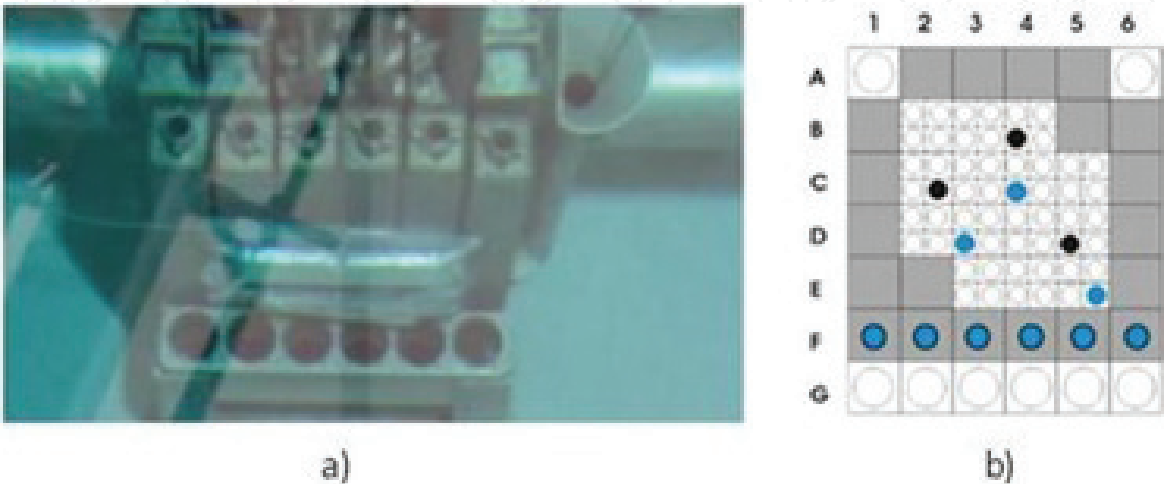


Figure 20. Rack with samples for irradiation.

controlled from ports 1 and 2 located in the neutron activation and delayed neutron counting rooms, respectively.

Additionally, there is a peripheral pneumatic transfer system that was part of the original design (1965) and is used to irradiate samples in a position adjacent to the core (Position A6). This old system was used from 1968 to 1992 for radioisotope production ( $^{24}\text{Na}$ ,  $^{32}\text{P}$ ,  $^{82}\text{Br}$ ,  $^{198}\text{Au}$  and  $^{99}\text{Mo}$ ) [17].

The rack containing the flux monitors and samples is positioned in the middle of the frontal face of the core, irradiated during 4 h at 30 kW (**Figure 20**) and subjected to a thermal neutron flux of around  $2.3 \times 10^{11}$  neutrons  $\text{cm}^{-2} \text{s}^{-1}$ .

**Table 1** presents a summary of the experimental conditions used for the analysis of short-, medium- and long-lived elements.

Characteristic	Value/description	
	Pneumatic transfer system	Rack system
Method	Direct comparator	Direct comparator
Measurand	Mass fraction	Mass fraction
Sample/comparator (mass)	$0.250 \pm 0.001 \text{ g}$	$0.200 \pm 0.001 \text{ g}$
Reactor power	20–30 kW	30 kW
Irradiation position	D3 or C4	G3–G4
Irradiation time	1–5 min	4 h
Flux monitor	Al + 0.1% Au 0.5 mg	Al + 0.1% Au 0.5 mg
Decay time 1	5 min	3–7 days
Reading time 1	5 min	3 h
Reading 1 geometry	50 mm	30 mm
Decay time 2	60 min	21–28 days
Reading time 2	10 min	4 h
Reading 2 geometry	10 mm	15 mm
Photon energy (keV)	Nuclide dependent	Nuclide dependent

**Table 1.** Experimental irradiation conditions.

#### 4. Automated sample positioning system for gamma spectrometry

Automation allows greater control of counting geometries, less error in positioning, increased productivity in the analyses, increased control and quality assurance of the analytical data and decreased doses received by the staff [18].

