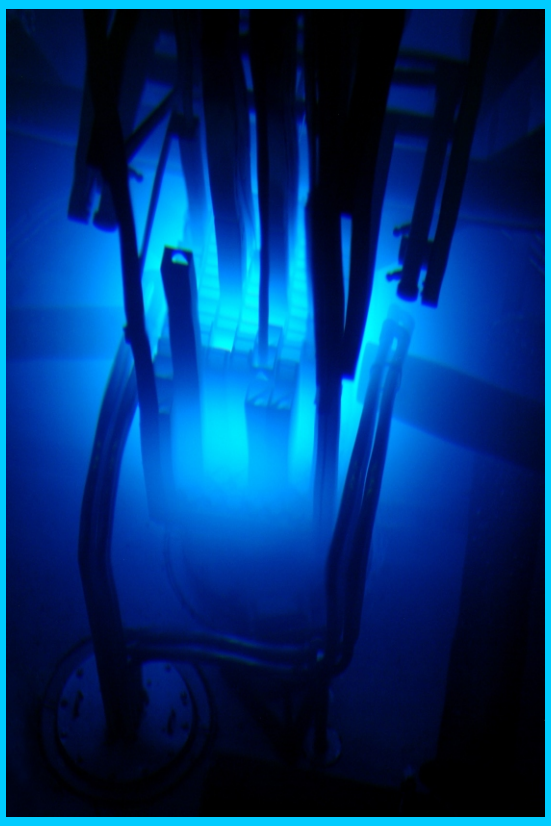
Nuclear Science and Technology



Introduction

The Program on Nuclear Science and Technology comprehends Nuclear and Condensed Matter Physics, Neutron Activation Analysis, Radiation Metrology, Radioprotection and Radioactive Waste Management. These activities are developed at the Research Reactor Center, the Radiation Metrology Center and the Radioactive Waste Management Laboratory. The Radioprotection activities are developed at all radioactive and nuclear facilities of IPEN.

The Research Reactor Center at IPEN is responsible for the operation and maintenance of the research reactor IEA-R1 and has a three-fold mission of promoting basic and applied research in nuclear and neutron related sciences, providing educational opportunities for students in these fields and providing services and applications resulting from the reactor utilization.

Specific research programs include nuclear structure study from beta and gamma decay of radioactive nuclei and nuclear reactions, nuclear and neutron metrology, neutron diffraction and neutron multiple-diffraction study for crystalline and magnetic structure determination, perturbed -angular correlation (PAC) using radioactive nuclear probes to study the nuclear hyperfine interactions in solids and neutron activation analysis, both instrumental as well as involving radiochemical separation applied to the fields of health, agriculture, environment, geology and industry. The research in the areas of applied physics includes neutron radiography, scientific computation and instrumentation.

During the last several years a special effort was made to refurbish the old components and systems of the reactor, particularly those related with the reactor safety improvement, in order to upgrade the reactor power. Primary objective was to modernize the IEA-R1 reactor for safe and sustainable operation to produce primary radioisotopes, such as 99Mo and 131I, among several others, used in nuclear medicine, by operating the reactor at 5 MW on a schedule of 120 hours/week continuous operation. A major operation was carried out in 2007, with the cooperation of the United States Department of Energy (USDOE), which enabled the transportation of a second batch of 33 spent fuel elements of US origin, stored in the wet storage facilities of the reactor pool since 2000, back to the United States. In the year 1999 a similar program transferred back to US 127 spent fuel elements.

The Radiation Metrology Center was established in October 2002. All activities of research and development, services, supervision of graduate and undergraduate students and courses performed at the Center are related to the development, improvement and establishment of new methodologies or products in radiation metrology, aiming to assure the safety of IPEN workers, community and environment. Services such as personnel and environmental dosimetry, high dose and accident dosimetry, metrology in diagnostic radiology and radiotherapy, calibration of instruments and radioactivity determination in foodstuffs and food commodities (imported and exported by Brazil) are offered to internal and external communities. The financial resources for research and development are supported by scientific governmental agencies.

The Radioprotection Service cares for the radiological safety of IPEN workers and general population, through radiation control programs and using national and international standards. Research related to the main activities is also performed.

The Radioactive Waste Management Laboratory receives, characterizes, treats and stores radioactive wastes generated at IPEN and at external communities, using national and international standards. Several research projects related to the main activities are also undergoing.

Study of the crystalline and magnetic structures of materials by neutron diffraction

The IPEN Neutron Diffraction Group is, presently, involved in studies of Rietveld quantitative phase analysis employing both neutron and X-ray diffraction patterns. The X-ray diffraction patterns are measured at different laboratories. The neutron diffraction patterns are measured in the IPEN-CNEN/SP PSD neutron powder diffractometer. The neutron powder diffractometer was constructed at the IPEN machine shop and installed at the IEA-R1 research reactor of the Institute. It is an extensive upgrading of the old IPEN-CNEN/SP multipurpose neutron diffractometer "Aurora". The old diffractometer was a single-detector instrument with a boron trifluoride (BF^3) detector and a flat copper mosaic single crystal monochromator. The main modifications introduced in the old instrument was the installation of a position sensitive detector (PSD) and a double-bent monochromator. The PSD is formed by eleven ^{3}He linear detector elements clamped together at each end to form a rigid plane. Placed at a distance of 1,600 mm from sample, the PSD spans an angular range of 20 of a diffraction pattern, with a quite good resolution. An extended powder diffraction pattern can be obtained by moving the detector and collecting the data in 20 segments in a 2 interval ranging from 5 to 130. In order to increase the neutron beam flux at the sample position, a focusing Si perfect single crystal monochromator was installed in the instrument. The monochromator is made of 9 vertically stacked Si single crystal blades, mechanically bent in the horizontal plane and quasi-bent by segmentation in the vertical plane. With a take-off angle of 84, the monochromator can be positioned to produce 4 different wavelengths, namely 1.111, 1.399, 1.667 and 2.191 A (nominal values). A rotating-oscillating collimator (ROC), placed at the entrance to the detector shield, eliminates parasitic scattering from furnace or cryorefrigerator heat shields in the vicinity of the sample, while only reducing the scattered intensity by ca. 10%. The collimator also makes the PSD less sensitive to the ambient background leaking in through the shielding entrance. A beam shutter installed in the instrument protects the operator during sample manipulation or installation of any device at the sample position. Recently, a sapphire filter has been installed in order to remove fast neutrons from the neutron beam that impinges the sample. This lowers the background level in the diffraction patterns obtained with the instrument. In comparison to the former instrument, the new diffractometer has a better resolution and is ca. 600 times faster in data acquisition. The IPEN-CNEN/SP PSD neutron powder diffractometer has been designed mainly for crystalline and magnetic structures determination and for application of the Rietveld method in quantitative phase analysis, although other different studies can be performed after an analysis of viability. Utilization of this instrument is open for cooperative studies with the latin-american scientific and technological communities.

Hyperfine interactions in solids

Experimental measurements of hyperfine interactions (interactions between the nuclear moments and magnetic field or the electric field gradient) provide a very sensitive and accurate method to investigate condensed matter phenomena in many different solids. A large variety of phenomena in solid materials, in general, originates from small differences in their electronic structure. In this perspective, it is of specific interest to investigate new material and compounds in order to understand the origin of such phenomena from an atomic view. The hyperfine interactions technique involving the measurement of Perturbed gamma-gamma Angular Correlation (PAC) is being used to investigate a series of intermetallic compounds and metal oxides which present interesting properties like superconductivity, magnetic order, phase transitions, etc. Biological materials like proteins and DNA are also a recent subject of investigation. The PAC techniques uses radioactive nuclei implanted in the solids, which can probe magnetic hyperfine field (mhf) and electric field gradient (efg) in determined sites of crystalline structure of the material and provide information about the electronic charge and spin structure around the probe. This information makes possible to investigate properties of the crystal structure and or the origin of magnetic interactions in the material. Due to the proximity of a nuclear research reactor, our laboratory can use a variety of special radioactive probe nuclei such as ^{140}La, ^{111}Ag, ^{111m}Cd besides the usual ones like ^{111}In and ^{181}Hf. A 4-BaF_2 detector spectrometer setup is available in the laboratory and a 6-detectors spectrometer has been set up which incorporated improvements in the associated electronics in order to maximize the detection efficiency. A methodology using the ^{6}Li ion beam from the Pelletron accelerator in IFUSP to implant ^{111}In probe into the sample through ^{108}Pd(6Li,3n)^{111}In nuclear reaction was developed and it is also available. The compounds which are being investigate are: Metal Oxide: The PAC technique has been used to study the hyperfine interactions in the magnetic and paramagnetic regions of the distorted perovskites RTO_3 where R = rare-earth element and T = Cr, Fe, Co, Mn, using dilute ^{111}In $\rightarrow$ ^{111}Cd and ^{181}Hf $\rightarrow$ 181Ta nuclear probes which were introduced into the samples through a chemical process and ^{140}La $\rightarrow$ ^{140}Ce produced by irradiating the sample with

quadrupole interaction parameters as well as the magnetic hyperfine field was obtained for each compound. Diluted Magnetic semiconductors: new families of semiconductors, which are doped with magnetic materials, are under intensive investigation as they can be used for spintronics. ZnO doped with Co, Mn, Fe, Ni, Cr and V are being investigated by PAC in order to understand the origin of the magnetism in these compounds. Rare-earth based compounds: series of intermetallic compounds based on rare earth elements show different magnetic behaviors and exhibit very interesting physical phenomena like Fermi liquid behavior, Kondo effect, etc. These properties are not well understood yet, and nuclear techniques are very suitable to investigate the microscopic origin of such phenomena. In our laboratory, we have studied heavy fermions compounds $CeIn_3$, and CeT_2X_2 where (T = Mn, Pd, Rh and X = Ge, Si) with PAC technique using ^{140}Ce and ^{111}Cd probe nuclei. Other families of intermetallic compounds such as RAg , RNiIn and RPdIn where R is a rare earth element are also being investigated. Ab-initio calculations: the hyperfine interaction parameters can be better understood if the electronic structure of the material is known. A very precise ab-inito method of electronic structure calculations based on the density functional theory using a local density approximation is being used to help in the interpretation of hyperfine interaction parameters through the WIEN97 code. The first-principles full potential linear augmented plane-wave (FP-LAPW) calculations of the electronic structure and hyperfine fields have been performed for the intermetallic compounds $CeIn_3$ and $CeMn_2Ge_2$ $CeMn_2Si_2$. A study of the changes induced by the presence of Zn or Ni impurity at Cu site in $CuAlO_2$ delafossite was also carried out by using FP-LAPW calculations. Ab-initio calculations for the series of compounds like RAg have been initiated.

