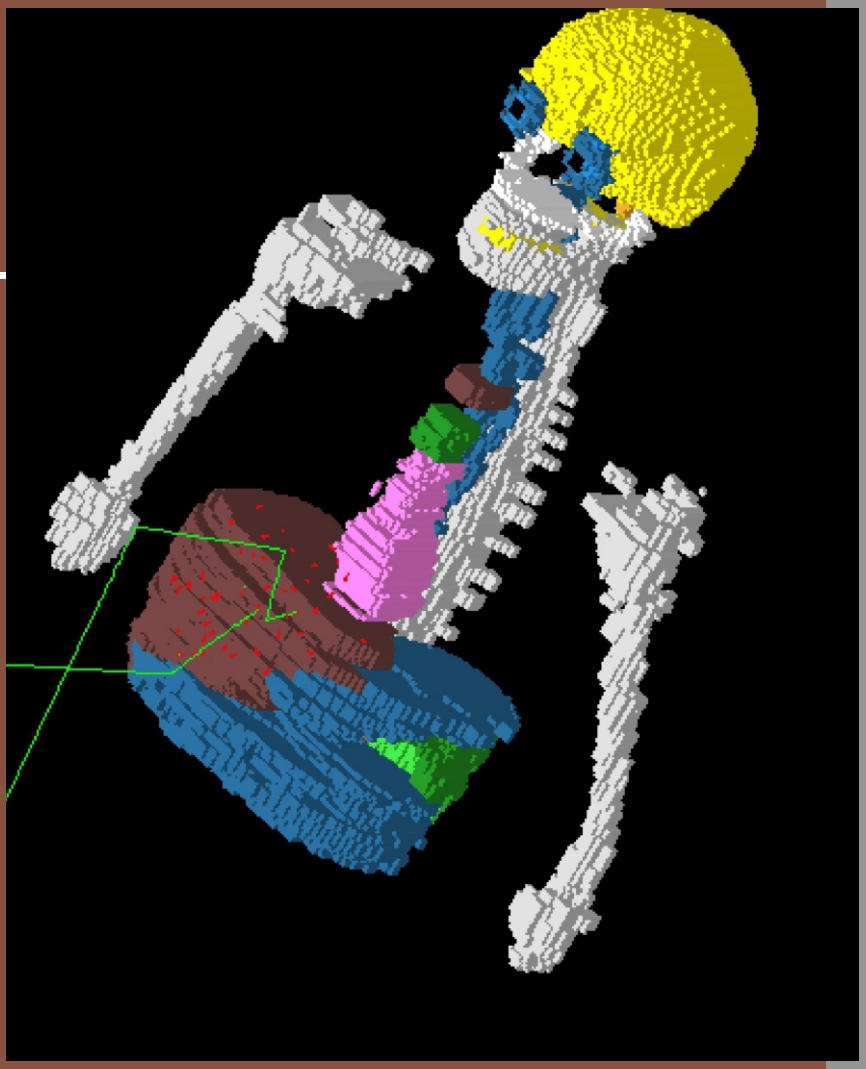
Nuclear Science and Technology



Part of the antropomorphic phantom MAX06 (R. Kramer et al.) with activated glass microspheres inside the liver as simulated with the GEANT4 Monte Carlo toolkit

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 computing 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 ^{99}Mo and ^{131}I, among several others, used in nuclear medicine, by operating the reactor at 5 MW on a schedule of 120 hours/week continuous operation. During the period between 2008 and 2010 the power has been raised from 2.0 to 3.5 MW and it is planned to go up to 4.5 MW during 2011.

At the Radiation Metrology Center, 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.

Study of the crystalline and magnetic structures of materials by neutron and X-ray 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, inside and outside IPEN. The neutron diffraction patterns are measured in the high-resolution neutron powder diffractometer 'Aurora' installed on the IEA-R1 research reactor at IPEN-CNEN/SP. Although the IEA-R1 is a low-flux reactor, installation of a position sensitive detector (PSD) allows a quite fast measurement of patterns with good resolution. A double-bent silicon monochromator installed in the diffractometer permits measurements with four different wavelengths, namely 1.111, 1.399, 1.667 and 2.191 Å (nominal values). Figure 1 is a photograph of the high-resolution neutron powder diffractometer 'Aurora' installed on the IEA-R1 research reactor.

Figure 1. The neutron powder diffractometer 'Aurora'

Presently, only room-temperature patterns can be measured. In near future, a helium cryostat will be installed allowing measurements close to 4 K and above. 'Aurora' has been designed mainly for crystalline and magnetic structures determination and for application of the Rietveld method in quantitative phase analysis though 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 which are produced by neutron irradiation in the IEA-R1 research reactor of IPEN, 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(^{6}Li,3n)^{111}In nuclear reaction was developed and it is also available. The compounds which have been investigated are:

1) 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 $^{181}Hf \rightarrow {}^{181}Ta$ nuclear probes which were introduced into the samples through a chemical process. The quadrupole interaction parameters as well as the magnetic hyperfine field were obtained for each compound.

2) Diluted Magnetic semiconductors: new families of semiconductors, which are doped with magnetic materials in order to use the electron spin information , are under intensive investigation as they can be used for spintronics. ZnO, SnO_2, TiO_2 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.

3) Insulator oxides with large bandgap as HfO_2 and CeO_2 are promising materials to replace SiO_2 as a gate dielectric to prevent leakage current in complementary metal oxide semiconductor (CMOS) transistors. Thin films and nano-structured powders of these materials are under investigation using PAC spectroscopy in order to obtain an atomic scale characterization of their properties under different temperatures.

4) 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.

5) PAC spectroscopy is also being used to investigate biomolecules of EDTA and DNA molecules of different mouse lineages (A/J, C57BL/6, B6AF1, BXA1 and BXA2) infected by the strain Y of *T. cruzi*. This parasite may cause the Chagas disease when transmitted to humans. The main objective of the present work is to investigate the neighborhood of the sites to which the ^{111}In-^{111}Cd probes are bond in the DNA molecules of the different mouse lineages by measuring nuclear quadrupole interactions in order to compare them and establish which nitrogenated base the probe are bonded to.

6) 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 WIEN2k 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 and oxides such as HfO_2 and ZnO are also being carried out.

Applied Nuclear Physics, Instrumentation and Scientific Computing

The Group of Applied Nuclear Physics, formed in 2008, performs research mainly in the areas related to development of instrumentation and methods for radiation measurements, scientific computing, measurements and analysis of nuclear data, and application of nuclear techniques in several areas.

Instrumentation

The recent technology of silicon photomultipliers brought new perspectives for compact gamma ray detectors, which can be used in X and gamma ray tomography techniques. New concepts of position sensitive detectors have been studied with the aim of reaching spatial resolution that are adequate to build a positron emission tomography equipment. These studies involve Monte Carlo simulations in parallel with experiments, in order to obtain an instrument with optimized characteristics. A fully digital multiparametric system will be used in the signal acquisition and processing of the several tomograph detectors. Also related to instrument development, a multipurpose ionization chamber was developed in collaboration with the IEE-USP. This chamber has several applications in measurements of scattered radiation from medical equipments. Moreover, a module was developed for checking the gamma-ray coincidences using the pre-amplifier signal as input to quantify the ^{18}F retained in the chimney filters during the FDG production.

Scientific Computing

Computing is a tool as important as experimentation and theory in solving the scientific challenges in the nuclear science of the twenty-first century. Our group has been working with Monte Carlo simulation softwares, mainly Geant4, applied to dosimetry, medical instrumentation and detector studies. We also developed a scientific software applied to Instrumental Neutron Activation Analysis; the software is currently undergoing a complete re-engineering to include k_0 Neutron Activation Analysis methodology. To accomplish this goal, energy efficiency calibration is being incorporated as well as all the other calculations necessary to obtain element concentrations via k_0 technique. The programming language of choice for this software was Python together with Q_t graphics library for the user interface. Two problems were analyzed with Monte Carlo simulations using the GEANT4 package: studies of dosimetry of ^{32}P activated glass microspheres used in brachytherapy, in collaboration with the Center of Materials Technology - IPEN, and simulations of gamma-ray position-sensitive detectors composed by LYSO scintillators coupled to silicon photomultipliers. A software was developed with the aim to help a nonexperienced user to compute the detection efficiency curve and to transfer the efficiency from the point source geometry to other large source geometries. This was built in the Visual Basic platform, and the efficiency transfer method was done with analytical calculus and with numerical simulations using the Monte Carlo method using the GEANT4 tollkit. It was also developed an empirical method for the efficiency determination for large sources. Studies for the implementation of a computer cluster formed by around hundred cores and some GPU units were carried on, and funds were obtained for this implementation in the next years.

Nuclear Data
Nuclear applications often require a good degree of knowledge on several parameters of the nuclei involved, both regarding the safety of the experiment and the reliability of the results. In this sense, the half lives of several radionuclides produced by neutron irradiation (^{193}Os, ^{155}Sm, ^{52}V, ^{101}Mo, ^{101}Tc, ^{127}Te) have been determined with better precision than found in the literature. Also, nuclear reaction cross sections and nuclear structure have been studied using the Pelletron heavy-ion accelerator of USP. Also, parameters related to the k_0 NAA method were studied using a rigorous statistical treatment of the uncertainties involved in the process using both covariance matrices and the least squares method. The aim of this work is to contribute to an enhancement of the international database of k_0 parameters, as well as to reduce the discrepancies found in it. These include the detection-related parameters (detection efficiency and gamma spectrum analysis), the neutron field characterization parameters (α and f), as well as the nuclide-specific ones (k_0 and Q_0).

Methods for Nuclear Radiation Measurements
With the purpose of improving experimental results on nuclear spectroscopy, a thorough analysis of nuclear peak-fitting software has been performed, as well as the development of a method to determine precise gamma transition intensity values from gamma-gamma coincidence and angular correlation experiments. On a different front, viability studies were performed aiming towards the implementation of a PGAA (Prompt Gamma Activation Analysis) facility in the IEA-R1 reactor. This technique is complementary to the regular NAA method and has several applications in the industrial, medical and environmental fields, for instance.

Neutron imaging techniques

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 to inspect radioactive objects. 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 can be obtained in real-time. For both cases the radiographs are 2-D projections of the internal structure of the object. The neutron imaging 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 micra range were also developed. The IPEN posses also a digital system to process images with which has provided services and developed high level researches. In this period (1988-2007) four MSc and three PhD thesis have been advised. The figures 2 and 3 show examples of some typical images obtained in the present facility. Between 2009 and 2010, the working group has installed in the same facility a neutron tomography equipment able to provide 3D images and 3D digital films of the internal structure of the same objects. The figure 4 is an example of 3D images obtained by using this equipment. As has occurred in other countries, the availability of a neutron tomography equipment will spread the use of the neutron imaging techniques in Brazil since this technique reaches the industrial, technological and research fields not reached by the conventional radiography techniques.