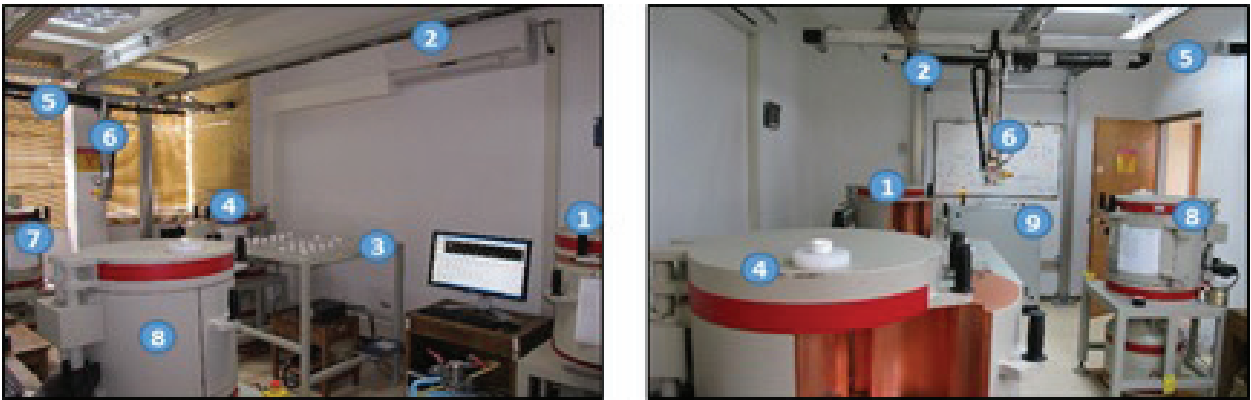


The automated system developed in the NAA laboratory fulfills the following objectives: Programming of analysis sequences, opening and closing of shields for sample positioning in the detector, and communication with the software for data acquisition. This has improved productivity by enabling 24/7 operation, and as a side benefit there is also less exposure to ionizing radiation.

This system is based on electromechanical components that can handle up to 64 sample readings in the same sequence of analysis, for which each step includes the collection of the sample from a sample rack, the positioning of the sample in the detector, data acquisition at pre-defined reading times, and the return of the sample to the rack where the other samples are located.

All of this is possible by means of a high precision positioning system based on linear actuators. The system is controlled by a human machine interface (HMI) where execution commands can be programmed. **Figures 21–24** show how everything is set up.

In principle, the presence of personnel in the gamma spectrometry room is limited while the positioning system is in operation. However, shielding for the sample rack is to be installed in the near future for radiological protection purposes.



**Figure 21.** Positioning system: 1. Detector A; 2. X-axis linear actuator; 3. Sample rack (64 positions); 4. Detector B; 5. Y-axis linear actuator; 6. Z-axis linear actuator; 7. Detector C; 8. Detector D; 9. Control panel.



**Figure 22.** Positioning system: 1. HMI; 2. Emergency stop; 3. System status; 4. Electronic components; 5. Power control.



**Figure 23.** Positioning system: 1. Electric motor to open/close shielding; 2. Shield sensor for opening; 3. Shield sensor for closing; 4. Emergency stop; 5. Sample gripper; 6. Detector sample support; 7. Actuator motion limit sensor.



**Figure 24.** Positioning system.

Precision is determined by servomotors that provide the movement, which have a resolution of 1,048,576 pulses/revolution per axis. Coupled to the previously described servomotors, there are linear belt actuators (Accuracy  $\pm 1.0$  mm) with their respective guides for alignment and friction reduction throughout the working area.

The system has two spatial adjustment options, point-to-point which displays the 64 positions of the sample rack and the 4 detector positions; and also single-point which is used to correct a common mismatch in all points using the point-to-point option, defining the first position of the rack, and allowing for the automatic adjustment of all the other positions. Both of these options are password protected for security reasons.



The point-to-point adjustment option must be used to change the counting geometry on the detectors, which must be performed prior to the execution of the sequences as required by the operators.

This positioning system greatly reduces manual efforts during the analysis of radioactive samples; the only manipulation required by our staff is the setup of the 64 samples in the rack. The idea of the use of this rack is minimizing Radiation Exposure, and thus enhancing the safety and well-being of personnel.

## 5. Data acquisition

In order to offer a quality service, the laboratory has made important updates in its instrumentation; acquiring four state-of-the-art HPGe Canberra Detectors for the measurement of gamma radiation and a novel sample positioning system.