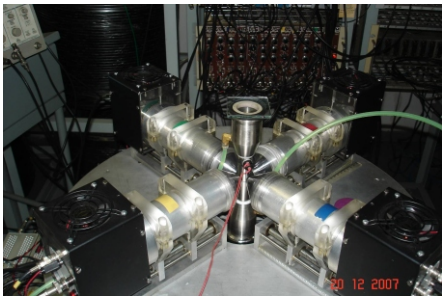


Figure 1. Angular correlation Spectrometer with 4 conical BaF^2 detectors and a sample heating furnace

Activities at the Nuclear Metrology Laboratory

In recent years the Nuclear Metrology Laboratory (LMN) has been involved in developing procedures for standardization of important radionuclides applied in nuclear medicine and as reference standards for semiconductor detectors. The primary systems used by LMN for this type of standardization are two 4πβ-γ coincidence systems consisting of a proportional counter, coupled to one or two 3"x 3" NaI(Tl) crystals, and another coincidence system employing a plastic scintillator detector in 4 geometry, called 4π(PS)β-γ which was recently developed at LMN. The disintegration rate is obtained by the application of the efficiency extrapolation technique. During the period of 2005-2007, the following radionuclides have been standardized by this technique: ^{51}Cr, ^{54}Mn, ^{55}Fe, ^{90}Sr-^{90}Y, ^{182}Ta and ^{201}Tl. The Nuclear Metrology Laboratory at IPEN has also been involved in the development of radioactive water-equivalent solid sources prepared from an aqueous solution of acrylamide, polymerised by high dose ^{60}Co irradiation. The sources have been prepared with ^{57}Co, ^{137}Cs and ^{133}Ba radioactive solutions and have density similar to water as well as good uniformity. Therefore, they are suitable for performing constancy and accuracy tests, mainly for Medical Radionuclide Calibrators. As a complementary activity related to radionuclide standardization the LMN has been heavily involved in Monte Carlo simulation of the extrapolation curves obtained in the 4πβ-γ coincidence technique. For this purpose the response functions of beta and gamma detectors have been calculated by means of two radiation transport codes, namely: MCNP and PENELOPE. These response functions are used as input data for another code developed at the LMN, called ESQUEMA. This code makes use of the Monte Carlo method for simulating all detection processes involved during radionuclide decay, being able to predict the beta and gamma detection spectra, including coincidence events and secondary radiation emission such as conversion electrons, X-ray and Auger electrons. This methodology has been successfully applied to achieve primary standardizations of ^{35}S, ^{133}Ba, ^{134}Cs and ^{182}Ta. This approach has got an enthusiastic approval during the ICRM 2005 and 2007 international conferences. Another field where LMN has been involved is neutron measurements. In 2007 activities were started on covariance analysis of the k_0 Nuclear Activation analysis (NAA)

methodology. In this period parameters involved in gamma-ray spectrometry were analyzed. An additional research area has been thermal and resonance neutron cross section measurements. This work has been performed in collaboration with the Institute of Physics from the University of São Paulo. In this period, the cross sections and resonance integrals for ^{41}K (n, gamma)^{42}K and ^{165}Ho (n, gamma)^{166}Ho nuclear reactions have been determined experimentally.

Radiation spectroscopy and spectrometry / radioactive decay

The Radiation Spectroscopy and Spectrometry Laboratory (LEER) focuses its work in the measurement of radiations, specially gamma transitions, and its scope can be divided in three main lines:

Nuclear Data
Using single gamma spectroscopy coupled to gamma-gamma coincidence and angular correlation analyses, the group at LEER have been measuring relevant nuclear data on nuclei produced via neutron irradiation in the IEA-R1 reactor, as gamma transition energies, intensities and electromagnetic nature, determining transition placements in the decay scheme, deducing beta-feeding for the excited levels, and measuring thermal neutron cross activation sections;

Nuclear Instrumentation and Methodology
Due to the exploratory nature of the work undertaken by the LEER, the group has also been developing methods and methodologies to allow a better and quicker analysis of the experimental data; this includes the development of both data reduction and analysis procedures and data analysis softwares, as well as the development of a neutron irradiation facility that doesn't rely on a nuclear reactor;

Semi-Parametric Neutron Activation Analyses
The group at LEER has also been developing a line on neutron activation analyses that replaces the use of certified standards with neutron flux monitors, using the Cd-ratio technique with Au foils.

Highlights 2005-2007:

- Analysis of the decay of ^{155}Sm, using both singles spectroscopy and gamma-gamma coincidence to find new gamma transitions and propose new excited levels, as well as performing theoretical calculations using nuclear models;

- Analysis of the decay of ^{193}Os via gamma-gamma coincidence and angular correlation, finding several new gamma transitions, proposing new excited states and strongly improving the literature information on transition intensities, nuclear models were also used to assess the Physical nature of the low-lying excited states;

- Development of an Am-Be neutron irradiator, using Monte Carlo techniques to assess its performance and to model a new one with enhanced flux, either using more Am-Be sources or a d-d neutron generator;

- Development of methods to extract precise gamma transition intensities from both gamma-gamma coincidence and angular correlation data;

- Determination of reference values for humans on whole blood for several elements using NAA;

- Elemental analyses on genetically modified food using NAA;

- Determination of several elements' concentrations on biological samples using NAA.

Neutron radiography tecnique

The neutron radiography (NR) is a non - destructive testing technique commonly employed to inspect the internal structure of objects. Because of the neutron - matter interaction characteristics this technique is largely employed to inspect hydrogenous rich substances (oil, water, adhesives, rubber, etc) even wrapped by thick metal layers as well as radioactive samples. The radiography is obtained by irradiating the object in an uniform neutron beam and a converter screen transforms the transmitted neutron intensity into ionizing radiation which is able to sensitize a film forming the image. The screens consist of strongly neutron absorbing elements (gadolinium, dysprosium, lithium) and the films are the conventional for X-ray films and the track-etch foils. Alternatively, neutron scintillators are also used as converter and in this case the light emitted sensitizes either a film or the CCD of a video camera. In the last case the radiography is obtained in real-time. For both cases the radiographs are 2-D projections of the internal structure of the object. The neutron radiography - NR activities at IPEN - CNEN/SP began in 1988 and the primary objective of the working group was design and construct an operational facility which is installed at the 5 MW IEA-R1 nuclear research reactor. From 1992 to 1997, the group has developed several radiography techniques by employing metallic dysprosium, gadolinium and boron converter screens together with conventional X-ray films and track-etch foils. In 2001 the facility has been improved and a real - time system was installed. Furthermore two new radiography techniques, by using electrons and alpha particles, to inspect objects with thickness in the m range were also developed. Nowadays the IPEN posses also two digital system to process radiographic images with which has provided services and developed high level researches. In this period (1988-2007) four Msc

and one PhD thesis have been advised and in the present three new PhD thesis by using this same neutron radiography facility are under advising. The figures 2 and 3 show examples of some typical obtained radiographs at the present facility. The NR working group intends to install a neutron tomography system in the NR facility of IPEN-CNEN/SP. Since the neutron radiography is able to provide images of objects which could not be investigated by the standard radiography techniques with X-ray or gamma radiation, the proposed tomograph will be very useful because it is the best alternative to investigate these same objects in 3-D. As has occurred in other countries, the availability of a neutron tomograph will spread the use of the neutron radiography technique in Brazil since it will reach those industrial, technological and research fields not yet reached by the conventional radiography techniques.

Highlights 2005-2007:

- Development of the neutron induced radiography(NIR) techniques to inspect thin samples.
- Development of a quasi real-time device to inspect radioactive samples by the neutron radiography.
- Development and implementation of a digital system to inspect radiography images in track detectors.
- Development and implementation of a digital system to characterize track detectors.

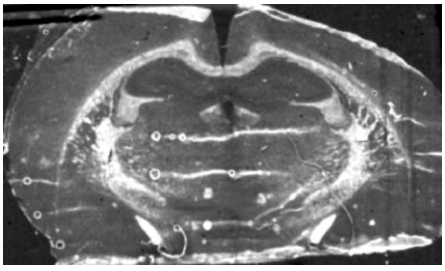


Figure 2. Biological tissue (brain)

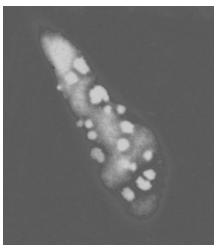


Figure 3. Bacteria colony

Scientific Computing applied to the Nuclear Area

Computing is a tool as important as experimentation and theory in solving the scientific challenges in the nuclear science of the twenty-first century. Central to the mission of our group is enabling the use and exploration of computational science to investigate fundamental problems in nuclear and related areas that require massive calculations and have broad scientific and economic impacts. Examples of these problems include dosimetry and micro-dosimetry modeling and simulation, radiotherapy and nuclear medicine modeling, simulation and planning, nuclear experiments modeling and simulations, and many more. The group's mission also encompass the development of new scientific software for analysis, visualization and exploration of data, employing advanced technologies from computer science and related areas. All this is done with a strong commitment to leverage student's abilities in scientific computing applied to the nuclear area, contributing to form competent scientists for the Brazilian Nuclear Program. Scientific Computing is considered nowadays a third mode of science, besides experimentation and theory. It now plays an equal role to theory and physical experiments in discovery-driven scientific research. Numerical simulation technology offer the possibility to model and simulate, maybe otherwise impossible, experiments that require expensive and inaccessible systems. It's even possible to model a complete nuclear physics facility, including radiation sources, filters, detectors and other parts, execute the simulation and validate the data against published experimental results. To accomplish such studies, however, solid computing skills are required. It's essential to deal with several programming languages, distributed processing and large scale software development, installation and management. Also the integration of several different software systems are essential to be able to successfully exploit computing systems for scientific use. Our group has been working with a Monte Carlo simulation software, mainly Geant4, applied to dosimetry, medical instrumentation and detector studies. Personal dosimeters as well as dose equivalent coefficients have been studied. X ray beams used in radiotherapy and diagnostic have been simulated; we also collaborate with international scientists who are also using Geant4 for dosimetry studies. We also developed a scientific software applied to Instrumental Neutron Activation Analysis; the software is now in the final testing phase before going to production.

