

Environmental Radioactivity

Maria José Madruga

In 2011, one of the highlight activities of the Environmental Radioactivity Group has been conducting the environmental radioactivity monitoring in Portugal after the Fukushima Nuclear accident that occurred in Japan on March 12. This finding reinforces the importance of the work developed by the Group particularly in the case of nuclear accidents in support of the activities of national (Portuguese Environment Agency, National Authority of Civil Protection, etc.) and international entities (EC, AIEA, etc.).

Legal activities:

One of the main activities of the Environmental Radioactivity Group (GRA) concerns the Radiological Environmental Monitoring Programmes which are performed in collaboration with the Measurement Laboratories (LM), the Dosimetry and Radiobiology (GDR) and the Radiological Protection and Radioactive Waste Management (GRRR) Groups. These surveys were established according to the Recommendations of the Articles 35/36 of the EURATOM Treaty and its execution is legally attributed to ITN (Decree-Law 138/2005 of 17th August). In April, in the framework of the Article 35 requirements, an EC verification team visited the ITN in particular the GRA laboratories where the analyses concerning the radiological surveillance programme for the regions around old uranium mining sites are performed.

Research and development activities:

Current research activities are ongoing to investigate the levels of radioactivity in the environment and the biogeochemical cycles of natural and artificial radionuclides in the atmosphere (aerosols) aquatic and terrestrial environments.

Quality system:

Regarding the quality assurance of the radioanalytical techniques one of the objectives of the Group for this year was achieved. The accreditation concession audit for three radioanalytical techniques (gross alpha/beta in waters by proportional counters and by Liquid Scintillation Counting and tritium determination in

waters by LSC) was conducted by the Portuguese Accreditation Body (IPAC-Instituto Português de Acreditação). The accreditation concession of these techniques by IPAC is foreseen for 2012.

One GRA staff member (Irene Lopes) was nominated (since September 2011) the UPSR Quality Manager.

Participation in intercomparison exercises:

The Group participated in collaboration with the Measurement Laboratories in the following international inter-comparison exercises: determination of radionuclides in soil and water organized by the IAEA/ALMERA (Seibersdorf, Austria); determination of ¹³⁷Cs, ⁹⁰Sr and ⁴⁰K in wild bilberry organized by the EC (IRMM, Geel, Belgium) and determination of natural and artificial radionuclides, global alpha/beta and tritium in water organized by the Consejo de Seguridad Nuclear (CSN, Spain). Regarding the indoor radon the GRA participated in two exercises: one, organized by NIRS (National Institute of Radiological Sciences, Chiba, Japan) and the other one, by the University of Cantabria, Spain. The results published in 2011 are in good agreement/compatible with the reference values.

Technical services:

The technical services developed by the Group were carried out under contract with companies or by request from enterprises or Government organizations. The technical services concern the evaluation of the radioactivity levels in public water supplies (Decree-Law 306/2007), mineral waters, indoor radon measurements (Decree-Law 78/2006; NT-SCE-02), foodstuffs, biological samples, etc.

Participation in national and international technical and scientific committees:

GRA members are involved in several committees and working groups from IAEA, EC and CTBT.

Education and Training:

GRA members are involved in education and training activities in radioanalytical techniques, environmental radioactivity and radiological protection.