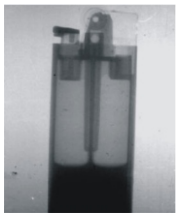


Figure 2. Lighter

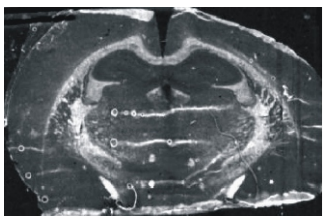


Figure 3. Biological tissue (brain)

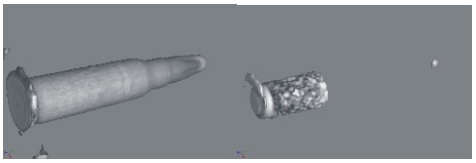


Figure 4. 3D image. Gun bullet (left); powder in detail inside the metallic housing (right)

Radiation spectroscopy and spectrometry radioactive decay

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

Nuclear Data
Using single beta and gamma spectroscopy coupled to coincidence and angular correlation analyses, the group have been measuring nuclear data on nuclei

produced via neutron irradiation in the IEA-R1 reactor, as gamma and beta transition energies, intensities and electromagnetic nature, beta feeding for the excited levels and measuring of thermal neutron cross activation sections and half live of beta decay.

Nuclear Instrumentation and Methodology
The group has 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. Also, preparation of texts for teaching physics as well as didactic proposals involving gamma spectroscopy measurements for high school students.

Gamma Spectrometry Applied to Health and Environment Areas
For clinical analysis biological materials in humans and animal models and obtain reference values for use in diagnosis of different pathologies. These data are relevant to both veterinary medicine and to public health areas.

Highlights 2008-2010
- Analysis of the decay of ^{127}Te isotope using both singles and gamma-gamma coincidence for providing literature information and text nuclear models.
- Beta-decay lifetimes for ^{28}Al, ^{52}V, ^{24}Na, ^{42}K, ^{56}Mn, ^{101}Tc, ^{101}Mo, ^{127}Te, ^{140}La, ^{155}Sm and ^{193}Os (LEER, Pelletron Laboratory/IFUSP and FEI/SP).
- Development and application of software for gamma spectroscopy analyses for high school students: applied at Escola Estadual Professora Maria Aparecida Nigro Gava, São Paulo city.
- The thermal neutron cross section of ^{48}Ca and ^{81}Br using the ko-factors in NAA (LEER and UNESP/ Botucatu).
- Development of methods and software's to extract precise gamma transition properties from both beta and gamma- gamma coincidence and angular correlation data as well as for neutron flux determination (LEER, CDTN and Pelletron/IFUSP).
- Determination of reference values for humans on whole blood and serum for several elements of clinical relevance (Br, Ca, Cl, Fe, K, Mg, Na and Zn) using NAA and EDXRF techniques (LEER and CQMA at IPEN and UNISA).
- Analysis in whole blood of athletes using NAA (LEER and LABEX /UNICAMP).
- Characterization of ions in biological materials (serum, blood, urine and organs) of wistar with Acute Renal Insufficiency using gamma spectrometry (LEER and UNIFESP).
- Analysis of elements in human blood of patients with chronic kidney disease using neutron activation analysis (LEER and CRCN).
- Reference values in blood elements in crioulo breed horses, white rabbits, mice of several species and Golden hamster using NAA and EDXRF techniques (LEER and Butantan Institute).
- Reference values in blood for Elemental analyses mice (NZB , SJL , A/J, I_{max}, H_{III}, BALB/$_c$, C57BL, L_{III} , I_{min}, SJL, B_{10} and AJ) using NAA (LEER and CBT at IPEN and Instituto Butantan).
- Quantitative evaluation of blood elements by NAA in mice immunized with snake venoms: Bothrops jararaca, B. jararacussu, B. alternatus, B. moogeni and B. neuwiedi and a also venom mixture with B.atrox from Rio Negro from Amazonia (LEER and Butantan Institute).
- Plant Nutrient Distribution Analysis using NAA (LEER, FEI and IFUSP).
- Potassium incorporation in fruits of South American tropical species (LEER and LARA/UFF).
- Multielemental Nuclear Analysis of soil reference material (LEER and CDTN).
- Quality control of hyperimmune sera by Chromatography, Instrumental and k_o standardization techniques (LEER, CDTN and Butantan Institute).
- Multielemental analysis of extracts of Achornea Gandulosa, Davilla Elliptica and Davilla Nitida using TXRF and NAA techniques (LEER and UNESP/Botucatu).
- Mineral characterization of the ration managed in the diet of equines used in the antivenom production (LEER and Butantan Institute).

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. The disintegration rate is obtained by the application of the efficiency extrapolation technique. During the period of 2008-2010, the following radionuclides have been standardized by this technique: ^{51}Cr, ^{57}Co, ^{111}In, ^{177}Lu, and ^{198}Au. The Nuclear Metrology Laboratory participated in the international comparison of ^{177}Lu standardization sponsored by BIPM in 2009. The Nuclear Metrology Laboratory at IPEN has also been involved in the determination of X-ray and gamma ray emission probabilities per decay of ^{51}Cr, ^{177}Lu, ^{198}Au and ^{241}Am. The measurements were carried out by means of HPGe planar and REGe spectrometers, both with a Be window. The development of radioactive water-equivalent solid sources prepared from an aqueous solution of acrylamide have been continued, by using bisacrylamide and ammonium persulphate for its polymerization. The sources have been prepared in cylindrical geometry with ^{133}Ba radioactive solutions with density similar to water as well as good uniformity. A new data acquisition electronic system for 4πβ-γ coincidence

measurements is under development which allows simultaneous recording of pulse height and time of occurrence for each nuclear event. Measurements of ^{60}Co have been performed and compared to the conventional system. 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 $4\pi\beta$-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. During the period of 2008-2010 this methodology has been successfully applied to achieve primary standardizations of ^{57}Co, ^{22}Na, ^{111}In, ^{177}Lu, and ^{198}Au. 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 determination of the cross section and resonance integral for the ^{165}Ho (n,γ)^{166}Ho nuclear reaction has been completed.

Nutritional Studies In Foodstuffs And Diets

Food safety is a major public concern worldwide. During the last decades, the increasing demand for food safety has been stimulating research regarding the risk associated with consumption of foodstuffs contaminated by pesticides, toxic elements and/or toxins. The necessity of healthy and good quality diets requires the ability to detect the presence of possible contaminants, as well as, nutritional composition of the diets. In terms of health and nutritional safety, to know the levels of nutrients and/or toxic elements consumed by the population through foodstuffs has become of great importance. Neutron Activation Analysis, NAA, has become an important and useful research tool due to the methodology's advantages. These include high accuracy, small quantities of samples and no chemical treatment has been successfully used on a regularly basis in several areas of nutrition and foodstuffs.

Studies carried out in the period 2008-2010:

A Brazilian Total Diet Study: evaluation of essential elements

Total Diet Study (TDS) entailing the analysis of a Market Basket (MB) means the approach adopted worldwide in estimating the daily intakes of analytes of interest by chemical analysis, for a large-scale population over a specific period of time. The MB consists of foods reflecting a defined total diet, based on amounts of food consumed, provided by representative national surveys. The MB involves preparing food in the manner in which they would be usually consumed (table-ready). This approach has been encouraged by the World Health Organization (WHO) due to the fact that dietary habits vary in every country. The MB has been the most adequate method to assess the dietary intakes of nutrients. Although there are many studies in several countries concerning Total Diet Studies, in Brazil they are still scarce. For this study, the methodology for the first Brazilian TDS for the São Paulo State population and its respective MB was developed. This MB corresponds to 72% of the daily food intake for São Paulo state population. This current study involved essential steps to establish a TDS: 1) information about food consumption obtained from the National Household Food Budget Survey "POF 2002-2003" conducted by the Brazilian Institute for Geography and Statistics from July 2002 to June 2003; 2) development of a Market Basket including 71 foods which belong to 30 food groups previously defined. The selection criteria were the foods consumed more than 2g/day/person; 3) sampling in restaurants of the University of São Paulo; 4) kitchen preparation of foods: discarding inedible portions (bone, fruit peels, etc), preparing ready-to-consume foods, individually and mixing foods of the same food group in proportions based on available consumption data. The weights of raw food consumption data were corrected for edible portions and for the ready-to-consume foods. The Instrumental Neutron Activation Analysis methodology was successfully applied to estimate the concentrations of Na, K, Ca, Fe, Zn and Cr in food groups that compose the MB. The contribution of each food group to the total daily intake of these elements was also calculated.

Essential elements in pre-term and term human colostrum

Deficiency of minor and trace elements can lead to various disorders in the early stages of child development. Trace element requirements, during early childhood are more critical due to faster growth rates. Human milk is recommended as the only source of nutrients for infants up to 6 months. In this study, human colostrum samples from two groups of newborns according to their gestational age were studied: a pre-term group and a term group. Samples were collected by manual expression from the first to the fifth day after birth. After sampling, human milk samples were frozen and freeze-dried until the analyses. In this study, Ca, Cl, Fe, K, Mg, Mn, Na, Se and Zn were determined in 15 pre-term colostrum samples and 15 colostrum term milk samples. The methodology applied was the Instrumental Neutron Activation Analysis (INAA).

Essential elements in cow milk and soy-based infant formulas

An infant formula is designed to supply nutrient requirements of neonates during the first months of life. 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. Numerous infant products have been produced and formulated to meet the nutritional needs of healthy full-term infants. Due to variation in nutrient contents from the food sources used to the formula preparations, specifications of nutrient levels, including mineral elements, have been set to simulate levels of these nutrients similar to human milk. Some commercial infant formulas are deliberately fortified with essential elements such as iron, zinc and copper to ensure that they provide infants nutritional requirements for trace elements. There are three major types of infant formula: cow milk, soy-based and protein hydrolyzed formulas. Instrumental Neutron Activation Analysis (INAA) was applied to quantify the essential elements Ca, Fe, K, Na, Se and Zn in three soy-based formulas and 14 infant formulas based on cow milk, being 2 samples for newborns of high risk and 3 for special requirements.