This software will allow the incorporation of several gamma spectrum analysis techniques. Also planned is the networked version, which will bring to the laboratory, besides the client/server technology, the database persistence and reliability, allowing, in the future, mining techniques to uncover hidden features in data. The programming language of choice for this software was Python. Being widely used by the scientific community around the world, Python features, such as rapid development, will allow new techniques and algorithms to be quickly tested in the software.

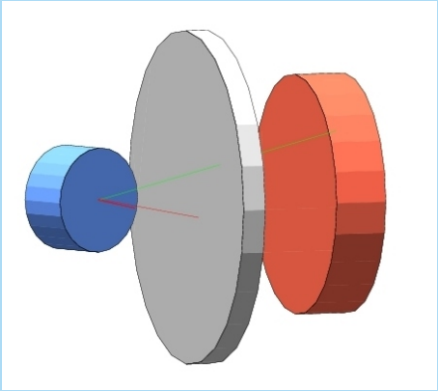


Figure 4. Simulation of a X ray tube: tungsten anode (blue cylinder) shot by an electron beam (red line), producing X rays (green lines) that cross the beryllium window (grey cylinder) and reach the volume to be irradiated (red cylinder)

Nutritional studies in foodstuffs and diets

The forms of malnutrition are numerous and varied, that may lead to serious health problems such as under-nourishment or obesity. In the developing countries, the nutritional problems are due mainly to specific nutritional deficiencies. About 2 billion people suffer from "occult" hunger: deficit of micronutrients such as iron, vitamin A, iodine and zinc. Children and women are particularly vulnerable of food insecurity. Information about trace element distribution is of significant importance in assessing the adequacy of human diets with respect to the intake of essential elements and in evaluating the potential health risks arising from exposure to toxic elements in foodstuffs. Essential and toxic elements have been measured extensively in Brazilian food and diets of different groups by Neutron Activation Analysis. Studies carried out in the period 2005-2007:

Iodine daily dietary intake in a group of Brazilian workers
The main role of iodine in nutrition arises from the important part played by the thyroid hormones in the growth and development of humans and animals. The recommended dietary allowance (RDA) for adult men and women is 150 g/day and the tolerable upper intake level (UL) for adults is 1,100 g/day. Iodine deficiencies can be prevented or reduced by increasing of its dietary intake through fortification of food. There is an intensive international effort to fortify the cooking salt with iodine in several countries, including Brazil. In this study, iodine dietary intake was evaluated through determination of iodine in duplicate portion diet samples by epithermal neutron activation. The collection of samples was carried out in a group of twenty-six workers from a steel industry of São Paulo city. The average daily dietary intake found for worker's group was 840 g/day, ranging from 400 to 1540 g /day. Some of the daily iodine intakes were about 10 times higher than the RDA value.

Total Diet Study: the nutritional elements variability
Since Total Diet Study (TDS) was first conducted in the U.S, in 1961, developed countries have been carrying out their own studies periodically. This study is the first attempt to establish a baseline for TDS in Brazil. Food consumption data source was from a recent household food budget survey (2002-2003) by the Brazilian Institute for Geography and Statistics, which contained 5441 foods. This study provides the nutritional elements contents of Na, K, Ca, Fe, Zn, Se and Cr in 37 most consumed foods (more than 2 g/day/person) in among people in the state of São Paulo, Brazil. Foods were grouped into 15 food groups, according to their chemical characteristics. Food samples from restaurants of the University of São Paulo were prepared table-ready and analyzed by Instrumental Neutron Activation.

Essential and toxic elements, natural and artificial radionuclides in edible mushrooms
Mushrooms are excellent nutritional sources since they provide proteins, fibers and mineral, such as K, P and Fe. They have also been the focus of environmental studies. In Brazil mushrooms are not consumed in large quantities by the general population since people know little about the nutritional and medicinal benefits that mushrooms offer. Hence, this study intends to contribute to a better understanding of the essential and toxic element content in edible mushrooms, which are currently commercialized in São Paulo state. Concentrations of As, Br, Cr, Cd, Fe, K, Na, Se and Zn and the activities of ^{40}K, ^{137}Cs, ^{238}U and ^{232}Th were determined in edible mushroom species: Shitake (Lentinus edodes), Shimeji (Pleurotus ssp), Paris Champignon (Agaricus bisporus), Hiratake (Pleurotus ssp) and Eringue (Pleurotus Eryngüi).

Essential element determinations in diet and light foods
The consumption of dietetic products, diet and light food, has become a significant part of the Brazilian population's eating habits. It seems to be connected to people's concern with their quality and style of life. Despite an increased emphasis on dietetic foods in traditional Brazilian diet, the knowledge of their chemical compositions in the field of nutrition and toxicology is restricted. The aim of this study is to determine essential elements in diet, light and their similar regular foods by Neutron Activation Analysis and to compare the results of these different categories. For sampling, diet, light and regular food samples: artificial sweetener, cappuccino and chocolate milk were collected in the market of São Paulo city.

Essential elements in milk formulas
The World Health Organization (WHO) recommends that infants up to 6 months be exclusively breastfed. Breastfeeding should continue along with complementary feeding until 2 years of age. However, this is not always possible due to the fact that not all mothers are able to produce sufficient amounts of milk for their infants. According to Codex Allimentarius, the best substitute

choices for maternal milk are infant milk formulas, and when prepared under proper hygienic conditions can be used to feed infants. There are no published data on element content in infant milk formulas marketed in São Paulo city. The objective of this study was to determine element (Br, Ca, Na, K, Fe, Rb, Se and Zn) content by Neutron Activation Analysis in the infant milk formulas consumed in São Paulo city and to compare with the printed labels.

Determination of mineral and trace elements in foodstuffs and diets from the Amazon region

The Amazon region is characterized by the availability of fruits rich in pro-vitamin A and fishes with high protein content. However, there is little information about the nutritional potentiality of these foodstuffs mainly concerning the content of fibre, phytate and minerals in the fruits and vegetables. The purpose of the present study was the establishment of the chemical composition of these foodstuffs by using the instrumental neutron activation analysis. About 25-30 different species of regional fruits and vegetables (only the edible part of these materials) were analyzed. This work is being carried out with the cooperation of Dr. Lucia K. Yuyama from INPA (Instituto de Pesquisas da Amazônia). Also different pre-school diets from this region were analyzed during this period.

Assessment of the Content of Mercury, Methylmercury and Other Elements of interest in Fish, Hair and Diets of pre-scholl children of the Amazon region

The Amazon region suffers mercury (Hg) impacts as a direct result of both natural and anthropogenic processes. Jaú National Park (PNJ) is the only National Park in Brazil that protects an entire black water basin (Jaú River), flood land and tropical reserve. These conditions favor Hg methylation in the aquatic biota. Preliminary studies of pre-school diets from PNJ communities have shown that these diets have a worrisomely high Hg content. The present study assessed total Hg content, micronutrients (Ca, Fe, K, Na, Se and Zn) and macronutrients (proteins, lipids, ash, energy, carbohydrate) in pre-school diets in the PNJ and surrounding communities. Furthermore, total and MeHg levels were also determined in hair samples of these children as well as those living in several neighborhoods of the city of Manaus. Included in this determination were the fish most consumed by these populations. From these results it was possible to evaluate the nutritional content of the diets and the exposure of the children to Hg and MeHg.

Study on the Element Migration from Plastic Packaging to food using Radiometric Method

in the last few years, problems related to food contamination by substances or elements that can be a risk to human health have been concerned, not only to government authorities, but also to general population as well. Within this context, plastic packagings can constitute a source of food contamination since plastic manufacturing processes involve the use of catalysts and different types of additives that may contain toxic elements. In this project a radiometric method was developed for element migration determination from plastic food packaging to food simulating or to the food itself, according to National Health Surveillance Agency (ANVISA) regulations. Element migration was determined for plastic packaging used for soft drinks, drinking water, milk, dairy products, juices and fatty foods and the results indicated the migration of Cd, Co, Cr and Sb present in these plastics. In some packagings, migrations of only some these elements were observed and in these cases the detection limits were determined. Results obtained indicated the potentiality of using radiometric method in the elements migration evaluation from plastic packages to their contents. Among the main advantages of radiometric method for element migration evaluation are its simplicity, no need of blank analysis and the possibility to obtain element migration to the food itself.