Research Team

Researchers

M. J. MADRUGA, Princ., Group Leader
F. P. CARVALHO, Princ.
J. A. CORISCO, Aux.
M. J. REIS, Aux. (25%)

Students

A.R. GOMES, FCT grant
E. ANDRADE, FCT grant
J. MELO, FCT grant

Technical Personnel

A. LIBÂNIO
A. MOURATO
I. LOPES
J. M. OLIVEIRA (70%)
M. M. MALTA

Radioactive Fallout in Portugal Following the Fukushima Nuclear Accident

M. Reis, M.J. Madruga, L. Silva, E. Andrade

Objectives

Following the nuclear accident that occurred in Fukushima Daichi (Japan) complex on March 12, 2011 short (^{129m}Te , ^{131}I , ^{132}Te , ^{136}Cs) and long lived anthropogenic radionuclides (^{134}Cs , ^{137}Cs) were released to the atmosphere and dispersed along the world reaching the European countries including Portugal, around two weeks after the accident (end of March 2011). In Portugal, the radioactivity in the atmosphere was measured using a continuous high volume (ASS-500) aerosol sampling station located at ITN *campus* (Sacavém). Samples of aerosols, grass, leafy vegetables and milk, which give a better indication of the atmospheric radioactive deposition in the case of a nuclear accident, were collected and analysed to the gamma emitter's radionuclides using high resolution gamma spectrometry analysis. Radioactivity on atmospheric aerosols (related to Fukushima accident) was also detected at Açores (S. Miguel island) by the radionuclide particulate station RN53, from the International Monitoring System of the Comprehensive Nuclear Test Ban Treaty Organization (CTBTO).



Fig. 1 Aerosol sampling station ASS-500, located at ITN *campus*.

Results

During about one month (end of March until end of April 2011), traces of ^{131}I , ^{137}Cs and ^{134}Cs , amongst others, were detected in aerosols particles, reaching the maximum values for ^{131}I ($0.9 \pm 0.1 \text{ mBq m}^{-3}$) ^{137}Cs ($0.13 \pm 0.01 \text{ mBq m}^{-3}$) and ^{134}Cs ($0.126 \pm 0.004 \text{ mBq m}^{-3}$) on the last week of March.

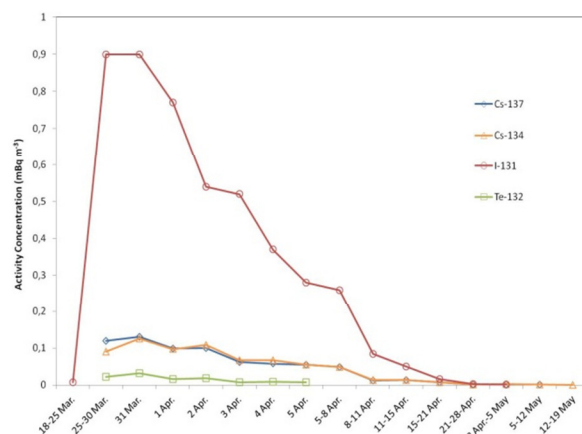


Fig. 2 Evolution of the activity concentration, at Sacavém, of several anthropogenic nuclides related to the Fukushima accident.

The fallout of those aerosols particles contributed to the maximum levels of radioactivity ($2.48 \pm 0.24 \text{ Bq m}^{-2}$ and $0.93 \pm 0.15 \text{ Bq m}^{-2}$ for ^{131}I and ^{137}Cs respectively) measured in grass samples, which were observed during the first week of April. Traces of ^{131}I were also detected in milk and leafy vegetables collected during the same time period. The isotopic ratio of some nuclides ($^{134}\text{Cs}/^{137}\text{Cs}$, $^{137}\text{Cs}/^{131}\text{I}$ and $^{137}\text{Cs}/^{132}\text{Te}$), together with the backward trajectories simulated using the HYSPLIT lagrangian model, clearly indicates that the detected anthropogenic nuclides originate from Fukushima (Japan). However, the levels of radioactivity observed do not raise any concern regarding the Portuguese population exposure and therefore no radiological protection measures were recommended by the competent authorities.