Toxic and essential element determination in edible mushrooms

Research interest about concentrations of elements in the fruit bodies of especially edible mushrooms has started in the late nineteen sixties and continued until now. Due to its high nutritional value, mushroom cultivation has been a good alternative

to supply protein to countries where the nutritional food value is very low. Due to low sodium concentration in mushrooms, they are also a very good food source for special diets for people with hypertension. This study determined the concentrations of As, Br, Co, Cr, Cs, Fe, K, Na, Rb, Se and Zn present in edible mushrooms acquired from São Paulo city markets or directly from producers of the Mogi das Cruzes, Suzano, Juquitiba and Mirandópolis cities. The obtained results confirm that mushrooms can be considered a good source of K, Fe and Zn. The low Na level is a good nutritional benefit for the consumer.

Determination of toxic elements in octopus samples by INAA

Molluscs of the Class Cephalopoda (squid and octopus) are considered of economic interest as they are a nutritional food source. Octopuses feed up of crustaceans, mollusks, fish and other cephalopods, giving them great potential as bioindicator organisms, providing qualitative information on the contamination of the environment. In Brazil, the lack of data on the sanitary-hygienic conditions makes it essential to carry out studies in this area, in order to ensure the delivery a safe food to consumers. The objective of this study is to determine the concentration of the elements arsenic, selenium and zinc in samples of octopus acquired in different points of the production chain. In the period 2005 to 2007, 121 samples of raw octopus were obtained in the following locations: free fairs, markets/fishmongers, supermarkets, industry and fishing terminals or warehouses in 4 municipalities (Guaruja, Santos, São Vicente and Praia Grande). The samples were dried, ground and homogenized. The methodology for element determination used was Instrumental Neutron Activation Analysis (INAA). Arsenic levels were above the limit of 1.0 mg kg^{-1} (in natura) for foodstuffs established by Brazilian legislation. Se and Zn concentrations were in accordance with literature values. Statistical analyses showed no difference among samples considering “in natura” weight, city or season.

Artificial and natural radionuclides in edible and wild mushrooms

Environmental biomonitoring has demonstrated that organisms such as crustaceans, fish and mushrooms are useful to evaluate and monitor both ecosystem contamination and quality. Particularly, some mushroom species have a high capacity to retain radionuclides and toxic elements from the soil and the air. The potential of mushrooms to accumulate radionuclides in their fruit-bodies has been well recognized. The levels of ^{137}Cs varied from 1.4 to 10.6 Bq kg^{-1}, 40K levels varied from 461 to 1,535 Bq kg^{-1}, ^{232}Th levels varied from 6.2 to 54.2 Bq kg^{-1} and ^{238}U levels varied from 14 to 66 Bq kg^{-1}. The ^{137}Cs levels in Brazilian mushrooms are in accordance with the radioactive fallout in the Southern Hemisphere. The artificial and natural activities determined in this study were found to be below the maximum permissible levels as established by national legislation. Thus, these mushroom species can be normally consumed by the population without any apparent risks to human health.

Another investigation of mushroom biological assimilation of radioactive elements belonging to the natural series of uranium and thorium was conducted on the Poços de Caldas plateau, Minas Gerais State, Brazil. This region has naturally high radioactivity, due to significant anomalies of predominant mineralized uranium and thorium. The soils of this region are also enriched with uranium and thorium when compared to the world average concentration. Several studies have been conducted on the incorporation of radionuclides in agricultural products of the Poços de Caldas plateau. The objective of this study is to determine of natural and artificial radionuclides in wild mushroom samples collected from different points of the Poços de Caldas plateau region.

Assessment of the Content of Mercury, Methylmercury and Other Elements of interest in Fish and School Children Hair from Cananéia and Cubatão coastal cities, São Paulo State, Brazil

The Cananéia-Iguape estuarine-lagoon complex, located in the southern coastal region of Sao Paulo State, is part of the Biosphere Natural Reserve (UNESCO) due to its environmental-cultural importance. It is a region of overall low pollution impact in the southern part of the hydrological system (Cananeia estuary) and is an Environmental Protected Area. Located in the proximities of one of the most important industrial complexes in Sao Paulo state and the most important port of South America, the Santos Estuarine system is highly susceptible to human activities from industry, urban sewage and polluted solid waste disposal. Cubatão city, located in this estuary, was considered as one of the most polluted cities in the world in the recent past. The present study assessed total Hg content, micronutrients (Ca, Fe, K, Na, Se and Zn) and macronutrients (proteins, lipids, ash, energy, carbohydrate) in fish most consumed for the population from Cananéia and Cubatão coastal cities. Furthermore, total and MeHg levels were also determined in hair samples of children from both cities in order to verify bioaccumulation of Hg in this populational group. From these results it was possible to evaluate the nutritional content of the fish consumed and the exposure of the children to Hg and MeHg in these coastal cities, Cananeia and Cubatão.

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 project "Toxic metal and trace elements assessment in sediments from Guarapiranga and Rio Grande reservoirs, Metropolitan region of São Paulo," was developed. These reservoirs are important and supply water for a greater part of São Paulo metropolitan region. The contamination of Rio Grande tributary, Billings reservoir, and Guarapiranga reservoir, by determining metal concentration and other elements of interest using three analytical techniques (INAA, AAS and ICP OES) were assessed in bottom sediment samples. The content of total and organic mercury in the sediments from Rio Grande tributary was evaluated. For this purpose an analytical methodology for Hg was developed and validated. The chosen chemical parameters for sediment characterization were Al, As, Ba, Cd, Cu, Cr, Fe, Pb, Mn, Hg, Ni, Se and Zn. 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 as well. 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. Also AAS and ICP OES analytical techniques were applied in order to assess toxic metals in the sediment samples. 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 the study of pollution caused by anthropic discharges. There are two main types of experiments that are generally used for this purpose: *passive biomonitoring*, in which the native organisms are collected and analyzed and *active biomonitoring*, in which organisms from a pristine area (like a mussel farm) are collected and transplanted to polluted sites. In the first phase of this work, the active biomonitoring approach was used and the marine bivalve *Perna perna*, very abundant in the coast of the State of São Paulo, Brazil, was transplanted from a mussel farm 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 (1.0 µg.g-1 for As and 0.30 µg.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 (1.0

µg.g-1 for Cd, 2.0 µg.g-1 for Pb, 0.5 µg.g-1 for Hg and 50 µg.g-1 for Zn). In the second phase of this work, a passive biomonitoring approach is being applied, and two types of organisms have been chosen for the study: the *Perna perna* mussel, collected in a mussel farm at the Cocanha beach (clean area) and in two sites at Santos Bay (Itaipu and Palmas) and the *Crassostrea brasiliana* oyster, collected at Cananéia (oyster farm) and at Bertioga and Santos Estuary. Inorganic trace elements are being determined : As, Br, Co, Cr, Fe, K, Mg, Mn, Zn and V, by INAA and Cd, Pb and Hg by AAS. Also ecotoxicological tests with the neutral red reagent are being made, in order to evaluate the level of stress of the organisms in the polluted regions. In 2009, this work with passive biomonitoring became part of the IAEA/ARCAL Project: "Regional Programme for the biomonitoring of contaminants in molluscs and fish to ensure seafood safety in Latin America and the Caribbean using nuclear analytical techniques" was started and also the analysis of fish consumed by the population of the city of São Paulo were included in the project. Samples of some of the most consumed fish species by the population (robalo, sardinha, pescada, salmão, corvina, tainha and anchova) were acquired at the CEAGESP, the main food distributor of São Paulo and are being processed for analysis of inorganic trace elements, also by the methods of INAA and AAS. The results obtained will be compared to the values of the Brazilian legislation for food contaminants.

Figure 5. Collection of the haemolymph of oysters for the ecotoxicological test

Production and characterization of biological reference materials

A certified reference material (CRM) is a reference material accompanied by a certificate, whose values are certified by a procedure which establishes its traceability to an accurate realization in which the value is expressed and each certified value is accompanied by an uncertainty at a given level of confidence. Certified reference materials are still not widely used in Brazil and other Latin American countries. The main reason is the high cost of these materials, since most of them are imported. Reference materials are important tools in the quality assurance of analytical results as they are used in method validation, calibration of instruments and in the realization of the traceability of analytical results to stated references. The Neutron Activation Laboratory has been involved in the development of Brazilian biological reference materials, as described below:

- As a contribution to the biomonitoring programs carried out by the Institute, all the steps for the production of a Brazilian mussel reference material using the *Perna perna* species were developed. Besides being used in biomonitoring studies, this mussel species has also economical importance due to its cultivation in aquaculture farms for human consumption. The production 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 were performed, with emphasis on the assessment of the stability of the material, its homogeneity status, residual water content and granulometric characterization. An international collaborative interlaboratorial study was performed for assignment of certified values. 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.

Figure 6. *Perna perna* mussel

- The International Atomic Energy Agency (IAEA) has been supporting several projects with the objective of laboratory capability improvement in Latin America. In this context, a new IAEA project ("Improvement of analytical quality through proficiency testing and certification of matrix reference materials using nuclear and related analytical techniques in the Latin American nuclear analytical techniques network"- ARCAL RLA 0214) has started in 2009. In this project, the Neutron Activation Laboratory is responsible for the preparation of a new fish reference material. Whitemouth croake (*Micropogonias furnieri*), also known as *corvina*, was chosen as it is the second fish in production in Brazil and it is widely distributed and consumed in the Latin American countries. For preparation, about 300 kg of fish was collected and only the edible parts were used. After preparation, the fish reference material will be tested for homogeneity and stability and characterized by the ARCAL participating laboratories with experience in this field and possibly by expert laboratories outside Latin America.