Correlation studies of trace element concentrations in human tissues and the Brazilian population health

Determinations of trace elements in human tissues are of great interest in the health area. With the improvement of analytical techniques and knowledge of the role of trace elements in human organism, the correlation studies between trace elements and their effects have became a challenge of many researchers. The NAA laboratory of IPEN over these years has analyzed different types of human tissues such as bone, teeth, lungs, hair and nails and interesting results have been obtained. In the period of 2005-2007 we focused in the analyses of blood serum of healthy elderly population and brain samples of normal and demented individuals of São Paulo Metropolitan region. Reliable protocol was established for serum sampling, reference population selection, control of possible sample contamination and quality control of the analytical results since recent studies have been concerned with reliability of the serum analysis results and their usability in clinical chemistry and medical investigations. To avoid contamination of sample the handling of the biological samples was performed inside a laminar flow hood (Fig1.). The evaluation of trace element levels in sera of an elderly population is of particular interest especially in regard to healthy longevity. The ageing is a period of life characterized by trace element imbalance due to many factors such as physiological alterations with lower absorption of nutrients, lower food intake and reduction of physical activities. On the other hand, presently

the fastest growing population group in the world is that over 60 years old, however data on serum trace element analysis for this group are very scarce. Serum Ca, Fe, Rb, Se and Zn concentrations were obtained by INAA at the IPEN-CNEN/SP and serum Cu, K, Mg, Na and P, as well as, biochemical and hematological parameters were determined at the Central Laboratory Division, HC-FMUSP. Serum element concentration ranges obtained for healthy elderly from the "Successful Ageing" Program of FMUSP indicated that they are within reference ranges and, 32% of elderly presented elevated total cholesterol levels but 93% showed desired HDL and 59% of desired LDL concentrations. Cerebral tissues of an over 50 year old population of both genders provided from the Brain Bank of the Brazilian Aging Study Group (BBBABSG) were analyzed in order to elucidate about neurodegenerative diseases resulting from the gradual and progressive loss of neural cells which leads to the dysfunction of the nervous system. Trace elements may contribute to the pathogenesis of cognitive disease since they may increase oxidative stress and also can influence the homeostasis of other elements and accelerate the aggregation of amyloid beta peptides. Concentrations of Br, Fe K, Na, Rb, Se and Zn were determined in different compartments of brain. Significantly higher concentrations of some elements were found in the hippocampus of demented individuals than those presented by the normal group in the corresponding brain parts. Partnerships: IBt and FMUSP. Financial Resources Agencies: FAPESP and CNPq.

Figure 5. Laminar flow hood for handling human biological samples

Environmental applications of neutron activation analysis

Nowadays one of the most dangerous kinds of pollution in the Earth's ecosystem is resulting from heavy metals dumping. Its increasing use in industries and other activities considered to be essential in modern human life, has resulted in a modification of natural geochemistry cycle of these elements, increasing their dispersion in the environment. Pollution studies require highly sensitive analytical techniques, with high precision and accuracy. Instrumental neutron activation analysis (INAA) has been used for the determination of heavy metals and other trace elements in different environmental samples.

Sediments

The study of the distribution of metals in sediments is very important from the point of view of environmental pollution. The sediment concentrates metals in aquatic systems, and represents a relevant contamination monitor. Studies of sediments from estuaries which have been polluted by heavy metals represent the comprehension of transportation phenomena in these complex ecosystems and the discovery of the pollution history. The contamination of Sepetiba Bay (Rio de Janeiro) using a multi-element approach was estimated from sixty bottom sediment samples. The elements Pb, Cd, Cu, Ni and Zn were determined. The results showed very strong contamination of zinc (up to 1000 µg g-1). Cd and Zn presented values which may cause hazards to aquatic organisms. The contamination of Rio Grande tributary, Billings reservoir, in the Metropolitan region of São Paulo, by determining metal concentration and other elements of interest using three analytical techniques (INAA, AAS and ICP OES) were assessed in bottom and core sediment samples. The chosen chemical parameters for this characterization were Al, As, Ba, Cd, Cu, Cr, Fe, Pb, Mn, Hg, Ni, Se and Zn. From these assessments the most adequate technique was selected for the routine analysis of sediment samples for each element concentration determination. The concentration values obtained for the metals As, Cd, Cu, Cr, Hg, Ni, Pb and Zn were compared to the Canadian Council of Minister of the Environment (CCME) oriented values (TEL and PEL values). The contamination of two estuarine systems: a lagoon-estuary complex area of Cananéia and Santos-São Vicente, located in the coast of São Paulo State was evaluated. Cananéia is considered as part of Biosphere Natural Reserve due to its environmental and cultural importance and is considered not polluted. Santos - São Vicente estuary is an example of environmental degradation in coastal systems of industrial origin. The assessment concerning the distribution of some major (Fe, K and Na), trace (As, Ba, Br, Co, Cr, Cs, Hf, Hg, Rb, Sb, Sc, Ta, Tb, Th, U and Zn) and rare earth (Ce, Eu, La, Lu, Nd, Sm, Tb and Yb) elements in sediment samples was

done by using INAA technique. Fifty bottom sediment samples were collected in each estuary in four campaigns: summer and winter of 2005 and 2006.

Soils

The urban environment quality is of vital importance as the majority of people now live in cities. Metals occur naturally in soil, but contents are generally increased in the urban environment due to anthropogenic activities. The platinum group elements Pt, Pd and Rh are the active components of car catalysts and are being spread into the environment to an as-yet incompletely known extent due to surface abrasion of the catalyst during car operation. São Paulo is a city with 19 millions of inhabitants which shows severe pollutions problems. There has been little research on metal concentration levels in soils of São Paulo. This study presents the results obtained for the concentration levels of potentially toxic elements (As, Ba, Cr, Cu, Pb, Sb and Zn) and platinum group elements in urban soils of São Paulo (green areas, public parks and soils near streets and avenues with high traffic density). The results obtained showed concentration levels of the analyzed elements higher than the values considered as reference values for soils in São Paulo, according to the Environmental Protection Agency of the State of São Paulo guidelines. These results suggest an anthropogenic source and indicate a potential damage to soil quality.

Biomonitoring of marine and atmospheric pollution

- Biomonitoring of coastal areas using marine organisms is an attractive approach for studying pollution caused by anthropic discharges. Most of the experiments are based on collection and analysis of native organisms (passive biomonitoring), but this method has the disadvantage of dealing with many natural variations. In this work, the marine bivalve Perna perna, very abundant in the coast of the State of São Paulo, Brazil, was transplanted from a mussel farm (active biomonitoring) and used for biomonitoring of four sites (Itaipu and Ilha das Palmas, in Santos and TEBAR oil terminal and Ilha Bela ,in São Sebastião) , situated in coastal regions close to domestic and/or industrial discharges. Hg, Cd and Pb were determined in the transplanted organisms by AAS and As, Ca, Co, Cr, Fe, Na, Se and Zn were determined by INAA. After the transplant experiments of the organisms to the sites of study, a rise in concentrations was observed for all elements, depending on the season and site of study thus indicating the applicability of the Perna perna mussel as biomonitor. It could be observed that the concentrations of As and Se were always above the tolerance limits of the Brazilian legislation (1.0µg.g-1 for As and 0.10µg.g-1 for Se), in all sites of study, including the control site. For the potentially toxic elements Cd, Hg, Pb and Zn, the observed for all elements, depending on the season and site of study thus indicating the applicability of the Perna perna mussel as biomonitor. It could be observed that the concentrations of As and Se were always above the tolerance limits of the Brazilian legislation (1.0µg.g-1 for As and 0.10µg.g-1 for Se), in all sites of study, including the control site. For the potentially toxic elements Cd, Hg, Pb and Zn, the concentrations obtained were always below the maximum limits established by the legislation (1.0µg.g-1 for Cd, 2.0µg.g-1 for Pb, 0.5µg.g-1 for Hg and 50µg.g-1 for Zn). As for Cr, in fall and winter the control point had levels below the ones established in the legislation, but after the transplant of the organisms, these levels raised to concentrations above the limit. In spring and in summer all the points, including the control, presented Cr above the limit, except for the Ilhabela site.

- Reference materials are important tools in the quality assurance of analytical results as they may be used in method validation, calibration of instruments and in the realization of the traceability of analytical results to stated references. This study is a contribution to metrology in chemistry in Brazil and describes the preparation of a mussel reference material using the Perna perna species. This mussel species is abundant in most part of the Brazilian seashore and is currently being used in biomonitoring studies in our laboratory. It also has economical importance due to its growing cultivation in aquaculture farms for human consumption. The preparation of the reference material included sampling, sample pretreatments, freeze-drying, grinding, sieving, homogenization and gamma ray sterilization. Physical and chemical characterization following internationally agreed recommendations are being performed, with emphasis on the assessment of the stability of the

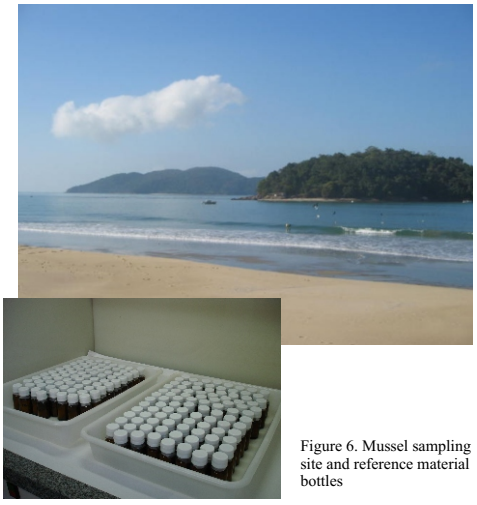


Figure 6. Mussel sampling site and reference material bottles

material, its homogeneity status and the layout for certified values assignment via an interlaboratorial program. Preliminary results (with associated uncertainties) for the concentration of elements such as As, Br, Co, Cr, Fe, K, Na, Se and Zn determined by INAA and Cd, Hg and determined by AAS show that the material is suitable to be used in environmental studies.