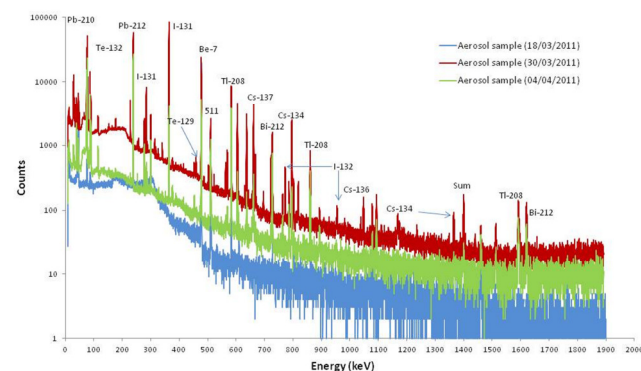


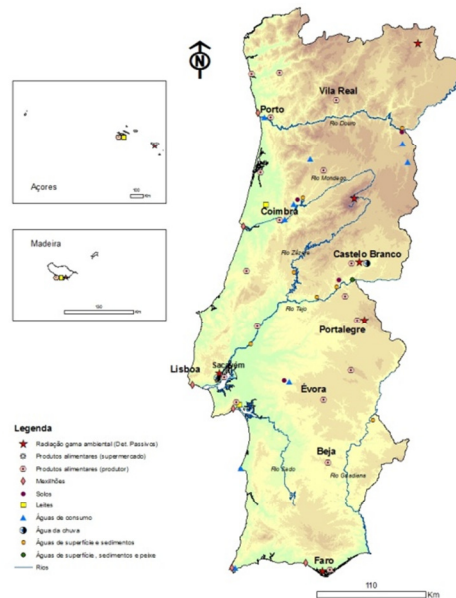
Fig. 3 Comparison of aerosol sample spectra before and after the detection of radionuclides from Fukushima.

Radiological environmental monitoring programmes

M.J. Madruga, F.P. Carvalho, M. Reis, J.G. Alves, J.A. Corisco, R. Trindade, I. Lopes, J. Abrantes, J.M. Oliveira, L. Silva, L. Portugal, M. Malta, A. Libânio, A. Mourato, G. Silva, L. Santos, A. Batista, A.R. Gomes, E. Andrade, G. Carvalhal, J. Melo, M. Pereira

The Radiological Environmental Monitoring Programmes performed according to Article 35 of the EURATOM Treaty include the Programme at a National Level, established by law in 2005 and which execution was legally attributed to ITN (Decree-Law 138/2005, 17th August), the Source Related Programme for the ITN *Campus* and the Specific Programme for the Regions around Old Uranium Mining Sites. The radiological environmental survey has as main objectives to quantify the artificial and natural radionuclide levels in the environment (aquatic, terrestrial and atmospheric) considered as direct pathways of contamination to man and to evaluate the external radiation levels in order to assess the potential exposure of the Portuguese population. The monitoring programme at a national level concerns the radioactivity measurements of environmental and foodstuff samples collected along the country. The monitoring programme carried out for the ITN *campus* includes measurements of external radiation levels (gamma dose) and determination of radionuclide activity concentrations in relevant environmental samples (aerosols, rain water and soils) in order to check the operation conditions and discharge controls adequacy and to maintain a continuing record on environmental radionuclide levels related to the source. Monitoring of the regions around old uranium mining and milling sites in Portugal was performed. Water, soil, and vegetable samples were analysed for uranium series radionuclides. Radon measurements and aerosol samples were collected near the uranium mining waste dumps.

During 2011 about 560 samples (aerosols, rainwater, surface water, drinking water, sediments, fish, mixed diet, complete meals, milk, soils, etc.) were collected accordingly to international sampling procedures and a total of about 1970 analyses were performed for the determination of artificial and natural radionuclides, using gamma and alpha spectrometry, alpha/beta measurements and liquid scintillation technique. Integrated measurements of the ambient dose equivalent with thermo luminescent dosimeters were also performed. The results show that the Portuguese population was not exposed to radioactive contamination levels higher than the radioactive background. Therefore, there is no need to adopt any measures for radiological protection of the population. All the data are published in Internal Reports made available in the ITN website (<http://www.itn.pt>) and included in the European Radioactivity Environmental Monitoring Database (REM) located at the EU Joint Research Centre, ISPRA (Italy).