The NAA relative method uses gamma radiation emitted by the radioactive nuclei from activated samples and compares it to the radiation emitted by a reference material with similar characteristics.

The characteristic gamma energies of each radionuclide are measured using one of the four solid-state semiconductor detectors (GC-3018 and GC-7020) coupled to LYNX® digital signal analysers controlled by the Canberra's Genie 2000 (v3.3) software. Each of these systems is calibrated weekly in energy, and its efficiency is checked monthly using a gamma check source kit consisting of  $^{241}\text{Am}$ ,  $^{22}\text{Na}$ ,  $^{133}\text{Ba}$ ,  $^{137}\text{Cs}$ ,  $^{155}\text{Eu}$  and  $^{60}\text{Co}$  electro-deposited point sources.

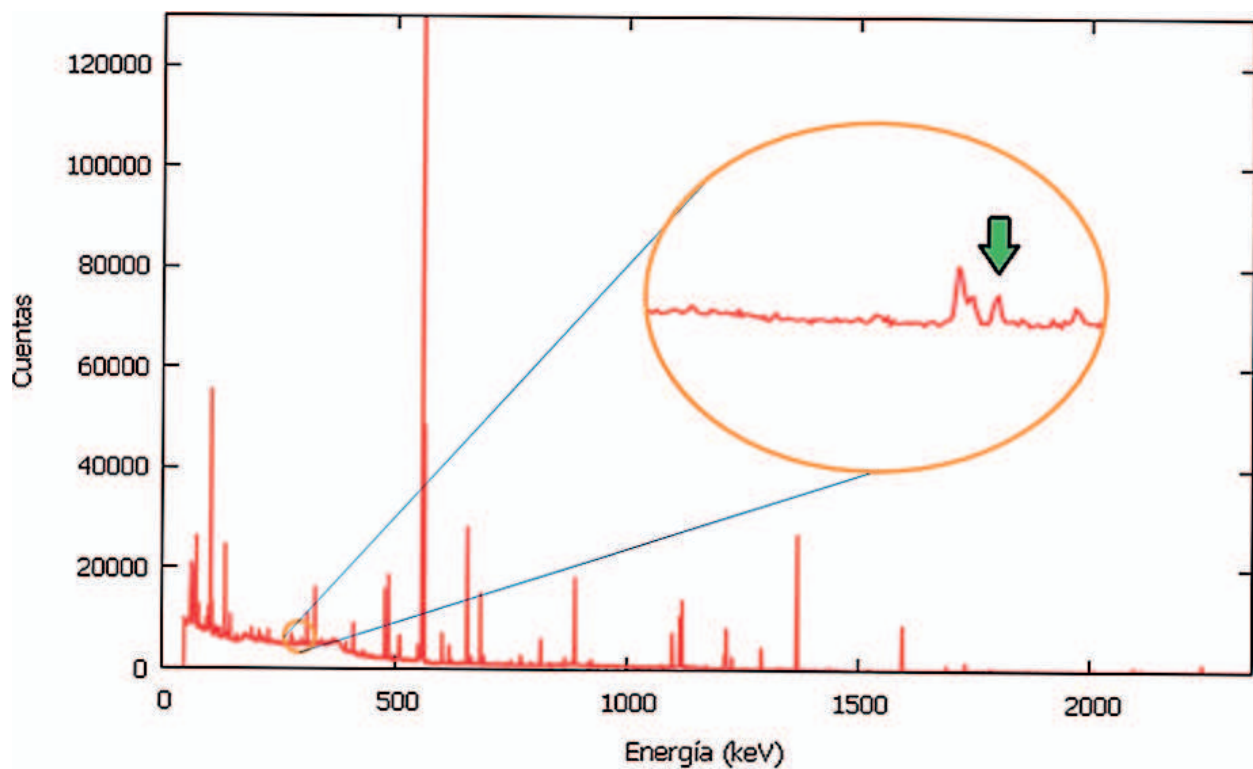
Depending on the irradiation, system used (**Table 1**), and once pre-determined decay times are reached, radiation measurement is performed by using one of the HPGe detectors [19].