- Lichens have long been considered one of the most valuable air pollution biomonitors. As such, they have been widely used to assess trace element atmospheric contaminants. In order to contribute to the understanding of the air pollution profiles of São Paulo Metropolitan Region (SPMR), Canoparmelia texana lichenized fungi samples collected in the sites of unpolluted areas of Atlantic Forest, SP and in the sites near automatic monitoring stations belonging to the Environmental Protection Agency of the State of São Paulo (CETESB) were analyzed by INAA. Element concentrations of As, Ba, Br, Ca, Cl, Co, Cr, Cs, Fe, K, La, Mn, Mo, Na, Rb, Sb, Sc, Se, U and Zn were determined in these analyses. Distribution maps of element concentrations found in lichen analyses were drawn and high concentrations of Ba obtained in lichens from São Caetano do Sul and surrounding regions that make up an industrial area called ABC Paulista Region. These high concentrations of Ba can be attributed to several industries and petrochemical complex located in this region. Br distribution in the SPMR indicates that its emission source can either be from vehicular or industrial origins. For the case of Co, lichens collected in sites located in the Eastern part of the SPMR presented high level of this element. The high concentration of Co in the São Miguel Paulista site can be associated to the emission of a metallurgical industry that produces about 600 tons of this element per year. As expected, the distribution maps of element concentrations showed the highest values around the petrochemical complex, metallurgical industry and in urban areas affected by vehicular emissions. Exploratory analyses revealed that the accumulation of toxic elements in C. texana may be of used in determining the human risk of cardiopulmonary mortality due to prolonged exposure to ambient levels of air pollution. Partnerships: IBt and FMUSP. Financial support agencies: FAPESP, CNPq and IAEA.

INAA: a link between past and present in retrieving of peoples' identities

The chemical analysis on archaeological ceramics samples can provide information about production centers, trade route identification, raw material, object exchange, and prehistoric people mobility patterns. This information is possible because differences in chemical composition are typically interpreted as evidence for different production locations. During 2005 and 2007 years, INAA method at IPEN-CNEN/SP was used on hundreds of archaeological ceramics specimens in sites from several states such as São Paulo, Pará, Sergipe, Mato Grosso do Sul and Amazonas in collaboration with various archaeologists. The ceramic powder samples are obtained by cleaning the ceramics' outer surface and by drilling using a tungsten carbide rotary file attached to the end of a flexible shaft, variable speed drill. After that, this material is dried in an oven at 105°C for 24h and stored in a desiccator. Approximately 100 mg of ceramic samples, the standard reference material NIST-SRM-1633b, and IAEA-Soil-7 are weighed in polyethylene bags and wrapped in aluminum foil. Groups of 8 samples and one of each reference material are packed in aluminum foil and irradiated in the research reactor swimming pool, IEA-R1, from IPEN-CNEN/SP at a thermal neutron flux of about 8.92 x 1012.cm-2.s-1 for 1h. Arsenic, Ba, K, La, Lu, Na, Nd, Sm, and Yb are measured after a 7-day cooling time and Ce, Cr, Cs, Eu, Fe, Hf, Rb, Sb, Sc, Tb, Th, Zn and U after 3 or 4 weeks' time. Etnoarchaeological and analytical approaches are tentatively combined in these projects to study cultural practices of clay selection and use. Emphasis to differences in raw material and characteristics resulting from cultural practice is given. One of this research's goals is to detect whether stylistic and morphological boundaries reflected at the aggregate level are also manifested at the compositional level. In the compositional studies, three main objectives guide the examination of compositional variability in raw material and in finished products. These objectives are: 1) to explore whether it is possible to detect chemical compositional differences between two closely situated archaeological sites in a single geological region 2) to evaluate the closeness of fit between clay compositions from specific sources and products manufacturing and finally 3) to seek explanation for aspects of observed compositional variability. These studies are made by means of data set using several multivariate statistical methods, such as Mahalanobis distance, cluster analysis,

principal components analysis, discriminant analysis, and other analysis like kernel density, Procrustes analysis, neural network to cite just a few of the statistical methods used. Figure 7 is an archaeological ceramic with details of lizards, reptilians including snakes, caymans that are common on Marajó Island. Combinations of human faces and reptilian imagery on large funerary vessels illustrate the close link between human and animal in Marajoara symbolism.

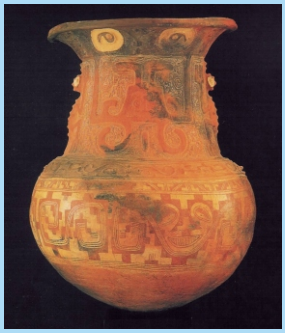


Figure 7. Funerary urn from Marajó Island (Musée Barbier-Mueller, Genève, Suisse)

Determination of rare earth elements, U, TH, and other trace elements in geological samples by neutron activation analysis for geochemical studies

Trace elements, including U, Th, Ba, Rb, Ta, Cs, Co, Hf and rare earth elements (REE), have been widely used in geochemical and petrogenetic studies due to the information they can provide about the formation and weathering of rocks. Instrumental neutron activation analysis (INAA) has been used as a powerful tool in these studies due to the high sensitivity, precision and accuracy in these trace elements determination. INAA provides multielemental analysis in concentrations of about mg kg-1 to ng kg-1, without the sample chemical attack. The Neutron Activation Analysis Laboratory at IPEN (LAN-IPEN) has several research projects in collaboration with different Universities (USP, UNICAMP, UFRGS) aiming trace elements determination in different kinds of rocks by INAA. The analytical procedure consists of weighing aliquots of about 100 mg of the powdered rock and of the geological reference material used as standard in polyethylene bags. Samples and reference materials were submitted to a neutron flux of about 1012 n cm-2 s-1 for 8 hours at the IEA-R1 nuclear reactor at IPEN. The measurements of the induced gamma-ray activity were carried out in a GX20190 hyperpure Ge detector (Canberra). The multi-channel analyser was a 8192 channel Canberra S-100 plug-in-card in a PC computer. The resolution (FWHM) of the system was 1.90 keV for the 1332 keV gamma-ray of 60Co. The gamma-ray spectra were processed by using the VISPECT gamma-ray software which locates peak positions and calculates the energies and net areas. The LAN-IPEN has participated in the GeoPT - International Proficiency Test for Analytical and Geochemical Laboratories The program GeoPT is designed to be part of the routine quality assurance scheme of analytical geochemistry laboratories. The organizers of the tests evaluated statistically all contributed data by using the Z-score criterion, which consider "satisfactory" the results falling between ±2. The elements Ba, Cs, Co, Hf, Rb, Fe, U, Th and the rare earths La, Ce, Nd, Sm, Eu, Tb, Yb and Lu were analyzed by INAA) and, as a general trend, Z-score values between ±2 were obtained for most elements confirming the good quality of the analytical results.

Characterization of micronutrients and contaminants in fertilizers and agroindustrial by-products

Utilization of fertilizers, containing macro and micronutrients, is essential for obtaining productivity necessary to feed increasing world population. Brazilian fertilizer legislation considers the total concentration of micronutrients as being the guarantee of their presence in fertilizers. This fact allows the use industrial by-products as micronutrient source for fertilizers, which brings the risk of presence of high amounts of inorganic contaminants that may lead to soil contamination, affecting harvest and the quality of the agricultural products. Elements B, Cl, Co, Cu, Fe, Mn, Mo, Ni and Zn are considered as micronutrient for the plants and microorganisms. However, they are toxic at high concentration in soil solution. The elements As, Cd, Hg and Pb are considered toxic for the animals. Uptake of these elements by plants consumed directly or indirectly by humans can be one avenue of entry to food chain. Besides the presence inorganic constituents mentioned, the analytical method of INAA is a tool very adequate for the monitoring of no routine elements such as: Br, Eu, La, Sc, Sm, Th and U, which may have influence in the environmental impact over the time. This project is being conducted in partnership with CENA/USP and EMBRAPA. Steps carried out in 2005-2007:

- Multielemental characterization of thirty-one commercial fertilizers from different manufacturers: eighteen of the type F.T.E. (Frited traces elements) and thirteen brazilian phosphate fertilizers.

- Multielemental characterization of five organic fertilizers: three sewage sludge, “biossolid”, from different treatment plants and two composed of urban garbage.

- Phytoavailability evaluation of micronutrients and contaminants in twelve commercial fertilizers of the type F.T.E. and in six brazilian phosphate fertilizers. To assess the microelemental phytoavailability, rice plants (Oryza sativa IAC 201 cultivar) were grown in pots containing 1 dm^3 of soil (FIG. 8). The trial was performed at green house of Centro de Energia Nuclear na Agricultura-USP, Piracicaba, SP, Brazil. The Typic Quartzipsamment and Typic Oxisols were the soils used in this study.

Figure 8. Partial view of green house

Environmental radioprotection

An assessment of radon and thoron concentrations was carried out in the most visited caves of the natural state park in Brazil, 'Parque Estadual do Alto Ribeira' (PETAR), by using Makrofol E nuclear track detectors placed in twin chambers. Tour guides were evaluated for committed effective doses caused by radon and thoron inhalation, giving results between 0.2 and 4.0 $mSv.y^{-1}$ for radon and 0.005 and 0.017 $mSv.y^{-1}$ for thoron. Phosphogypsum, a fertilizer industry by-product, has been worldwide stockpiled, posing environmental concerns. Since this material contains natural radionuclides in significant concentrations, its use as a building material and in agriculture has radiological implications. In order to confirm the feasibility of the use of phosphogypsum, an experimental house was recently built, having one of its rooms entirely lined with this new material. A comprehensive radiological evaluation was performed, including measurement of the external gamma exposure and radon concentrations. The results show that the annual increment in the equivalent dose to a hypothetical inhabitant of the house will remain below ICRP 60 dose limit for public of 1 $mSv.y^{-1}$. In order to study the radiological impact of using phosphogypsum in agriculture, a conservative scenario was defined considering a long term exposure due to successive annual applications of phosphogypsum in agriculture for 10, 50 and 100 years. The doses evaluated due to the ingestion of food produced by using soils tilled with phosphogypsum were always below 0.5 $mSv.y^{-1}$, and of the same order of magnitude as those obtained by using Brazilian phosphate fertilizers The Environmental Radiometric Laboratory at IPEN-CNEN/SP performs, on a regular basis, analysis of potassium-40, cesium-134 and cesium-137 concentrations in foodstuffs and food commodities imported and exported by Brazil. During the 2005-2007 period the radioactivity present in all analyzed products was within the international limits, allowing concluding that consumption of such goods offered no health risk derived from radiation.