Figure 7. *Micropogonias furnieri*, the second fish in production in Brazil

Correlation studies of atmospheric pollution and cardio-respiratory diseases through atmospheric pollutant biomonitoring in São Paulo Metropolitan area

Atmospheric pollution is today one of the many problems facing mankind. This problem affects everything from the natural environment to human health and to climate. As result authorities of all over the world have become very preoccupied with the adverse effects derived from pollution. Health problems due to atmospheric pollution also affect São Paulo city, the capital of the State of São Paulo In order to contribute to effective public polices it is important to study the pollutants, their origins as well as to identify the spatial gradient of air pollution in order to explore its possible association with health effects. In order to evaluate pollution levels in São Paulo Metropolitan region a systematic sampling of *Canoparmelia texana* lichenized fungi species was performed in several subprefectures as well as in a reference region located at the Intervales Park, Atlantic Forest, SP considered clean region. Concentrations of elements were determined by neutron activation analysis. Elements Ca, K, Mg presented concentrations at the percentage levels, Ba, Br, Cd, Cr, Fe, Mn, Na, Rb, V, Zn, La and Ce at the levels of mg kg-1 and the elements As, Co, Cs, Hf, Sb, Sc, Se, Th, Nd, Sm, Eu, Tb, Yb and Lu presented the lowest concentrations at the levels of $\mu g\ kg^{-1}$. The occurrence of Sb can be associated to the emission of plastic material incineration. The elements Cr and Fe also presented quite similar pattern distribution and their origins can be associated to industrial emissions and terrigenous origin, derived from the deposition on lichen of solid particles stirred up by the wind. The elements Zn, Ba and V presented similar distribution in the studied areas and they may be associated to vehicular sources. The origin of Ba can be attributed to the use of diesel as a fuel and V of gasoline. Zn and Mn may be associated to industrial origin and also to brake and tire wear emissions. High concentrations of Ca found in some lichen samples can be associated to cement used in construction of buildings. Besides, high concentration of K was found in lichen from clean region of control site. K is an essential element for lichens and in polluted area this element is present in low level due to the stress caused by pollutants. The mortality rates due to the diseases of circulatory and respiratory systems (ICD I00 to I99 and J0 to J99) for two groups of individuals: children younger than 5 years and adults over 45 years of age and living in the Municipality of São Paulo were obtained from databases maintained by the municipal government on the websites:

http://ww2.prefeitura.sp.gov.br//cgi/deftohtm.exe?secretarias/saude/TABNET/SIM/obito.def
Http://ww2.prefeitura.sp.gov.br/cgi/deftohtm.exe?secretarias/saude/TABNET/POP/pop.def.

Based on slow growth of lichen and its exposure period to pollution we use mortality data for a period of 5 years from 2005 to 2009. The statistical treatment of Pearson's correlation applied to the results of lichen element concentrations and mortality rates indicated significant positive correlation for the elements Co, Mn and Zn for adults. (Partnerships: FMUSP, IBt. Financial Resources Agencies: CNPq and IAEA)

Evaluation of trace elements in human tissues to study the health of the population

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 become 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 2008-2010 we focused in the analyses of brain samples of a normal elderly population since little is still known concerning their element level concentration and progress in understanding the role of elements in nervous system diseases has been hampered due to a lack of data of elements in different compartiments of human brains. Thus we decided to investigate trace elements in the hippocampus and frontal cortex tissues. The hippocampus is a major component of the brain and plays an important role in long term memory. It is also one of the first regions of the brain that suffers damage from Alzheimer´s disease. The frontal cortex is a sheet of neural tissue that plays a key role in memory, attention, perceptual awareness, thought, language, and consciousness. Brain samples of an over 50 year old population (mean age of 77.6 ± 8.7; range, 51-95 years) of both genders were provided by the Brain Bank of the Brazilian Aging Study Group (BBBABSG) of the São Paulo University, Medical School. Severity of cognitive impairment was assessed with the Clinical Dementia Rating scale (CDR). A CDR 0 (zero) indicates no cognitive impairment and CDRs of 0.5, 1, 2 and 3 indicate questionable, mild, moderate and severe dementia, respectively. Brain samples of a group of individuals of CDRs 0 (normal) were collected for this study from the

elderly residing in São Paulo city. Slices of brain tissue were dissected from the hippocampus and frontal cortex. Special care was taken to avoid sample contamination with metals during handling. The dissected brain tissues of each area were then homogenized, freeze-dried and analyzed by neutron activation analysis. Concentrations of Br, Fe, K, Na, Rb, Se and Zn were determined. A comparative study based on two different age groups of individuals carried out for the hippocampus and frontal cortex samples indicated that the element concentrations of hippocampus and frontal tissues from group aged 51 to 75 year group did not present significant differences from those found for the 76 to 95 years. Concentrations of results obtained in the hippocampus and frontal cortex tissues when compared in terms of genders showed that only Zn presented significantly higher concentrations in the hippocampus of males than those presented by the female group in the corresponding brain part. On the other hand, in the case of the frontal cortex no significant difference of element concentrations was verified between the genders. The results showed that the distribution of Fe, Se and Zn in brains of normal individuals is heterogeneous. Since certain cerebral diseases affect only small area of the brain it is important to select a defined area for elemental concentration analysis. Our results encourage us to continue research to obtain additional data, which can be used as base line values for normal brains and also to understand the functional activities of the specific brain region with regards to the elemental concentrations. (Partnerships: FMUSP. Financial Resources Agencies: CNPq)

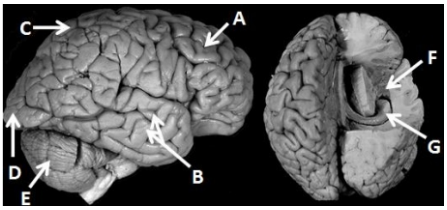


Figure 8. Photo of the brain areas for element composition determination. A. middle frontal gyrus, B. upper and middle frontal gyrus, C. pariental upper lobe, D. occipital lobe, E. cerebellar cortex; F. amygdala and G. Hippocampus

Elemental composition of herbal medicines sold over-the-counter in São Paulo city, Brazil

Medicinal plants have been used to treat diseases for thousands of years. This is true especially in third-world countries where herbal medicine has long played an important role in primary healthcare of the population. In Brazil, the use of herbal medicine is also very popular due to its immense flora, cultural aspects and to the popular belief that herbs, which are of natural origin, are safe and without undesirable side effects. Furthermore, over the last decades, public interest in natural therapies, namely drugs derived from higher plants, mainly the herbal medicines have increased expressively due the high cost of synthetic medications. Besides that they are sold over-the-counter without any prescription. Since the production of medicinal plants in Brazil is increasing and the future of this market is promising, the Regional Council of Medicine of São Paulo State approved the policy of the national medicinal plant drug and herbal medicines. It also created the municipal program for herbal medicine production. In addition, the National Health Surveillance Agency (ANVISA) published a resolution in order to orientate the use of plant based drugs. Within this scenario, it becomes important to evaluate element concentrations in medicinal plant materials since they can be active components or toxic to human health. In this study, neutron activation analysis (NAA) was applied to evaluate the element composition of herbal medicines from the plants (*Ginseng*, *Ginkgo biloba*, *Centella asiatica*, *Mulberry* and *Aloe vera*) from different origins or distributors. The variations of element concentrations obtained in the analyses of herbal medicines from the same plant species can be attributed to the environmental conditions and local soil characteristics where the plant was cultivated, to the part of plant utilized and the age of plant material utilized in its preparations. The high concentrations of Ca and Mg found in these samples can be associated to the absence of collateral effects of herbal medicines. The high concentrations of these two elements prevent stomach lesions. The high concentrations of K found in herbal medicines could be related to the diuretic actions of these products. K is present in natural diuretics, as well as, in drugs used for eliminating phlegm and to invigorate the stomach. In addition, it is known that K salts can regulate body fluids and also participate in cardiac muscle contraction. Toxic elements such as Cd, Cu and Hg were not detected in the samples analyzed and As and Sb were found in some samples but at very low concentrations. On the other hand, herbal medicines contain essential elements such as Ca, Fe, Cr, Co and Zn. Among the herbal medicines analyzed in this study, each capsule of Centella asiatica presented the highest quantities of most elements. The exceptions were found for Na in which the highest quantity of this element was found in Vera aloe. The findings of this study for over-counter herbal medicines are preliminary baseline information about inorganic constituents that can contribute to the understanding of the relationship between elemental contents and their therapeutical effects. (Financial Resources Agencies: CNPq)

Characterization of micronutrients and contaminants in plants grown in contaminated soil

In recent decades, anthropogenic activities, associated with industrial processes and mining, have been the major source of inorganic element enrichment in soils. One example is the need for wastewater treatment technology due to the increase of urban population and industrial development. In such treatment, sewage sludge is generated. This sewage sludge, after a proper microorganism reduction and physical conditioning, is usually known as biosolid. One of the alternatives to the final destination of this waste is the application in agricultural land, as fertilizer. The elements B, Cl, Cu, Fe, Mn, Mo, Ni and Zn are considered micronutrients for plants, but in high concentration in soil solution can reach levels toxic to plants and microorganisms. Other elements such as Ag, As, Be, Cd, Cr, Cu, Pb, Hg, Ni, Sb, Se, Tl and Zn are considered potentially harmful to human health depending on concentrations. Unlike organic contaminants, most inorganic elements do not undergo microbial or chemical degradation therefore their total concentrations remain in soils for a long time after their introduction. Due to the possible presence of these elements at toxic levels to plants, which can reach the food chain, the interest in the development of technologies for remediating contaminated sites has increased. The addition of substances capable of immobilizing toxic elements in the soil is a procedure that has been used for remediating contaminated sites. INAA is an efficient analytical method for monitoring several micronutrients and inorganic contaminants present in geological and biological matrices. This project is being conducted in partnership with CENA/USP. Steps carried out in 2008-2010:

- Determination of Antimony, Arsenic, Cadmium and Thorium in sugar cane juice obtained from cropland treated with sewage sludge. Elemental concentrations for As, Br, Ca, Cd, Co, Cr, Eu, Fe, La, Mg, Mn, Mo, K, Na, Sb, Sc, Sm, Th, Ti, U, V and Zn were determined in biosolid. Analysis of sugar cane juice showed no significant difference in concentration of As, Sb and Th in different doses of biosolid used, with the exception of Cd.

- INAA applied to multielement determination in a variety of lettuce grown in a contaminated soil and treated with phosphate. Super phosphate was used as a substance to immobilize toxic elements in soil. Different doses of super phosphate were added to a number of lettuce plant pots contain contaminated soil. The element concentrations absorbed in the leaves from lettuce treated with phosphate were compared with those absorbed in the leaves of a control plant. The use of 250 mg kg^{-1}(P) proved to be the most effective treatment to reduce the concentrations of Br, Ca, Cd, Cl, Co, Fe, K, Mg, Mn, Sb and Zn in lettuce leaves.

Archaeometric studies in archaeological ceramics using nuclear analytical techniques

The field archaeological chemistry dates to the 1700s, but the Archaeometric Studies Group from IPEN-CNEN/SP involvement began around 1996 with a new research program as a means of characterization of ceramics via INAA.

Studies in ceramics samples can provide information about productions 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. With the elements determined, attention is paid in establishing inter-sample similarity by means of advanced statistical methods like Mahalanobis distance, cluster analysis, principal components analysis, Kernel density, and other as Procrustes analysis, neural network to cite just a few of the statistical methods used.