Radiation measurement by samples coming from the pneumatic system is performed in some of the less efficient detectors (GC-1020 or GC-3018, depending upon availability). For complete analysis, two readings are carried out: the first, after 5 min of decay, positioning the vials individually at a distance of 50 mm from the detector and reading the corresponding spectra for 5 min. The second reading is done after the activity decays for an hour, at a distance of 10 mm and the spectra is read for 10 min. On the other hand, samples not going through the pneumatic transfer system are analyzed as follows: a first reading is done after a 4-day decay on one of the GC-3018 detectors, positioning the vials individually at a distance of 30 mm from the detector and reading the spectra for 3 h. A second reading is then performed after 21 days of decay on the higher efficiency GC-7020 detectors at a distance of 15 mm and the spectra is read for 4 h.

Flux monitors used during sample activation for flux corrections are also analyzed by gamma spectrometry on one of the GC-3018 detectors at a distance of 50 mm for 60 min. The samples obtained its corresponding gamma spectrum (**Figures 25 and 26**).



**Figure 25.** Detection geometry for check source verification.



**Figure 26.** Gamma spectra from soil sample irradiated for 4 h at 30 kW and a 5-day decay.

The elemental determination in the NAA is done by using the relative calibration (direct comparator method) [20]. This method uses a sample with known mass of the elements of interest (comparator or standard) and an unknown sample which is irradiated simultaneously. Taking into account that the amount of radiation emitted from the activation of the sample is proportional to the neutron flux, and this in turn to the mass of the irradiated element, it is found that the ratio between the mass fraction of the element  $x$  and the amounts of influence is given by:



The subscripts indicate parameters for the unknown sample and the comparator or standard. That is, the mass fraction of the unknown sample of the element (measurand) and the mass fraction of the element in the reference material.  $W$  is the total mass of the samples. The number of net counts of the energy of interest (keV) and the counting time of the gamma radiation are decay correction factors of the peak; these factors are used to obtain the neutron flux correction factor, which quantifies the gradient of the flux between the irradiation position of the sample and the comparator.

The neutron flux correction factor is determined as the ratio between the flux measured with the 0.1% Au-Al flux monitors at the sample position and that of the comparator. The neutron flux is proportional to the number of counts in energy range of  $^{198}\text{Au}$  and depends on other factors such as the neutron capture cross-section, isotope abundance, irradiation time,  $^{198}\text{Au}$  half-life, detection efficiency, and number of accounts for the emission energy of the radioactive isotope. Taking the relationship between the flux readings, all the terms except for the number of counts registered for the monitor at the sample position and for the monitor at the comparators position are canceled, obtaining:

This relationship is fulfilled, assuming that the irradiation conditions and counting geometries are similar.

## 6. Radioactive waste disposal

In compliance with national regulations, the Neutron Activation Analysis Laboratory has developed a simple scheme for the safe management of activated samples once they are no longer needed. These procedures are authorized and monitored by the National Nuclear Authority (Ministry of Mines and Energy). The aim of radioactive waste management is to isolate and apply protective measures to this type of waste so that there are no foreseeable future human health risks and/or negative effects on the environment.

Most of the radioactive waste corresponds to plastic materials (vials, racks, bags, tapes, etc.), as well as flux monitors and samples activated during irradiation, and also elements used in the decontamination of working areas (gloves, paper towels, plastic bags, etc.). Most of the waste is classified into three categories: Group 1 – Exempt Waste (EW), Group 2 – Very Short-Lived Waste (VSLW) and Group 3 – Very Low Level Waste (VLLW) [16].

The NAA Lab has a decay room for the temporary storage of VSLW, equipped with the necessary shielding and equipment for its safe handling. The room is locked and permanently monitored, admission is restricted to non-operating personnel, unless authorized otherwise. The stored waste is properly labeled and grouped into packages each workweek.

In order to classify and monitor temporarily stored waste, packages are analyzed by gamma spectrometry. Each package is analyzed separately in the GC-7020 detector at a reading geometry of 10 mm during 4 h, determining the activity of each of the radionuclides present.