Application of radionuclides in environmental studies

In Ubatuba coastal area, northernmost part of São Paulo Bight, time-series measurements of ^{222}Rn activities showed a negative correlation between SGD rates and tidal stage. Large fluctuations of SGD were observed at sites situated only a few meters on the shore (from 0 cm d^{-1} to 360 cm d^{-1}), as well as during few hours (from 0 cm d^{-1} to 110 cm d^{-1}), strongly depending on tidal fluctuations. The average SGD flux estimated from continuous ^{222}Rn measurements is 17 ± 10 cm d^{-1}. Integrated coastal SGD flux estimated for the Ubatuba coast using radium isotopes is about $7x10^3$ m^3 d^{-1} per km of the coast. The fluvial fluxes of the radium isotopes and dissolved barium for Ubatuba embayments, was evaluated in 2007 and covered 12 small rivers that belong to the major surface drainage system of such coastal area. Activity concentrations of ^{226}Ra in riverine waters discharging to Ubatuba embayments varied from 24 mBq $100L^{-1}$ to 705 mBq $100L^{-1}$ (in Perequê-Mirim River), while ^{228}Ra concentrations ranged from 1119 mBq $100L^{-1}$ to 3033 mBq $100L^{-1}$. Natural radium isotopes have been used as tracers to study the movement of shelf water, which is believed to be a primary source of iron to the Southern Ocean (Fig.9) and ^{234}Th as a tracer of particle flux in the upper ocean, since POC export via sinking particles is the primary mechanism of carbon sequestration in the Southern Ocean. In November 2007, the distribution of ^{223}Ra and ^{224}Ra in surface waters of Bransfield Straight and Drake Passage were determined.

Figure 9. Comandante Ferraz Brazilian Station at Antartica (EACF) at King George Island

First results indicate a marked increase in ^{224}Ra from Bransfield Straight close to Deception Island. These activities were in most cases lower than 0.1 dpm 100 L^{-1} of seawater. In the framework of AMANDES project, the distributions of the four natural radium isotopes, U and Th have been analysed in a set of 40 seawater and particulate samples collected during two campaigns carried out in October 2007 and April 2008 on board of R/V ANTEA in the Amazon region. In October 2007 campaign, ^{224}Ra activities up to 41 dpm 100 L^{-1} were observed in Amazon river waters, while ^{223}Ra activities ranged from 0.01 to 8 dpm 100 L^{-1}. The sedimentation rates and dating of a sediment core were determined by using ^{210}Pb method. The samples were collected at Lago Puruzinho, Amazônia Ocidental Brazil, in the same place where a previous Hg contamination was found. The age of sedimentary column was about 90 years and the sedimentation rate was estimated in 0.5 cm y^{-1}. The maximum concentration of Hg was found in 1982, the same period in which higher amounts of Hg were released in atmosphere.

Internal dosimetry

As a part of a continuous improvement of the monitoring programme for occupationally exposed workers at IPEN, a Web based system was developed to access the internal dosimetry database. The system was implemented using the Hypertext Preprocessor, PHP, and a PostgreSQL database. The database maintains information about worker identification, physical and chemical characteristics of the radionuclide, type of monitoring, measurement data and the dose.

External dosimetry

Aiming to improve the neutron dosimetry in Brazil a gamma-neutron mixed field dosimeter using the techniques of TLAD and SSNTD was developed. Commercially available materials were preferentially used. The dosimeter was projected to evaluate gamma, intermediate (Albedo) and fast neutron doses from ^{241}Am-Be sources.

High doses and accident dosimetry

In order to standardize a method for retrospective dose determination, the ESR response of tooth enamel irradiated with low energy X-rays and ^{60}Co gamma radiation was evaluated and compared with the ESR response of synthetic hidroxyapatite in the dose range between 0.2 - 10 Gy. Practical considerations of tooth selection, sample preparation and ESR spectrometer parameters settings were studied. The main dosimetric properties of Brazilian silicates of the jade family were studied for application in high dose dosimetry: tremolite, actinolite, rhodonite and diopside, using the thermoluminescent, thermally stimulated exo-emission and electronic paramagnetic resonance techniques. The results obtained showed the possibility of their use as irradiation detectors and for high dose dosimetry.

Metrology in diagnostic radiology

Computation systems were applied to simulate new ceramic compounds suitable to improve the radiodiagnosthic room shielding. Different ceramic compounds were formulated theoretically using SiO_2, Fe_2O_3, Al_2O_3, BaO, PbO e Ti_2O_3 compounds that were mixed in different concentrations and the shielding properties were simulated to choose the sample with the best shielding properties. Ceramic plates were produced and attenuation properties were evaluated and compared with the theoretical results.

A new X radiation system (160 kV) was characterized and the qualities from the IEC 61267 were established. As a part of its characterization, the spectrometry of this system was realized in order to verify the real kVp values. This system has been used to calibrate diagnostic radiology instruments. Since then, 490 instruments were tested. A special method to calibrate the CT ionization chambers was developed. A control and data acquisition program for the calibration of instruments for diagnostic radiology was established using the Labview software. The practical peak voltage quantity for diagnostic radiology clinical equipments was evaluated and a quality control program to be applied in dental diagnostic radiology systems was finalized. A methodology for correlations between doses and detectability in standard mammographic images was established. This methodology was applied in São Paulo State, with measurements performed in 50 health establishments using mammography units. Protective devices of five manufacturers were evaluated according to the international standards NBR/IEC 61331-1 and NBR/IEC 61331-3. Three different methodologies were applied for the determination of the attenuation equivalent. The discrepancy between the results obtained and the requirements of the standards show the need to adopt a compulsory certification process for protective devices, thus contributing for the increase of the radioprotection of users. An intercomparison program of radionuclide calibrators for nuclear medicine was established and performed for 67-Ga, 201-Tl and 99m-Tc sources at the Center for Radiopharmacy/IPEN.

Metrology in radiotherapy

The primary purpose of radiation therapy is to deliver sufficiently high dose to the tumor cells, maintaining at the same time the dose to surrounding normal tissues as low as possible. In this way, the Medical Physics Group at IPEN develops research projects in conjunction with some hospitals (A C Camargo, Clinicas, Albert Einstein, Syrian-Lebanese Hospitals and Pediatric Center Boldrini) in order to establish better procedures of clinical measurements for the new technologies in Radiotherapy, such as intensity modulated radiation therapy, radiosurgery and quality control on computerized treatment planning systems. As a result, new quality assurance programs will be implemented at these hospitals. The margins required to define a planning target volume (PTV) for adequate treatment of the mobile tumors such as prostate or those located in areas with less mobility as the ones in head and neck region, in the absence

of daily localization imaging based was studied. The impact caused by the PTV, in terms of dose, to the critical structures surrounding the PTV and its influence when inverse planning is used in the intensity-modulated radiation therapy (IMRT) was also evaluated. The results showed the importance of putting margins in the clinical target volume to assure an adequate treatment, and it was also showed that isocenter daily variation can cause an increase to the dose greater than the tolerance level to the critical organs. A DL-Alanine, Fe-II based gel dosimeter to be applied in Co-60 gamma-radiation fields in the dose range of radiation therapy procedures was developed. The dosimetric gel showed a linear behavior of the spectrophotometric response in the dose range between 0.5 and 40 Gy. The present DL-Alanine, Fe-II based dosimetric gel, presents an excellent potential for the determination of the dose fields in radiation therapy and its future application as a standard in the determination of these fields, in 3D, using the Magnetic Resonance Imaging technique (Fig. 10).

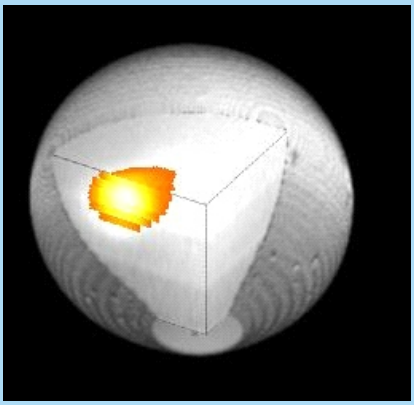


Figure 10. Head phantom image obtained with the magnetic resonance technique and irradiated using gamma knife system

Microdosimetry

Microdosimetric measurements were performed using a Wall-less Tissue Equivalent Proportional Counter - TEPC with spherical cavity. The fast neutron beam from a nuclear reactor was filtered using the heavily filtered fast neutron irradiation system (FNIS) to obtain a decrease of neutron radiation contamination by gamma rays. To obtain the neutron microdosimetric spectra, as the frequency distribution of lineal energy, the dose distribution of lineal energy with good precision, and another quantities as frequency-mean of lineal energy, dose- mean of lineal energy, absorbed dose, equivalent dose and average quality factor of fast neutron were evaluated. The obtained results were satisfactory, with the neutron microdosimetric spectra showing a gamma ray contamination under 5%, especially for dose distribution of lineal energy.

Products and Services

Determination of radionuclides in environmental samples

Radioactivity is measured on a routine basis, by using alpha and gamma spectrometry, gross alpha and beta counting and neutron activation analysis, in order to determine the contents of artificial and natural radionuclides in environmental samples. The following analyses are available: Determination of gross alpha and beta activities; ^{241}Am source leakage tests; Determination of uranium, thorium and radium isotopes; Determination of ^{210}Pb, ^{210}Po and ^{222}Rn; Determination of potassium-40, cesium-134 and cesium-137 concentrations in foodstuffs and food commodities imported and exported by Brazil.