Biomass combustion and release of radionuclides into the atmosphere

F.P. Carvalho, J. M. Oliveira, M. Malta

This Research Project funded by the FCT and coordinated by the University of Aveiro (Acronym BIOEMI; PTDC/AMB/65706/2006) was completed during the current year. In the time course of project characterization of radioactivity in wild vegetation including bush and forest, was made in order to assess the (organic and radioactivity) compounds released with the smoke into the atmosphere and potentially inhaled. During the occurrence of vegetation and forest fires in the summer period, field sampling was performed in closed collaboration with fire brigades. Filters with the aerosol particles collected near the fire were analyzed for naturally occurring radionuclides. The concentrations of the more volatile radionuclides such as ²¹⁰Po, ²²⁶Ra and ²¹⁰Pb in smoke particles, especially in the smallest ones (<0.43 µm), were found very high. People exposed long time to the fumes near fire fronts may inhale significant activities of smoke particles and thus of these radionuclides, and especially ²¹⁰Po. Activity concentration values measured indicate potential for significant radiation exposure and, thus, protection of the respiratory tract is needed to avoid not only the inhalation of smoke particles but also to avoid radiation exposure.

Definition of new ENdpoints to assess GENtoxic effects resulting from environmental exposures to Uranium, Uranium daughters and ionizing radiation in bioindicator species-ENGENUR.

F.P. Carvalho, J. M. Oliveira, M. Malta

This research project is funded by the FCT (Acronym ENGENUR; FCT PTDC/AAC-AMB/114057/2009) and aims at defining new markers for exposure to ionizing radiation from uranium mining tailings in the areas of past uranium mining and milling industrial activity. The mining area selected for work was Cunha-Baixa mine (Mangualde), and samples of wild rodents and amphibians were collected in the area for analyses of biological effects of radiation at genetic, chromosome and tissue level. Exposures of the earthworms using standard test protocols to assess the toxicity of uranium were carried out. The project achieved already significant results showing toxicity in untreated acid mine drainage and residual waters and soils containing uranium for the earthworms and wildlife. First results were presented to international conferences and published already or are in press in international journals.

Marine radioactivity

F.P. Carvalho, J. M. Oliveira, M. Malta

Man-made, such as ^{137}Cs and $^{239+240}\text{Pu}$, and naturally-occurring radionuclides, such as ^{210}Pb and ^{210}Po , are introduced in the ocean by atmospheric deposition and coastal discharges. Research and monitoring of radioactivity in marine organisms was carried out in order to pursue the assessment of radiation doses from naturally-occurring and man-made radionuclides to biota and to the human population through ingestion of sea food. Monitoring of radionuclide levels along the Portuguese coast was performed using mussels (*Mytilus galloprovincialis*) as bioindicator organisms collected at various sites at the open coast and in the main estuaries. Determination of radionuclides in mussels, particularly the naturally-occurring ^{210}Po and ^{210}Pb allowed better understanding of the accumulation of these radionuclides by molluscs and allowed making relevant contributions to methodologies used worldwide in coastal monitoring programs using mussels.

Project “CAPTAR”

F.P. Carvalho

Participation in the web-based science education project coordinated by the University of Aveiro and funded by the Calouste Gulbenkian Foundation. The project aims at producing a scientific periodical (CAPTAR: ciência e ambiente para todos) available on line (<http://captar.web.ua.pt/>) for education and motivation of students and young researchers in environmental sciences. Two journal issues were released in 2011. Celebration of the centennial of Marie Curie Nobel Prize for the discovery of polonium and radium was comprehended in the activities of the year.