Radioactive waste is discharged as conventional waste only when its activity reaches acceptable levels established by national regulations, with previous knowledge and consent of the Regulatory Authority. Procedures for the Management of Radioactive Waste are lined up with technical and administrative requirements established by national regulations. The following schematic allows for the safe management of radioactive samples and activated materials generated during practice.

- a. Minimization: Only materials and samples directly linked to national projects are irradiated in order to generate useful information for the economic and social development of the country.
- b. Segregation: Separating waste generated during short and long irradiations and that collected during decontamination activities.
- c. Pre-treatment: Waste package preparation, 16–20 samples per package.
- d. Classification: By activity concentration and half-lives of nuclides present in the sample.
- e. Characterization: Classification and monitoring of temporarily store waste are performed by Gamma Spectrometric analysis of the packages generated each workweek. Every package is analyzed separately in Canberra HPGe GC-7020 detectors, at a reading geometry of 10 mm during 4 h, these reading are then analyzed and the activity of each radionuclide present is determined.
- f. Storage: The NAA Lab has a decay room for the temporary storage of VSLW, equipped with the necessary shielding and equipment for its safe handling. The room is locked and permanently monitored, admission is restricted to non-operating personnel, unless authorized otherwise. The stored waste is properly labeled and grouped into packages each workweek.

## 7. Quality assurance

As part of the validation process for NAA using HPGe detectors and future accreditation of the laboratory under ISO/IEC 17025:2005 [21], the technique has been validated for the determination of rare earths such as La and Ce, and elements of interest such as U and Th in geological matrices. The following parameters were taken into account: selectivity, linearity, reproducibility, limits of detection and quantification, robustness and uncertainty estimation.

This process included the evaluation of detection limits and quantification of gamma radiation spectra obtained, according to the statistical criterion of Currie [22]. The following results show element concentrations in the sample in units of mg/kg. Ba: 129, Ce: 1.37, Co: 0.20, Cs: 0.29, La: 0.11, Rb: 10.06, Sb: 0.054, Sc: 0.024, Th: 0.27, U: 0.2. These results were comparable to those reported by other laboratories, thus demonstrating the competence of the NAA Laboratory on multielemental analysis in geological matrices.

Certified reference materials (rocks, soils, sediments and coals) are used as comparators for the implementation of the technique, and they serve as a basis to compare known quantities of an element with fractions of elements in the sample. Internal standards are used for routine control of the method, and these standards are geological materials with known concentrations reported by other laboratories worldwide that use NAA or other analytical techniques, these standards are evaluated under the same analytical conditions of the problem samples.

For the determination of uncertainty, the steps recommended in the Reference Guides [23, 24] were followed. First, the measurand was defined, establishing its relation with influence quantities and identifying them. Identifying those with greater contribution were evaluated according to their type: A or B. Finally, the combined uncertainty and the expanded uncertainties are quantified.

The uncertainty estimation was done following the bottom-up approach. The procedure consisted of establishing the measurand, identifying and quantifying the sources of uncertainty and finally determining the combined and extended uncertainties. The sample’s mass, standard’s mass, neutron flux gradient, counting geometry differences, and sample count statistics were evaluated as sources of uncertainty. Sample and standard count statistics as well as differences in irradiation geometry were identified as the main contributors to the uncertainty [25, 26]. The combined relative uncertainty for the studied elements oscillates between 2 and 8% (**Table 2**).

The results of the evaluation of performance: limits of detection, intermediate precision, robustness, veracity and uncertainty, meet the requirements established for the test method;

Element	Isotope	Half-life (days)	Energy 1 (keV)	Energy 2 (keV)	LOD (mg/kg)	Precision (%)	Veracity (%)	Uncertainty (%)
La	La-140	1.68	1596.2	487.0	0.11	6.1	97.4	4.0
Sb	Sb-122	2.72	692.8	–	0.05	9.5	103	1.9
U	Np-239	2.36	106.1	228.2	0.25	6.9	100	8.9
Ba	Ba-131	11.5	216.1	496.3	129	5.1	96.1	8.8
Ce	Ce-141	32.5	145.4	–	1.37	2.8	101	9.9
Co	Co-60	1925	1173.2	1332.5	0.20	3.2	95.6	5.8
Cs	Cs-134	754	604.7	795.9	0.29	2.1	103	6.9
Rb	Rb-86	18.6	1076.6	–	10.1	5.8	95.9	6.7
Sc	Sc-46	83.8	889.3	1120.5	0.02	3.5	98.3	6.2
Th	Pa-233	27.0	312.2	–	0.27	3.3	97.6	8.3

**Table 2.** Multi-elemental validation results.



establishing a line of work toward further validation of more elements and offering the scientific community a proven method according to international standards [27].