A typical procedure used in our laboratory consist in cleaning the ceramic's outer surface and 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 to 10 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 10^{12} $cm^{-2}.s^{-1}$ 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 week's time.

During 2008 to 2010 years, the procedure was used on hundreds of archaeological ceramics specimens in sites from Manaus and Marajo island in collaboration with various archaeologists and geologist.

In Manaus the sites are in the district of Iranduba, 30 km southwest of Manaus, in the region located on the left margin of the Solimões river, next to the merging of the Negro river. The site is formed by a farm which is in a fertile valley in the central Amazon. This region contains both dry and flooded land (swamp). The dry land, which is not inundated by the annual river's flooding, is sustained by sedimentary rocks from the Alter do Chão and Novo Remanso formations from the Cretaceous and Miocene periods, respectively. The swamp area, inundated by annual floods, corresponds with the Quaternary sedimentary deposits from the Solimões and Ariaú river flood plains.

The Marajoara ceramic are from Marajó Island on the Amazon river delta area and are highly elaborated ceramics by means of a process of cultural change that occurred within communities that inhabited the area 3500 years ago. Radiocarbon dates place the period of major growth and expansion of Marajoara culture between the 5th and 14th centuries. The Marajoara style seems to be related to different regions within the Marajoara domain, as well as to different chronological periods. The archaeologists noticed that the urns were buried together with other ceramic objects, such as stools, figurines, miniatures, plates, vases, tangas (pubic covers), and a variety of ornaments, The designs had a symbolic significance of a social or religious character with highly complex ceremonial wares in form and decoration. Decorative techniques involve slip, painting, incision, excision and scraping. The ceramics are tempered with ground potsherds (grog) and two plain types that also define two different types of paste that can be found in all decorative types. Some pottery tempered with crushed ashes of a tree bark known as caraipé (*Licania scabra*) are also found. The use of caraipé as temper material was introduced in the Island AD 500. This material was used in the Amazon Basin towards the end of the first millenium and it is associated with Polychrome Tradition.

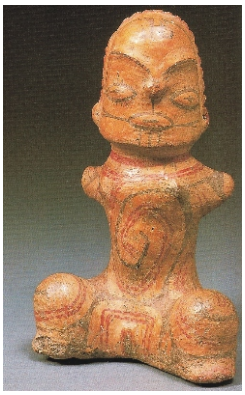


Figure 9. Female figurine

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: 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.

Neutron activation analysis and electron spin resonance for fossil samples dating

The Electron Spin Resonance (ESR) dating is based on the fact that ionizing radiation can create stable free radicals in insulating materials, like tooth enamel and bones. The concentration of these radicals - determined by ESR - is a function of the dose deposed in the sample along the years. The accumulated dose of radiation, called Archeologial Dose (AD), is produced by the exposition to environmental radiation provided by U, Th, K and cosmic rays. If the environmental dose rate (D_{an}) in the site where the fossil sample is found is known, it is possible to convert this dose into the age of the sample by the equation: age=AD/D_{an}. The annual dose rate coming from the radioactive elements present in the soil and in the sample itself can be calculated by determining the U, Th and K concentration. Therefore, the determination of the dose rate depends on the concentration of these main radioactive elements. Neutron Activation Analysis has the sensitivity and the accuracy necessary to determine U and Th with this objective. Recently our group studied some samples of pleistocene mammal teeth from Rio Grande do Sul and Stegomastodon teeth from north-eastern Brazilian megafauna.

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 been working in collaboration with the University of São Paulo (USP) and University of Campinas (UNICAMP), analyzing trace elements in different kinds of rocks by INAA.

The analytical procedure consisted 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 10^{12} 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 ^{60}Co. The gamma-ray spectra were processed by using the VISPECT gamma-ray software which locates peak positions and calculates the energies and net areas.

Analyses of rocks of the Rio Capim belt, São Francisco Craton, were performed in collaboration with the Geoscience Institute of UNICAMP. The results obtained corroborated to support the proposition that the Rio Capim belt was a Palaeoproteorozoic intra-oceanic arc sequence that collided with a continent, of which the Mesoarchaean Uauá block is a remnant.
In order to obtain additional constraints about the Paraná Magmatic Province basalt genesis, analyses of tholeiitic rocks from flows and sills from Northern region, particularly those from Northern São Paulo, Southern Minas Gerais, and Southern Goiás States were performed in collaboration with the Astronomy, Geophysics and Atmospheric Sciences Institute of the University of São Paulo.

Radiological impact of using phosphogypsum in agriculture and as building material

Phosphogypsum is a by-product of the phosphate fertilizer industry. It is produced by precipitation during wet process of phosphate rocks, thus posing serious problems with its utilization and safe disposal. In Brazil, three main industries are responsible for the production and storage of about $5.5x10^6$ tons per year. Phosphogypsum may contain trace metals and radionuclides of U and Th series. Since, in Brazil, phosphogypsum has been used for many years as soil amendment, it is important to know their availability in the environment. The main objective of this study is to evaluate the radionuclides and metals transfer in the soil-to-plant system. To accomplish this task an experiment was carried out in green house (Figure 10), where two major crop groups (soya bean and corn) and leafy vegetables (lettuce) were grown in two types of soil (clay and sandy) amended with phosphogypsum. The transfer-factors were evaluated for the metals (As, Cd, Cu, Ni and Pb, Ba, Co, Cr, Fe, Zn and REE) and for the radionuclides U, Th, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po. The addition of PG to the two soils studied, in the dosage of 1.0 g dm^{-3} for the clayish soil and 0.4 g dm^{-3} for the sandy soil, did not alter significantly the levels of radioactivity, metals and REEs in the final mixture and consequently the TFs obtained for lettuce, soya and corn. Therefore, the impact of one single application of Brazilian PG as soil amendment does not imply in any additional risk due to the transfer of radionuclides, metals and REEs to crops. This project had financial support from CNPq and was conducted in partnership with CDTN.

Phosphogypsum can also be used as building material. In order to evaluate the radiological implications of its use, an experimental house was built, having some of its rooms entirely lined with phosphogypsum. Measurements of samples of phosphogypsum plates from different origins resulted in values of 0.2 to 2.6 for the external radiation index, thus justifying a more detailed investigation. In this study, the application of a previously developed computational model to forecast external doses indoors was described. A comprehensive radiological evaluation was performed, including measurement of the external gamma exposure and radon concentrations in one of the rooms of the house. The results show that the annual increment in the effective dose to an inhabitant of the house will remain below the 1mSv limit for every reasonable scenario. The radon measurements were carried out over a period of 18months, in order to determine the long- term average levels of the indoor radon concentrations. The results obtained are below $200Bqm^{-3}$, the recommended investigation level for radon. The radon exhalation was also evaluated from plates and bricks manufactured with phosphogypsum and from phosphogypsum piles from two fertilizers producers, Ultrafertil, located in Cubatão and Fosfertil, located in Uberaba. This project had financial support from CNPq and FAPESP and was conducted in partnership with Fosfertil.

Figure 10. Lettuce grown in the green house

Assessment of atmospheric pollution in the vicinity of a tin and lead industry using lichen species Canoparmelia texana

This study examines the viability of using Canoparmelia texana lichen species (Figure 11) as a bioindicator of air pollution by radionuclides and rare earth elements (REEs) in the vicinity of a tin and lead industry. The lichen and soil samples were analyzed for uranium, thorium and REEs by instrumental neutron activation analysis. The radionuclides ^{226}Ra, ^{228}Ra and ^{210}Pb were determined either by Gamma-ray spectrometry (GRS) (soils) or by radiochemical separation followed by gross alpha and beta counting using a gas flow proportional counter (lichens). The lichens samples concentrate radionuclides (on the average 25-fold higher than the background for this species) and REEs (on the average 10-fold higher), therefore they can be used as a fingerprint of contamination by the operation of the tin industry. This project had financial support from FAPESP.

Figure 11. Canoparmelia texana lichen

Chemical and radiological characterization of clay minerals used in pharmaceutics and cosmetics

Clays have been used for therapeutic purposes, as active ingredients or as excipient in formulations for a variety of purposes. Despite their wide use, little information is available in literature on their content of trace elements and radionuclides. The

purpose of this study was to determine the elements (As, Ba, Br, Cs, Co, Cr, Eu, Fe, Hf, Hg, La, Lu, Rb, Sb, Sc, Sm, Ta, Tb, Yb, Zn, and Zr) and the radionuclides (U-238, Th-232, Ra-226, Ra-228, Pb-210 and K-40) in Brazilian clays as well as the health and radiological implications of the use of these clays in pharmaceutical formulations.

Radioactive and stable elements' concentration in medicinal plants from Brazil

Since the early days of mankind, plants have been used as food and for medicinal purposes. Still, little information exists in literature about the activity concentration of U-238 and Th-232 decay products, as well as stable element concentrations in Brazilian plants. Activity concentrations of Ra-226, Ra-228 and Pb-210, and chemical concentrations of As, Ba, Br, Cs, Co, Cr, Cu, Eu, Fe, Hf, La, Lu, Rb, Sb, Sc, Sm, Ta, Tb, Yb, Zn and Zr were determined in ten samples commonly used in Brazilian medicinal plants.

Measurement of activity concentration of natural radionuclides for the assessment of radiological indices

The main external source of background radiation exposure of the population are the terrestrial radionuclides like the single occurring radionuclides as ^{40}K and the radionuclides of the ^{238}U and ^{232}Th decay series, with half-lives of the same order that the age of the earth.

The activity concentrations and the gamma-absorbed dose rates of the terrestrial naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K were determined in commercially-used granites from Paraná state (18 samples) and from Espírito Santo State (6 samples), Brazil, and in superficial sand samples for 16 locations throughout the coast of the Great Victory, metropolitan region of the state of Espirito Santo State. All samples were measured by high resolution gamma spectrometry in triplicates after a 30-days ingrowth period.

For Paraná State granites, preliminary results show activities concentrations varying from 4 ± 1 Bq.kg^{-1} to 79 ± 3 Bq.kg^{-1} for ^{226}Ra, 7 ± 1 Bq.kg^{-1} to 142 ± 6 Bq.kg^{-1} for ^{232}Th and 214 ± 14 Bq.kg^{-1} to 1626 ± 77 Bq.kg^{-1} for ^{40}K. All results are within the range of literature values for similar rocks.