Radiocaesium Adsorption/Desorption on Geomaterials from “Raña” Deposits

M.J. Madruga, I. Paiva, E. Andrade, M. Gonçalves¹, A. Mateus²

The grain size fraction $< 63\mu\text{m}$ of the *raña* deposits matrix, comprising abundant clay minerals (smectite 15\AA , kaolinite, and illite) and quartz, besides minor amounts of iron hydroxides, was examined for radiocaesium (^{137}Cs) adsorption/desorption in order to test its use as liner/backfill/buffer for low and intermediate level waste (LILW) repositories. The ^{137}Cs adsorption was studied as a function of Cs^+ concentrations in the presence of Mg^{2+} (strongly hydrated competitive ion) using a batch method. The ionic strength of the magnesium solutions was adjusted to concentrations similar to those found in surface natural waters streaming in the region where the *rañas* were sampled. The simulation of *in situ* conditions for ^{137}Cs desorption in case of an incident/accident scenario considered two different procedures, static and dynamic. The ^{137}Cs adsorption is quite rapid, attaining the maximum saturation value after two weeks of contact time. Higher adsorption percentage values were obtained for lower Cs concentrations, in presence of Mg^{2+} . Desorption studies reveal that the solid/liquid equilibrium was more quickly attained in the static system (1-2 days) than in the dynamic system (around two weeks). In addition, the ^{137}Cs retention ranges between 60% and 70% in the static system, being just around 30% in the dynamic system. As expected, cation exchange between Cs and competitive ions is more efficient in the dynamic system. In both systems, the strongly hydrated ion Mg^{2+} , may lead to a wedge-effect in the FES pool, allowing a Cs penetration into the clayed-matrix and a consequent decrease in ^{137}Cs desorption levels. In the static system, the Mg and Cs concentrations in solution reach a dynamic equilibrium; consequently, no further ^{137}Cs is released without renewal of the solutions. Contrarily, in the dynamic system the renewal of Mg-solution leads to an equilibrium breakdown, leading to an increase of ^{137}Cs release. According to these experimental results, the clay fraction of *raña* deposits can be faced as a potential liner/backfill/buffer component to be used in LILW repositories.

¹ Fac. of Sciences/Univ. of Lisbon and CREMINER-LARSys

² Fac. of Sciences/Univ. of Lisbon and CeGUL

Total column and surface ozone variability over the Iberian Peninsula: Dynamical and chemical atmospheric factors - DYNOZONE

M. Reis, M. J. Madruga, E. Andrade, L. Silva

The DYNOZONE project was approved and funded by FCT. Is a cooperation project between New University of Lisbon, Nuclear and Technological Institute, Aveiro University and Fernando Pessoa Foundation for Teaching and Culture. The project officially started on October 2010. The main objectives of the project are to identify and justify, on a scientific base, the no photochemical origin of some ozone events, namely the stratosphere-troposphere exchange contribution to elevated surface ozone concentrations, as opposed to local production or long range transport from external pollution sources. During 2011, a ten years time series of naturally radioactive atmospheric tracers (namely ^7Be as a stratospheric tracer) were compiled and made available to the project team in order to correlate radionuclide and ozone data, providing a possible confirmation of the stratospheric influence on surface ozone high concentration events. A High Volume Sampler was acquired, as planned, to be installed in different locations and follow, on a daily basis, stratospheric intrusion events.

Building the pieces for a radioeco-toxicological characterization of the upper section of Tejo estuary from the Samouco shoals to the slimy site of Hortas

J.A. Corisco, L. Portugal, M. Almeida

During the decade 2000-2010 there has been observed an uncommon increase of bivalve catching in the left side of Tejo estuary, in the sedimentary planes extending from the village of Samouco upstream to the village of Alcochete. Native species *Scrobicularia plana* and specially the invasive species *Tapes japonica* appear to be the most prominent. The concerned area of interest has acquired in former decades a very distinct radiological status, from the fact that downstream, in the village of Barreiro, the phosphate industry of Quimigal – currently disabled – was a source of natural long lived radionuclides discharge in the environment. This project started with the intention of making a statement on the distribution of natural radionuclides in both biota and sediments in the area of interest, but quickly that interest has expanded towards the analysis of multiple pollution sources including heavy metals, once their presence resulting from human activities may contribute to a complex ecotoxicological scenario affecting human populations and environmental quality.