As part of the application for the accreditation process under the ISO/IEC-17043 standard [29], and the need for constant validation in performance and quality assurance, the laboratory participates in annual IAEA-WEPAL (Wageningen Evaluating Programmes for Analytical Laboratories) Proficiency Tests and Inter-laboratory Comparisons, obtaining excellent results and positioning its metrological competence, this being a major step toward accreditation under ISO/IEC 17025 [21].

## 8. Data applications

The Colombian Laboratory for Neutron Activation Analysis, CNAAL, is an installation oriented to the generation of high-quality analytical data that contribute to the geoscientific knowledge of the national territory, represented in the characterization of our valuable mineral and hydrocarbon resources. This potential of CNAAL's analytical technique can now be applied in vast areas of the country, which for decades were the scene of a long, costly and painful armed conflict, which ended in 2016 with the signing of the Peace Agreements between the Colombian State and the FARC guerrillas, the oldest in our continent.

Our laboratory has focused its analytical capabilities on the exploration of rare earth elements, which according to the OECD study [30] present a relatively favorable scenario for the search for these strategic minerals that present a greater supply risk taking into account its typical scarcity.

Rare earth elements (REEs) are central in information and communications technologies and green technologies, which is one of many reasons that justify studies in this area. In this way also, OECD's Cost of Inaction and Resource Scarcity; Consequences for Long-term Economic Growth (CIRCLE) Project "...aims at identifying how feedback from poor environmental quality, climate change and natural resource scarcity are likely to affect economic growth in the coming decades" [28].

Additionally, the characteristic mobility of REE is useful for the study of petrogenetic processes and the study of the geochemical cycle of uranium and other associated energy minerals.

Other applications planned for neutron activation analysis technique are related to: advances in the validation of analytical methods to determine elements, quality assurance by ISO/IEC 17025, continue with successful participation in the IAEA – WEPAL proficiency test and promotes future developments to generate impact researches on selected topics on geological materials characterization (rocks, soils, sediments, minerals and hydrocarbons), forensic sciences (element traces in crime scenes), archeometry (studies of provenance of bones, paintings, pottery, coins) and environmental sciences (mobility and accumulation of eco-toxic elements in humans, from technological and industrial processes and evaluation of environmental impacts in the biotic

components), among others. In order to consolidate the credibility of our results, the LAAN has participated in a series of intercomparison exercises whose results have been improved to date.

Recent advances in the characterization of the neutron flux (thermal and epithermal) of the modern Colombian nuclear reactor IAN-R1 (upgraded in 2015), allow the CNAAL a window of opportunity for the implementation of the “ $k_0$  – NAA method”, to improve the analytical capabilities of the laboratory, placing it at the level of other facilities of similar characteristics in other countries [29].

## 9. Conclusions

The Colombian Neutron Activation Analysis Laboratory and the Nuclear Research Reactor IAN-R1 area singular scientific and technological facilities in our country, located at the Colombian Geological Survey (Servicio Geológico Colombiano). CNAAL plays an exceptional role oriented to cover technical topics in sustainable exploitation of mineral resources and hydrocarbons, developments in nuclear sciences and researches in environmental and forensic sciences. At this point, the laboratory is available to the entire scientific and academic community of the country and recently upgrading of the Nuclear Research Reactor IAN-R1 (2014), also contributes to these national goals.

The CNAAL has state-of-the-art technology and competent personnel, which allows it to expand the coverage of research services in the country, through the Neutron Activation analysis in the execution and development of research projects.

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## Author details

Guillermo Parrado\*, David Alonso, Julián Orozco, Mary Peña, Andrés Porras, Martha Guzmán, Nelson Acero and Mauricio López

\*Address all correspondence to: gparrado@sgc.gov.co

Colombian Geological Survey (Servicio Geológico Colombiano), Bogotá D.C., Colombia

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