For Espírito Santo State granites, preliminary results show concentrations varying from 31 ± 10 Bq.kg^{-1} to 219 ± 29 Bq.kg^{-1} for ^{232}Th, from 17 ± 2 Bq.kg^{-1} to 270 ± 20 Bq.kg^{-1} for ^{226}Ra and from 498 ± 21 Bq.kg^{-1} to 1481 ± 60 Bq.kg^{-1} for ^{40}K. The southern region of Espírito Santo State shows the highest values for ^{226}Ra, ^{232}Th and ^{40}K. The lowest values of concentration for the same radionuclides were observed for north region.

For Espírito Santo sands, ^{226}Ra concentrations varied from 3 ± 1 Bq.kg^{-1} to 738 ± 38 Bq.kg^{-1}, with the highest values for the central locality of the Camburi beach. ^{232}Th concentrations varied from 7 ± 3 Bq.kg^{-1} to 7422 ± 526 Bq.kg^{-1}, with the highest values for Areia Preta beach. ^{40}K concentrations varied from 14 ± 6 Bq.kg^{-1} to 638 ± 232 Bq.kg^{-1}, with the highest values for Areia Preta beach. Radium equivalent and the external hazard index results showed two distinct groups. In the first one, for the majority of the beaches, the radium equivalent activities are in the range from 15 ± 6 Bq.kg^{-1} to 257 ± 18 Bq.kg^{-1}, below of the lower limit of 370 Bq.kg^{-1} recommended by OECD for the safe use of building materials and external hazard index was below the acceptable limit of 1 suggested by UNSCEAR. In the second one, for Setibinha, Curva da Jurema, South and Central locations of Camburi and Areia Preta beaches, the radium equivalent results are in the range from 818 ± 37 Bq.kg^{-1} to 11228 ± 577 Bq.kg^{-1}, exceeding 2 to 30 times the recommended minimum value for safe application in the civil construction industry and the external hazard index surpassed almost 3 to 40 times the UNSCEAR suggested limit.

Radioactivity in surface, underground and drinking waters of high natural radioactivity regions of Brazil

The levels of gross alpha and beta natural radioactivities in surface, underground and drinking waters consumed by the urban and rural population living in Lagoa Real Uranium Province of central south Bahia state, Brazil were determined in several samples collected both from the dry and rainfall season, from urban public supply of drinking water and, in the rural area, from dug and drilled wells and also from small dams and reservoirs supplied with rainfall. After pre-concentration, the samples were evaporated under an infrared lamp on inox planchets and subsequently counted on a Berthold LB 770 thin end-window low-background proportional counter. Preliminary results show natural radiation levels varying from 0.02 ± 0.001 Bq/L to 0.80 ± 0.04 Bq/L for gross alpha activity and from 0.010 ± 0.006 to 3.0 ± 0.2 Bq/L for gross beta activity. The overall results show that, for the Caetité and Livramento de Nossa Senhora regions there is no significant difference between the rain and dry seasons. Also, for these two regions, all values are within the 2004 WHO recommended levels of 0,5 Bq/L for gross alpha and 1 Bq/L for gross beta activities. Some values of Lagoa Real region exceed the 2004 WHO recommendation levels, however, as we are dealing with natural radiation in a well-known high background radiation area, more studies are needed. For a complete assessment, further uranium concentrations determinations for the same samples will be performed.

Radon and thoron dosimetry with nuclear tracks detectors

Radon, thoron and their progeny are always present in the open atmosphere, but are found in higher concentrations in the mineral waters and mud used for therapeutic and recreation purposes in spas, in confined atmospheres of underground workplaces like natural caves, where tour guides are exposed to these radionuclides, and at facilities dealing with great uranium and thorium concentrations.

Radon and thoron levels at several workplaces are determined using Solid State Nuclear Track Detectors (SSNTD), exposed at least three months over a time period of more than one year. The committed effective doses are evaluated for workers and general public according to the ICRP procedures.

At "Termas de Araxá" spa, the effective committed dose due to ^{222}Rn and ^{212}Pb inhalation was evaluated for workers and patients. Radon measurements were carried out with Makrofol E SSNTD over a period of 21 months. The ^{212}Pb air concentration was assessed through the modified Kusnetz's method. Doses received by the spa workers were below 20 mSv.y^{-1}, suggested by ICRP 60 as an annual effective dose limit for occupational exposure. The radiation doses for the patients were below the mean annual effective dose due to natural sources estimated to be 2.4 mSv.y^{-1}.

The monitoring of radon in natural caves of three touristic parks of São Paulo state, Morro Preto and Santana caves, located at PETAR (Alto Ribeira State Park), Caverna do Diabo, in Jacupiranga State Park and Caverna Colorida, in Intervales State Park. Radon measurements between October 2003 and November 2005 and further between April 2009 and June 2010 were carried out with CR-39 SSNTD. The annual effective dose was calculated considering the most realistic scenario, 52 working weeks and an equilibrium factor of 0.5. Doses received by the tourist guides were below 20 mSv.y^{-1}, suggested by ICRP 60 as an annual effective dose limit for occupational exposure.

At IPEN, São Paulo, Brazil, the nuclear materials storage site deals with considerable uranium and thorium concentrations. The occupational committed effective dose due to ^{222}Rn, received by workers was assessed through the radon activities determined with Makrofol E SSNTD, exposed over a period of 21 months. 13 measurement stations were spread inside the storage site and one was placed at the anteroom used for material transfer. The committed effective dose results varied from 4.1 to 5.4 mSv y^{-1} at the nuclear materials storage site and from 2.9 to 3.7 mSv y^{-1} at the ante-room, according to the working time. All effective committed doses received by workers were below the 20 mSv y^{-1}, suggested as an annual effective dose limit for occupational exposure by ICRP 60.

Determination of natural radionuclides from U-238 and Th-232 series, trace and major elements in sediment cores from Baixada Santista and evaluation of impacted areas

Baixada Santista is the region of higher population of the coast of São Paulo Sate, where is located the largest port and the most important industrial complex of Latin America. As a direct result of the industrial and port activities and the large population growth this region is considered highly impacted because it has received in recent years a considerable load of industrial and domestic effluents. Ten sediment cores, distributed in the ecosystems: estuary of Santos-Cubatão, estuary of São Vicente, channel of Bertioga and Santos Bay (Figure 4) were collected in order to determine the concentration of trace and major elements and natural radionuclides from the ^{238}U and ^{232}Th series using the techniques, neutron activation analysis, X-ray fluorescence and gamma spectrometry. No enrichment was found for the major elements in the ecosystems studied; the highest concentrations of ^{232}Th and ^{238}U were found in ecosystems that are under the influence of the industrial complex. In the four studied ecosystems the most critical elements found and that deserve the attention are the elements As, Br and Sb, because they are enriched in different degrees. Comparing the obtained values of As, Cr and Zn elements in the sediment cores with the values of TEL and PEL index for sediment quality, it was verified that the studied region presents As levels higher than TEL in all the sampled locations; the Cr element presented superiors values than TEL in Santos-Cubatão estuary and Bertioga channel. The Zn element presented values higher than TEL for some core slices of Santos-Cubatão estuary, for one core of São Vicente estuary, one core in Bertioga channel and Santos Bay. Taking under consideration CONAMA nº 344/04 resolution, all the sediments can be considered as class 1, and are below the reference values for the decision to manage the dredged material.

Figure 12. Sampling location in Ilha das Palmas, Santos Bay

Application of nuclear and isotopic tracer techniques to study land-ocean interactions in São Paulo State Coastal Plain and in Bransfield Strait, Antartica

The distribution of natural Ra isotopes was studied in surface, groundwater and estuarine water samples collected from dry and wet seasons (2009-2010) campaigns performed in Ribeira Valley, Southern São Paulo State. Results evidenced that there is a prevalence of Ra-228 isotope in all the set of samples analyzed, although the activity concentrations of Ra isotopes are representative of natural background levels, showing low or minimal human intervention. In the set of samples studied along Ribeira do Iguape River, Cananéia and Iguape outlets, the higher concentrations of Ra were observed in bottom waters, indicating the diffusion of Ra-228 from sediments recently deposited as a potential source of the increased concentrations of this isotope when compared with others. Fluxes of Ra for Cananéia outlet are strongly influenced by tidal oscillations, which modulate the increase and decrease of Ra concentrations in response of the respective waters salinity gradient. In Iguape outlet and in the hydrochemical stations performed along Ribeira do Iguape River it was observed a linear relationship between the amount of suspended matter and the increase of Ra-228 activity concentration. Th-234 was used as a tracer of organic carbon fluxes distribution in the Bransfield Strait in order to evaluate its influence in the CO^2 drawdown, since POC export via sinking particles is the primary mechanism of carbon sequestration in the Southern Ocean (Figure 13). Because of its very particle reactive behavior, Th-234 is removed from a parcel of water in only two ways, through decay and through particle flux. Therefore, a steady-state 1D activity balance can be used to calculate its flux. Fluxes up to 15.274 dmp m^{-2} d^{-1} were estimated, the highest value observed in Station 09 at 794 m depth. POC exported fluxes derived from the disequilibrium Th-234/U-238 model varied from 0.6 to 16,000 mmol C m^{-2} d^{-1}.

Figure 13. Water sampling in Baia do Almirantado

Dosimetric Materials

The main objective of this research area is the development of new dosimetric materials with high sensitivity, low cost and easy obtaining. Single crystals of $CaSO_4$:Tm were obtained under controlled crystal growth conditions, such as temperature, atmosphere and velocity, aiming to improve the TL sensitivity of the crystals and investigate its dosimetric properties in order to evaluate the applicability of this material to dosimetry. The preliminary results show that $CaSO_4$:Tm presents high sensitivity and reproducibility and may be applied to radiation dosimetry.

Other Rare Earth elements, namely, Ce, Nd, Eu and Tb, and the post-transition metal Tl were also assessed as $CaSO_4$ activators, with Ce and Eu being the best candidates for further studies. Both $CaSO_4$:Ce and $CaSO_4$:Eu present high sensitivity, good reproducibility, linear dose-response in the range of 0.01 to 10 Gy and energy independence to photon beams. Preliminary results suggest that $CaSO_4$:Ce response is very dependent on radiation type and energy while $CaSO_4$:Eu is nearly independent of these parameters, but both dosimetric materials present a linear dose-response for high energy electrons in the tested range of 0.01 to 0.50 Gy.

Aluminum oxide composes the modern TL and OSL radiation dosimeters. TL and OSL phenomena are related to inserted impurities on the crystalline structure of α-Al_2O_3. The studies include the dosimetric properties of alumina samples obtained by electrofusion, adsorption and coprecipitation. Electrofused alumina commercially available was used to produce pellets glass mixed and sintered. Adsorption and coprecipitation were the methods used to introduce metal ions to alumina crystal lattice. The alumina pellets produced by means of sintering at high temperature are being characterized.