Internal dosimetry

The whole-body and thyroid measurements are routinely performed on IPEN workers, visitors, trainees, and contract workers. During 2005-2007, the Laboratory carried out occupational monitoring of workers involved in radioactive waste management, radioisotope production, research, students and visitors. In this period 1.453 of thyroid and 1.496 whole body measurements were carried out. Routinely the analyses of biological samples from people occupationally exposed to radionuclides with risk of internal contamination are performed. The most frequently requested radionuclides, and the total number of analyses during the period 2005-2007 were: U-nat., 635; U (isotopes),103; ^{232}Th, 83; ^{3}H, 87; γ emitters, 27; Actinides, 46. In addition to bioassay measurements, the Laboratory participated in national (Brazilian Intercomparison Programs) and in international intercomparison (Promotion du Controle de Qualite des Analyses de Biologie Medicale en Radiotoxicologie) programs. The dose calculation follows the measurements of the activity in excreta or in body tissues. These calculations are based on the mathematical models recommended by the International Commission on Radiological Protection, and adopted by the Brazilian Nuclear Energy Commission, according to the type of the radionuclide and the practice. During the period 2005-2007, a total of 2972 evaluations were performed for dose assessment, as part of the internal monitoring program. No worker received dose higher than the annual limit on intake.

Dosimetric pellets of $CaSO_4$:Dy

The Dosimetric Materials Laboratory has produced the $CaSO_4$:Dy crystals and dosimetric pellets during many years. The pellets are destined mainly to the personal, area and environmental monitoring. Furthermore, they are applied to high dose, accidental and medical dosimetry. During 2005 to 2007 about 15000 pellets were produced to fulfill the solicitations from many institutions.

External dosimetry
Around 1500 users are individually monitored monthly for external exposure, using trunk and extremity thermoluminescence dosimeters. There are 100 locations of environmental thermoluminescence dosimetry and 278 locations of workplace monitoring. The routine individual monitoring service for external exposure satisfies completely the regulatory norms of CNEN (Brazilian Nuclear Energy Commission). During 2005 to 2007 12,506 TLD evaluations for different applications were performed.

Calibration of Instruments
Many types of instruments were calibrated: several kinds of ionization chambers, pen dosemeters, survey meters (including superficial contamination detectors), alarm dosemeters, activimeters, clinical dosemeters, and others. The distribution, in terms of calibration levels was: 3% for radiotherapy, 8% for diagnostic radiology and 89% for radiation protection level. In this period, 6599 instruments were calibrated: 14% from IPEN, 20% from hospitals and clinics and 66% from industries. Besides this service, 475 samples including thermoluminescent dosimeters, alanine and other samples were irradiated, with beta, gamma and X radiations for dose-response curves.

Radioprotection Service

The main task of the Radioprotection Service of IPEN is to provide IPEN workers and general population with adequate protection against ionizing radiation. The Radioprotection Service implements adequate procedures and monitoring techniques according to the national and international regulations and standards.
The Radioprotection Service delivers professional radiation protection advice to departments which are involved in the use of radioactive material or ionizing radiation.
The group helps radiation employers to comply on their statutory obligations by providing Radiation Protection Advisers who may be integrated into existing project teams to ensure that all work, whether it be small movements of radioactive materials or large complex engineering operations, is carried out within a robust system of radiological protection.
The services include advice on compliance on statutory requirements for work with ionizing radiations, and they include:

- Preparation of local rules;
- Designation of radiological areas;
- Control and accounting of radiological material;
- Restriction of exposure;
- Dosimetry (internal and external);
- Occupational and environmental radiation and contamination monitoring;
- Dose assessment;
- Contingency planning and radiological risk assessment;
- Statutory training in all areas of radiological protection.

When required, the Radioprotection Service can provide the following services: assistance in the preparation of radiological protection aspects of safety documents; review of radiological protection aspects of safety documents; advice and assistance on radiological aspects of categorization of plant and plant modifications; participation in safety audits; support to engineering projects; analysis of transport packages and waste contents, including assistance with waste characterization; investigation of abnormal dosimetry results; routine reports on personal dose statistics; provision of appropriate radiological information for reports; personal protective equipment including respiratory protection; emergency and incident support and investigation (including advice to plant operations staff and site emergency services outside normal working hours); and response to national emergency response.
The Radiation Protection Advisers can provide a comprehensive support to all operations and facilities; as commissioning, and modifications to decontamination and decommissioning. The Radioprotection Service is available to the customers 24 hours a day, 365 days a year.

During 2005-2007, the IPEN/ Radioprotection Service cooperation in the field of emergency situations in Brazil has been carried out according to the expected work program.
Concerning the program for the improvement of infrastructures at IPEN, the Radioprotection Service is managing the activities of radiological survey of access areas under the direction and instructions of Radioprotection Service staff.
Finally, the Radioprotection Service is updating in a continuous way its procedures in order to fulfill the new legal requirements derived from the regulations.

Nuclear and radiological emergency response

IPEN is an operational unit of the Protection System for the Brazilian Nuclear Program (SIPRON) that is a group of organizations with the objectives of the integrated planning, the combined action and the continuous execution of measures in order to assure the nuclear safety in the country and to respond to radiological and nuclear accidents in Brazil. IPEN also takes part in the implementation of the Emergency Situation plan that was developed by the National Commission of Nuclear Energy (CNEN) to respond to nuclear or radiological emergencies, as loss of radioactive sources and accidents during the transport of radioactive material.
The Nuclear and Radiological Emergency Response Team (NRERT) of the Radioprotection Service is responsible for the evaluation and first response to situations of nuclear or radiological emergencies in São Paulo state. NRERT works with other federal, state and local agencies to monitor, contain, and clean up the release of radioactive material while protecting people and the environment from harmful exposure to radiation. All the thirty-five notifications of reportable incidents occurred during the period 2005-2007 were cleared up.
Other important points to consider were the investments in infrastructure, as new facilities and radiation monitors for the emergency response, and the review of the Radiological Emergency Plan of IPEN.

Figure 1. Radiological emergency response

Training in radiation protection at IPEN

The Radioprotection Service is responsible for the development and implementation of training in radiation protection for a range of users and applications of ionizing radiation. This service has been established to satisfy: the training requirements for IPEN workers for various levels; to emergency response personnel, such us fire fighters, civil defense personnel; and to provide and disseminate information in radiation protection education for students and community.
Workers who are occupationally exposed to ionizing radiation need more extensive and deeper training to ensure that radiation is used safely, than those who have a low potential exposure as a result of occasional activities in areas where exposure may occur. The training of the principles of radiation protection are based on the recommendations of the CNEN (National Commission in Nuclear Energy) and IAEA (International Atomic Energy Agency).
The competences are acquired, developed and maintained through a programme of regular training. The courses are offered, such as basic training, refresher training and on the job training. The content and level of the courses offered are established for each category of persons to be trained.
Training in radiation protection is provided by classroom based training, and on the job training, and the courses provide the theoretical background necessary for the radiological protection requirements for ionizing radiations.
In the last three years, the courses were offered periodically. Upon completion of the course, an examination will be given to authenticate completion of the program requirements. A Certificate of Achievement was presented to those who have successfully completed the course, and a permanent record of training completion will be on file in the Radiation Protection office.
The basic course covers the general principles of occupational radiation protection in the following subject areas: basic radiation physics, definitions and units of radioactivity, principles of radiation protection against external and internal exposures, biological effects of radiation, the risk and assessment of such exposures, instrumentation, inventory and contamination control, emergency response, and the Federal regulations and IPEN procedures. After this basic course all workers are trained in this specific practice in each work area.

Occupational epidemiology

Epidemiology is the study of the distribution and determinants of disease in human populations. Occupational epidemiology involves investigating the frequency of occurrence and causal factors for health effects that have non occupational as well as potential occupational etiologies. The main evidence for the presence or absence of various health outcomes is provided by epidemiological investigations. It is necessary, however, to review carefully and understand the methodological issues and limitations of them in order to properly interpret the results. As background of the evaluation and as starting point, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), presented these studies.
The main objectives of the research group, started in 2006, are:

- To get a solid introduction and a detailed study of the basic epidemiologic methods including the special features of occupational epidemiology;
- To assess the different types of epidemiological study, the applications, advantages, and limitations of the major types of observational and experimental studies, emphasizing the many possibilities for errors in epidemiological for a clear understanding;
- To use epidemiological principles and methods to the practice application (ex., study designs), in particular the radiation epidemiology;
- To determine the possible health consequences of workplace exposures (exposure standard setting) and to recommend remedial efforts when indicated.

Environmental risk assessment

Evaluation of the contamination risk caused by lightning rods disposed at uncontrolled garbage dumps

The Radioactive Management Laboratory (LRR) is conducting experiments to evaluate the contamination risk caused by lightning rods disposed at uncontrolled garbage dumps, using lysimeters as shown in figure 1, filled with waste and radioactive sources in order to evaluate the risk of contamination caused by radioactive lightning rods. The generated leachate is periodically monitored and analyzed to determine its characteristics as pH, Eh, solid content. Microbial growth is also being evaluated using a direct method to count the number of cells in a culture. The equivalent dose to members of the public was calculated considering the ingestion of drinking water, the most probable mode of exposure. The final result was about 145 times below the effective dose limit of 1 $mSv.year^{-1}$ for members of the public, established by the International Commission on Radiological Protection (ICRP), demonstrating that the risk caused by lightning rods disposed at uncontrolled garbage dump is low. Microbial growth may be affected by the radioactive element (Am-241), however more assays have been conducted to elucidate this behavior.