DL-Alanine ($C_3H_7NO_2$) is an amino acid tissue equivalent traditionally used as standard dosimetric material in EPR dosimetry. Recently it has been studied to be applied in gel dosimetry, considering that the addition of alanine in the Fricke gel solution improves the radiation induced ferric ions production. The spectrophotomety evaluation technique can be used comparing the two spectrum wavelengths bands: 457 nm band that corresponds to ferrous ions and 588 nm band that corresponds to ferric ions concentration to evaluate the dosimetric properties of this material. The performance of the Alanine gel solution developed at IPEN was studied using spectrophotometry technique. The obtained results indicate that signal response dependence for clinical photons and electrons beams for Alanine gel dosimeter is better than 3.6 % (1σ) and the energy dependence response is better 3% (1σ) for both beams. These results indicate that the optical response is energy

independent in the studied dose range and clinical photons and electrons beams energies.

Quartz is a crystalline material abundant in the nature, found with different colors and properties according to the region where it is extracted. This material has been largely studied to be applied as dosimetric material, mainly for retrospective dose evaluation in case of accident dosimetry (Fig. 14). The TL response of green quartz powder samples from Brazil was evaluated to characterize this material to be applied in radiation dosimetry. The evaluation techniques employed were: thermoluminescence, electron spin resonance and spectrophotometry. Different heat treatments pre and post irradiation were evaluated with the purpose of to eliminate the low temperature picks and increase the TL sensitivity. Dosimetric properties such us reproducibility, dose response, dose rate and energy dependent response, TL and EPR signal stability were studied. The obtained results indicate that the green quartz is a promising dosimetric material.

Figure 14. Green quartz samples

External Dosimetry

The main research works developed in the external dosimetry area were:

Development of a passive individual gamma-neutron mixed field dosimeter calibrated to be used in ^{241}AmBe sources. The dosimeter uses the techniques of Solid State Nuclear Track Dosimetry (SSNTD) and Albedo Thermoluminescence (TL) Dosimetry.

The intrinsic efficiency of $CaSO_4$:Dy TL dosimeters, developed and commercialized by IPEN to be applied in individual, area and environmental dosimetry, was evaluated for 4, 6, 8, 12 and 16 MeV clinical electron beams at their depth of maximum ionization in polymethyl methacrylate (PMMA), Solid Water (SW) and standard liquid water phantoms aiming to use this material in personal and radiotherapy dosimetry. The reproducibility of the TL response is better than 2.4% and the TL response as a function of the dose, linear between 0.01 and 3.25 Gy. The obtained intrinsic efficiency indicates that $CaSO_4$:Dy dosimeters can be applied to the radiation protection for high energy electron beams generated by linear accelerators.

Aiming to evaluate the dose range that animals submitted to pulmonary radiographic exams are exposed and the relationship with the individual doses of owners and clinical staff, the entrance surface skin dose of dogs of different breed and sizes with cancer and with suspected pulmonary metastasis were evaluated. TL dosimeters of $CaSO_4$:Dy were used to entrance surface skin dose evaluation of 27 dogs. Simulations of dog's irradiation were also carried out using a water phantom. Each procedure was carried out by the acquisition of three chest radiographic images, two latero-lateral and one ventro-dorsal of dogs with suspect of pulmonary metastasis. The obtained results has shown to be extremely important the assessment of doses involved in veterinary diagnostic radiology procedures both to protect the occupationally exposed workers and to optimize the delivered doses to the animals.

Microdosimetry

The main objective of this research is the dosimetric evaluation of semiconductor components (surface barrier detectors and PIN photodiodes) for applications in microdosimetry and dose equivalent measurements on low dose fields (fast and thermal fluxes) using an AmBe neutron source, the IEA-R1 reactor neutrongraphy facility (epithermal and thermal fluxes) and the Critical Unit facility IPEN/MB-01 (fast fluxes).

As moderator compound to fast neutrons flux from the AmBe source was used paraffin and boron and polyethylene as converter for thermal and fast neutrons measurements. Thermal neutron measurements of AmBe source and Neutrongraphy facility were developed using 10B converter through charged particles detection and for fast neutrons measurements polyethylene converter was used as proton recoil generator. The resulting fluxes were used to the irradiation of two semiconductor components (SSB - Surface Barrier Detector and PIN photodiode).

Monte Carlo simulation methodology was employed to evaluate analytically the optimal paraffin thickness. The obtained results were similar to the experimental data and allowed the evaluation of emerging neutron flux from moderator, as well as the fast neutron flux reaching the polyethylene covering the semiconductor sensitive surface. Gamma radiation levels were evaluated covering the whole detector with cadmium foil 1 mm thick, allowing thermal neutrons blockage and gamma radiation measurements.

The obtained results were in good agreement with other studies published. Using the obtained spectra an approach to dose equivalent calculation was established.

Internal dosimetry

A cross-species extrapolation biodistribution study using ^{177}Lu-DOTATATO has been developed to aid the dose estimation due to peptide receptor radionuclide therapy (PRRT) patients. Besides, this radiopeptide uptake by the Erlich tumor (ascetic form) has been studied as a possibility for test new Somastanine analogs radiopharmaceuticals.

Whole-body retention to workers after accidental inhalation of radionuclides is studied. The counting system used for measuring high-energy gamma emitters comprises two thallium-activated sodium iodide [NaI(Tl)] detectors. The data used in this assessment include whole body and thyroid measurements over specific periods of time. The effective half-lives are obtained to both whole-body and thyroid.

The photon spectra of reference radionuclides are calculated by using the transport code Penelope. The contributions from scattered photons to the spectra fields have been determined with regard to the quantities photon fluence. The mean photon energies calculated with respect to the mentioned quantity are studied. Differences in the design of the sources and their influence on the spectra are assessment. The dependence of the scattered photon component from the energy are examined by feeding the Penelope code with monoenergetic photons of different energies.

As a part of a continuous improvement of the monitoring programme for occupationally exposed workers at IPEN, a computational code for internal dose assessment was developed. The code is an agile and efficient tool for the designing, visualization and resolution of compartmental models of any nature.

The architecture of the system is conceived containing two independent soft wares: CBT (Computer Building Tools) and SSID (Smart Software for Internal Dosimetry). The first one is responsible for the set up and manipulation of models and the SSID is responsible for the mathematical solution of the models. Four different techniques are offered for the resolution of system of equations, including semi-analytical and numerical methods, allowing comparison of precision and performance of both. The software was developed in C# programming, using Microsoft Access database and XML standards for file exchange with other applications. Compartment models for uranium, thorium and iodine were generated for the validation of the CBT software. The models were subsequently solved by SSID software and the results compared with the values published in issues of ICRP.

Metrology in Radiotherapy

In the dosimetry area related to the medical applications of the ionizing radiation were studied:

The TL performance of $CaSO_4$:Dy and LiF:Mg,Ti dosimeters to photon and electron beams applied to radiotherapy was investigated. The TL response of these dosimeters was studied for 6 and 15 MV photons and 4, 6, 9, 12 e 16 MeV electron beams using PMMA, liquid water and Solid Water (SW) phantoms. Using a Varian linear accelerator Clinac 2100C (Fig. 15). The TL dose-response of both dosimeter and three phantoms types present linear behavior on the photon and electron dose range from 0.1 to 5 Gy. The obtained results indicate that the performance of $CaSO_4$:Dy dosimeter is similar to LiF:Mg,Ti dosimeters and this material can be an alternative in clinical electron beams dosimetry.

Figure 15. Clinical electron beam irradiation set up

The complex cancer treatment techniques require rigorous quality control. The Fricke xylenol gel (FXG) dosimeter has been studied to be applied as a tridimensional dosimeter because it is possible to produce 3-D FXG phantoms of various shapes and sizes.

The optical response of the Fricke xylenol gel (FXG) dosimeter developed at IPEN using 270 Bloom gelatine from porcine skin made in Brazil, for clinical electron beams from a VARIAN® electron linear accelerator in energy range from 6 to 16 MeV to reference depth, using a water phantom and OA spectrophotometry technique was evaluated. The optical dose response is presented in Figure 16 to doses ranging from 0 to 40 Gy.

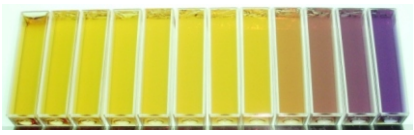


Figure 16. Optical dose responses to gamma radiation, photon and electron clinical beams of FXG solution

The performance of a head phantom filled with FXG solution was also evaluated using RMI technique to 3DCRT with multiple radiation fields and clinical photon beams. The obtained results indicates that the target volume can be clearly observed for all MR images of the FXG phantom irradiated with 6 MV clinical photon beam and, in the case of coronal image, the radiation beam projection and the overlap of different radiation fields used can also be observed. These results

encourage additional tests using complex treatment techniques and they indicate the viability of applying the studied phantom for routine quality control measurements and in 3DCRT and IMRT treatment planning.

$^{90}Sr+^{90}Y$ clinical applicators have to be periodically calibrated, according to international standards and recommendations. Four calibration methods of clinical applicators were studied, comparing the results with those of the calibration certificates. The methods included the use of a standard applicator calibrated by the National Institute of Standards and Technology, and an Amersham applicator, of the Calibration Laboratory (LCI) as reference; a mini-extrapolation chamber developed at LCI; and thermoluminescent dosimetry. The distribution of the depth dose in water, important in dosimetry of clinical applicators, was determined. The results obtained were comparable to the data of the IAEA standard. Furthermore, a dosimetry postal kit was developed for the calibration of clinical applicators using the thermoluminescent technique.

Metrology in Diagnostic Radiology

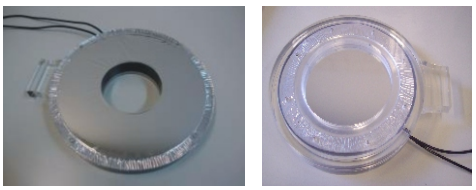
During the period of 2008-2010 new radiation qualities were characterized at the Calibration Laboratory of IPEN, São Paulo, in order to adequate its procedures to the IAEA new code of practice, TRS 457 (Dosimetry in Diagnostic Radiology: An International Code of Practice). A quality control methodology was developed for the instruments used in interventional diagnostic radiology. All X-radiation qualities were established following the recommendations of the IEC 61267 standard. Procedures to calibrate instruments used in conventional diagnostic radiology, computed tomography and mammography measurements were also improved. All these radiation qualities are available to new dosimetric materials irradiation.

The behaviour of the main X-ray equipment of the Calibration Laboratory, that operates in the range from 25 kV to 150 kV, was studied using a non invasive PTW meter, model DiavoltTM, and an ORTEC spectrometry system, model NOMAD-PLUS 92X. To complete the diagnostic radiology radiation qualities establishment, four special phantoms (two for mammography and two for conventional radiology) were developed in order to study the entrance air kerma in diagnostic radiology patients.

The influence factors of the radiopharmaceutical vial dimensions used for activimeter calibration at IPEN was studied with the objective to establish a quality control programme and to determine the correction factors for the geometry of the vials. They are used for distribution of radiopharmaceutical and activimeters calibration, using a NPL-CRC Secondary Standard Radionuclide Calibrator System with traceability to National Physical Laboratory (NPL) and calibrated with a P6 vial type with different dimensions than that of IPEN. The results showed a maximum variation of 22% for ^{201}Tl, and the minimum variation was 2.98% for ^{131}I. The correction factors must be incorporated in the routine calibration of the activimeters.

Also in this research field, some ionization chambers were developed to be used in X-ray beams, diagnostic radiology level. A ring-shaped graphite ionization chamber was designed and assembled to monitor X-ray beam intensity (Figure 17a). International organization protocols recommend the use of monitor chambers to assure the correct dose delivery. This ring-shaped ionization chamber has the advantage over commercial monitor chambers, also called transmission chambers, of not interfering in the radiation beam spectra. All characterization and response stability tests were performed with the ionization chamber, and the results were within the international recommendations. Also, a transmission chamber, with double sensitive volumes, was developed to compare to commercial transmission chambers (Figure 17b). Its response was within international recommendations.

A double faced ionization chamber was built for quality control program in mammography X-ray beams. This ionization chamber was made of PMMA and has two collecting electrodes of different materials: aluminum and graphite. Both sides of this ionization chamber have the sensitive volume of 6.0 cm^3. The double faced ionization chamber was characterized in standard mammography X-ray beams established at the Calibration Laboratory. The behavior of the developed ionization chamber was adequate when compared to international criteria.



(a) (b)

Figure 17. Monitor ionization chambers developed at IPEN

High Doses and Accident Dosimetry

A simple and inexpensive polymeric material whose optical properties can be observed, quantified and related to absorbed dose was studied. Commercial polycarbonate (PC) is a new type of film detector that suffers yellowing upon radiation exposure.The color change (Fig.18) was used as dosimetric property to quantify gamma absorbed dose. The dosimeter consists of a piece of polycarbonate film (3 x 1 cm^2) 3 mm thick and the used evaluation technique was spectrophotometry. PC films were irradiated with gamma doses between 1 and 150 kGy. The optical response was measured using a UV - VIS spectrophotometer. The

main dosimetric characteristics studied were: pre- and post-irradiation stability, dose response, environmental conditions influence and response dependence with dose rate. PC films are easy to prepare and analyze. The influence of environmental conditions was observed and must be corrected. Polycarbonate dosimeters present good stability and reproducibility and linear behavior in the dose range studied indicating that the dosimetric characteristics are suitable to determine high gamma doses.

Figure 14. Green quartz samples

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;
- Determination of natural and artificial gamma emitters;
- Determination of uranium, thorium and radium isotopes;
- Determination of ^{210}Pb and ^{222}Rn;
- Determination of radionuclides in foodstuffs and food commodities imported and exported by Brazil.

High Doses Dosimetry
The High Doses Dosimetry Laboratory of Ipen developed a dosimetric system based on alanina/ESR that presents good characteristics for use in gamma fields such as: wide dose range from 10 to 10^5 Gy, low fading, low uncertainty (<3%), no dose rate dependence and non-destructive ESR signal readout. The detector is encapsulated in special polyethylene tube that reduces the humidity problems and improves the mechanical resistance. A computer program to extract signals from noise spectra based on the wavelet transform was developed in order to allow the use of the dosimetric system at radiotherapy dose ranges. The dosimetric system was validated by IDAS program from IAEA and is available to high doses measurements.

Dosimetric pellets of $CaSO_4$:Dy
The Dosimetric Materials Laboratory developed and patented the $CaSO_4$:Dy crystals growth system and the method to produce the Teflon® sintered pellets. The $CaSO_4$:Dy crystals are growth in the sealed system and they are cold pressed with the binding material to making the 6 mm diameter pellets (Fig.19). The $CaSO_4$:Dy pellets are destined to the individual, area and environmental monitoring through the thermoluminescence dosimetry. Besides they could be applied to the high doses, retrospective and to the clinical dosimetry. They are utilized to solid state dosimetry research too. The pellets of several materials such as quartz, topaz and jade could be also produced by means of the same referred method. The requests of $CaSO_4$:Dy crystals and pellets from many institutions have been attended by the laboratory.

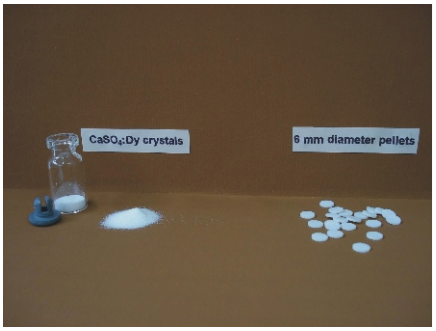


Figure 19. $CaSO_4$:Dy crystals and sintered pellets

Routine External Dosimetry
The external dosimetry applies the thermoluminescent (TL) technique and $CaSO_4$:Dy based dosimeters completely developed at IPEN to carry out individual external exposure, area and environmental monitoring. The individual monitoring service for external exposure is accredited by CNEN (Brazilian Nuclear Energy Commission) regulatory committee, CASEC (Essays and Calibration Services Evaluation Committee) and it completely satisfies CNEN's regulatory norms, besides presenting a brake in IPEN's quality control system, being responsible for the external exposure monitoring of all occupationally exposed individuals at IPEN, other autarchies, governmental offices and even some private facilities. The area and environmental monitoring, which are not under the scope of any regulatory office, are also performed in different society sectors, such as research institutes, universities and private foundations and companies. Aiming to improve individual external exposure, area and environmental monitoring services and give society a return of the budges invested in them, papers based on the results obtained by the services have been present in the most relevant scientific reunions over the triennial period of 2008-2010.

Internal dosimetry
The whole-body and thyroid measurements are routinely performed on IPEN workers, visitors, trainees, and contract workers. During 2008-2010, the Laboratory carried out occupational monitoring of workers involved in radioactive waste management, radioisotope production, research, students and visitors. Routinely the analyses of

biological samples from people occupationally exposed to radionuclides with risk of internal contamination are performed. The most frequently requested radionuclides, were: U-nat, and U isotopes in urine. ^{232}Th, ^{3}H, γ emitters and Actinides, can also be requested. In addition to bioassay measurements, the Laboratory participated in national (Brazilian Intercomparison Programs) and in international intercomparison (Promotion du Controle de Qualité 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. No worker received dose higher than the annual limit on intake in the period 2008-2010 for dose assessment, as part of the internal monitoring program.

Calibration of radiation monitors and dosemeters

The Calibration Laboratory has since 1980 been calibrating instruments used in radiation protection and therapy measurements and belonging to hospitals, industries, clinics and other users located in São Paulo and in other parts of Brazil. Since 2000 calibration service is being offered to users of diagnostic radiology instruments with the establishment of standard radiation quality at this level (Fig. 13). At the radiation protection level there are special set-ups with gamma (^{60}Co and ^{137}Cs), beta (^{90}Sr + ^{90}Y, ^{204}Tl and ^{147}Pm), alpha (^{241}Am, ^{233}U, ^{238}Pu, ^{244}Cm, etc.) and low energy X radiations (60 kV). Clinical dosemeters (radiotherapy level) can be calibrated, using gamma (^{60}Co) or low energy X radiation. As reference system, a secondary standard ionization chamber is used, traceable to the Physikalisch-Technische Bundesanstalt, PTB, Germany, and to the National Laboratory of Metrology of Ionizing Radiation, Brazil. Instruments used in diagnostic radiology measurements can be tested in X radiation qualities, using a Seifert X radiation system (160 kV) and a reference system with four ionization chambers for diagnostic radiology measurements (mammography, computed tomography, fluoroscopy, radiation protection and conventional diagnostic radiology ionization chamber) traceable to the PTB, Germany. The types of instruments calibrated are: several kinds of ionization chambers, pen dosemeters, survey meters (including superficial contamination detectors), alarm dosemeters, activimeters, clinical dosemeters and others. Besides this service, samples including thermoluminescent dosimeters, alanina and others, using beta, gamma and X radiation were irradiated.

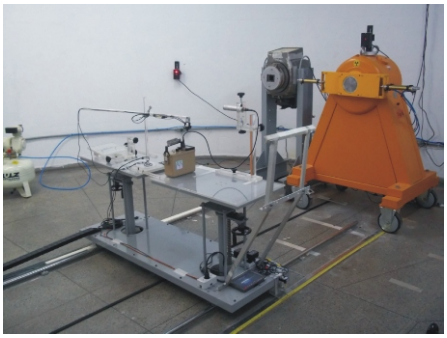


Figure 20. Gamma radiation calibration room general view

Nuclear Science and Technology

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Honor Mention and Awards

Maíra Tiemi Yoshizumi, PhD student, and Dr. Linda Caldas, supervisor, for best oral presentation at the V Congress on Applied Physics to Medicine, on September 2008, with the paper "Performance of a double-faced ionization chamber in standard beta radiation beams".

Luciana Caminha Afonso, PhD student, and Dr. Linda Caldas, supervisor, for best poster entitled "Study of the influence of scattered radiation at a gamma irradiator" presented at the International Youth Nuclear Congress, Interlaken, Switzerland, on September 2008.

Two studies developed in the LEER: "Reference values in whole blood of SJL/J mice using Neutron Activation Analysis" and "Biochemical values in whole blood of horses used for hyperimmune sera production" were awarded during the 9th International Conference on Nuclear Analytical Methods in the Life Sciences. September, 7 to 12.

Two studies developed in the LEER: "Blood Levels of Zinc in crioulo horses used in sera production" and "Sodium analysis in whole blood of athletes using NAA" were selected and awarded with financial support to be presented in the VIII Latin American Symposium on Nuclear Physics and Applications. December, 15 to 19, Santiago.