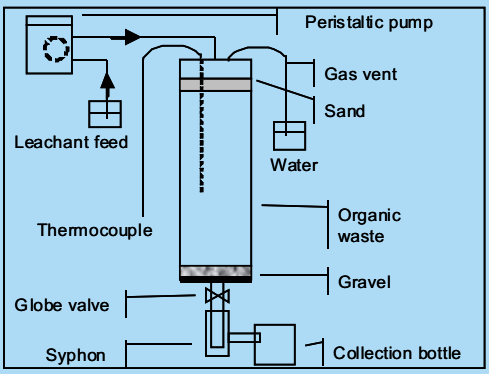


Figure 1. Sketch of the experimental design

A model for determination of screening level for radioactive elements in soil

At the present, decision about clean-up of Brazilian sites contaminated with radioactive isotopes is addressed on a case-by-case basis, since there is no general guidance or recommendation to support actions in early phases of the problem identification. For chemicals, CETESB - the governmental organization responsible for preventing and controlling environmental pollution in São Paulo State - established background values, prevention and intervention, as the first step to implement a remediation actions based on human health risk assessment.

The aim of this study was to develop a methodology for the establishment of target values for radioactive soil contamination, as far as possible consistent and compatible with the approach adopted by CETESB for sites contaminated with chemicals. The following steps have been addressed in this study: conceptual scenario and model development; codification of the equations in an electronic spreadsheet; selection of proper range and statistical distribution of the input values; derivation of the intervention levels for selected radionuclides using Monte Carlo methods. The mathematical model developed was mainly based on the equations used by the U.S. Environmental Protection Agency (EPA) and by the National Council on Radiation Protection and Measurements (NCRP) for soil screening purposes.
Intervention and prevention values are presented for an adult and for a 10 years old child, considering three exposure scenarios: agricultural, residential and industrial; the following radionuclides were considered: ^{3}H, ^{14}C, ^{32}P, ^{35}S, ^{45}Ca, ^{51}Cr, ^{90}Sr, ^{125}I, ^{131}I, ^{134}Cs, ^{137}Cs, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{232}Th, ^{238}U, ^{239}Pu and ^{241}Am. Quality reference values were determined for ^{40}K, ^{137}Cs, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{228}Th, Th-nat e U-nat. Results obtained in this study showed a good agreement with those reported by NCRP, considering that the equations and the input data used in both models are not the same ones.

Radioactive waste characterization, treatment and disposal

Characterization of ion exchange resins and activated charcoal

The Radioactive waste characterization program includes radioanalysis of wastes generated at IPEN and, in some cases, a proposition for their treatment. Our program follows the procedures outlined in CNEN-NN 6.09, about intermediate and low level waste acceptance criteria and some procedures are currently being implemented. The main subjects under development are characterization of ion exchange resins and activated charcoal generated at the research reactor water treatment system. The radiochemical characterization of the ion-exchange resins and activated charcoal wastes revealed the presence of significant amounts of 60Co and 137Cs, out of a list of 20 fission and activation products. The activities of the remaining radionuclides were below detection limits. Forty nine samples were taken from the 21 waste drums and analyzed by gamma spectrometry. As many as five samples were taken from some drums because of the stratification of the waste.

Optimization of the radioactive waste storage

The Radioactive Waste Management Laboratory started a project to apply the concepts of clearance levels and exemption limits to optimize the radioactive waste storage capacity, taking into account that a fraction of these wastes has decayed to a very low level and considering that “retrieval for disposal as very low level radioactive waste” is one of the actions suggested to radioactive waste managers. This study was carried out by determining the doses and costs related to two main options: either to maintain the present situation or to open the packages and segregate the wastes that may be subject to clearance, using the national and three international clearance levels. These doses and costs have been evaluated by using the technique to aid decision making, known as cost-benefit analysis. The results pointed out that to maintain the present situation is not the better option, even though some parameters of each scenario may vary.

Artificial neural network applied to isotopic characterization of radioactive waste drum

In this study, the Monte Carlo method and the artificial neural network approach have been applied for waste isotopic characterization in a 200 L drum. A system with 10 detectors and 5 cross sectioned drum layers was designed, with the detectors positioned parallel to the vertical axis of the drum. Situations where the drum layers are filled with a Co-60 source are created and the responses in each of the detectors are simulated using the MCNP-4C computer code. The Co-60 source can assume 10 relative intensities between 0 and 74 and the detector responses are combined to generate a data file, containing all the possible situations in which the sources can occupy the layers of the drum. The data file is separated in input and output files and then used for training a neural network, where the inputs are the detector responses and the outputs are the position and relative intensity of the sources in the radioactive waste drum. The results of this study show that the neural network, in the training condition, is able to evaluate satisfactorily the activity of the radioactive waste contained in the drum. Nowadays, other radionuclides are under investigation for this methodology.

Treatment and disposal of disused sealed sources

The R&D work undertaken at the LRR in connection with the management of lightning rods, sealed sources and smoke detectors is divided in designing the facilities to handle these sources; developing decontamination process; and developing the concept of a deep, fully-dedicated repository to dispose the sealed sources.

During the reported period, ancillary equipment were designed and fabricated for the hot cell designed to handle the sealed sources (Figure 2), under construction at the LRR. These include a mobile shielded transport overpack for the disposal containers; a hydraulic press stopper fastener to tightly close the disposal container; a workbench capable of holding the source's original shielding during the dismantling work.

For the lightning rods, an investigation on the decontamination process was started aiming at recycling of the metal scrap originated from their disassembling. Figure 2 shows the apparatus for monitoring the scrap.

The contribution of the LRR in final disposal of sealed sources is the development of a concept of repository that is technically and economically feasible for developing countries facing the problem of securing large inventories of disused sealed sources. An investigation work was started in this period aiming at the estimation of the durability of cementitious material under the deep borehole repository environment. This research is partially founded by AEA under a research contract that is part of the IAEA's Coordinated Research Project "Behavior of cementitious Materials in Multipurpose Packaging for Transportation, Long Term Storage and Disposal".

Figure 2. Apparatus for monitoring lightning rod plates

Use of *Saccharomyces cerevisae* in radioactive waste treatment

Waste management plays an important role in reducing the volume of radioactive waste streams, minimizing the cost of the final disposal and the impact on the environment. In this context, new research should focus on the development of simpler and cheaper techniques which may improve the waste processing. The use of biomass in processes concerned with the removal of heavy metals

and radionuclides offers significant potential in the treatment of waste-liquid streams. The aim of this work is to study the potential of the *S. cerevisae*, immobilized in bentonite, in removing Americium-241 from radioactive liquid streams occurring at the Radioactive Waste Laboratory of Nuclear and Energy Research Institute, IPEN - CNEN/SP. Parameters such as pH, biomass concentration and equilibrium time were evaluated. Preliminary results indicated that this technique may be an effective technique for the treatment of radioactive waste liquid contaminated with Am-241.

Degradation of radioactive organic waste using consortium and bacteria

The Radioactive Waste Management Laboratory is responsible for the treatment and storage of many radioactive waste streams, generated by the own Institute and other external radioisotopes users. The Laboratory receives liquid wastes, and sometimes they are constituted by a mixture of aqueous and organic phases. As they should not be directly immobilized in cement and also the small volume stored which does not justify the investment in an evaporator, the Laboratory is searching for a cheaper alternative and simpler methods of treatment. The aim of this work is to develop a technique to degrade organic compounds by using consortium and bacteria. Nowadays, liquid wastes were partially analyzed, and preliminary results revealed that the main organic compound is the tributyl phosphate; and for volatile organic compound it is the ethanol. The next step will be to test the degradation potential of the microorganisms in contact with the wastes.

Products and services

The Radioactive Waste Management Laboratory is responsible for reception, treatment and temporary storage of the radioactive wastes generated at IPEN, as well as those generated at many other radioactive facilities all over the country. The main features of the Laboratory include basically units for: waste reception and segregation; decontamination of small pieces; liquid waste immobilization and conditioning; in-drum compaction of compressible solids; spent sealed sources and lightning rods disassembling; primary and final waste characterization; storage of untreated and treated wastes.

Safeguards Service of the IPEN

The Safeguards Service - SS at IPEN acts in collaboration with several operators from different Materials Balances Areas (MBAs/IPEN).

It makes a annual planning schedule and pre-inventory procedures to accomplish the Physical Inventory Taking (PIT), check the Design Information Questionnaire (DIQ) and carry out Physical Inventory Verification (PIV) in order to realize the inspections of the Brazilian National Authority (National Nuclear Energy Commission / CNEN), Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) and International Atomic Energy Agency (IAEA) that complies with Quadripartite Safeguards Agreement (INFCIRC 435) in 2005, 2006 and 2007. This agreement was signed between Argentine, Brazil, ABACC and IAEA. This agreement adopted the Common System of Accounting for and Control of Nuclear Materials SCCC for Brazil/Argentine.

In 2007 the Safeguard Service carried out and followed the transfer fuel elements from IPEN Nuclear Research Reactor Facility to the United States, with CNEN and ABACC inspectors.

During the development of the Brazilian Nuclear Fuel Cycle in the 80´s, IPEN acquired from INB (Indústrias Nucleares Brasileiras) Uranium as U_3O_8, to be used as a raw material in the Nuclear Fuel Cycle Facilities.

The refund of this material was conducted and managed by the Technical Advisory (SAT) and the Safeguards Service (SS) of IPEN. They managed and executed the strategic planning, the coordination and supervision of the tasks.

This material was re-packed and distributed in several chemical compounds. They were standardized to attend the international standards.

During the tasks several measurements were performed such as air monitoring, dose rate etc and none of them presented values above the standards limits adopted by CNEN. A certain amount of it was sent to Canada to be processed into UF_6 and the remainder is expecting authorization from CNEN/RJ to be sent to Caetite Facilities in Bahia, Brazil.

Figure 1. Re-package of nuclear material

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"Analysis of serum and whole blood using NAA and EDXRF techniques for clinical investigation" - 12th Modern Trends in Activation Analysis - 2007.

"Citotoxicidade de ligas metálicas utilizadas como biomateriais" - LATINCORR 2006.