Field work has started in February 2011 by collecting biological (plants, bivalves) and sediment samples. Laboratory sample processing has been carried through ever since. High resolution gamma spectrometry of sediment samples has confirmed the presence of components of the natural series of ^{235}U and ^{238}U . Neutron activation analysis is being carried to complementary analysis of heavy metals both in sediments and biota. Scientific/technological partnership is being attempted and an encouraging meeting with a group from UNINOVA dedicated to environmental monitoring and robotics has opened a new perspective to this work. Project is still aiming for financial support, which will be attempted in 2012.

Accreditation of radioanalytical techniques

M.J. Madruga, I. Lopes, A.R. Gomes, J. Melo, J.A. Corisco

During this year a major effort was devoted to the final steps of the preparation for the accreditation. Improvements were introduced in the management and technical procedures and an internal audit by UPSR auditors was carried out. In December 2011, accreditation concession audit, according to the ISO/IEC 17025 requirements, was performed by two auditors of the Portuguese Accreditation Body (Instituto Português de Acreditação-IPAC) for the three techniques, gross alpha/beta in waters with proportional counters and Liquid Scintillation Counting (LSC) and tritium determination in waters by LSC. During the audit a systematic and independent examination was carried out to determine whether quality activities are implemented effectively and are suitable to achieve the objectives. The audit report highlights the good quality of the work developed, the experience, technical training and the spirit of openness to improvement of the staff involved.

Three papers concerning the quality assurance of these radioanalytical techniques were published at LSC 2010, Advances in Liquid Scintillation Spectrometry. Radiocarbon, Editor: P. Cassette, ISBN: 978-0-9638314-7-7, University of Arizona, Tucson, Arizona, USA. In the framework of the IAEA TC project, RER/ 0/ 031 - “Strengthening Sustainability of Nuclear Research and Development Institutes in the Modern Science and Technology Environment” one GRA staff member (Irene Lopes) has performed a scientific visit (two weeks) to the Central Mining Institute (Poland) taking knowledge of the implementation in this Institute of the Quality System, according to the standard ISO/IEC17025. Training in Sr-90, Ra-226/Ra-228, Rn-222 and uranium analysis in waters, using liquid scintillation counting technique and alpha spectrometry was also performed.

Radon in waters

M.J. Madruga, I. Lopes

Radon (^{222}Rn) is a radioactive naturally occurring gas, derived from the natural uranium series (^{238}U) and decay to short lived daughters (^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po) with a high solubility in waters. The highest activities of ^{222}Rn are found in groundwaters due to its emanation from granite rocks. The consumption of waters with high levels of radon could give a high contribution to the effective dose from ingestion affecting the human health. So, it is important the knowledge of the radon levels in waters. In this context, determination of ^{222}Rn by Liquid Scintillation Counting (LSC) technique is ongoing and an intercomparison exercise in collaboration with STUK institute (Finland) was performed during this year, with good results. A collaborative protocol with Instituto Superior Técnico (IST) laboratories is envisaged for the radon analyses in Portuguese waters.

Radionuclide Particulate Station RN53 of the International Monitoring System for the Verification of the Comprehensive Nuclear Test Ban Treaty (CTBT)

M. Reis

The radionuclide particulate station RN53, installed in São Miguel - Açores, belongs to the International Monitoring System (IMS) network for the verification of Comprehensive Nuclear Test Ban Treaty (CTBT), signed and ratified by the Portuguese State. The station is managed by ITN, with the collaboration of two local operators from the Meteorological Institute. The station was certified on December 2010 and, since then, was providing data to the International Monitoring System of the CTBT. Between 25 of March and 16 of May, the station registered some isotope detections (iodine and cesium) related to the Fukushima NPP accident. On October 2011, one of the station operators participated in a training course financed by CTBTO: "Technical Training Programme (TTP) for Radionuclide Station Operators with ORTEC Equipment", held at Oak Ridge, USA.

Services**Radioactivity in drinking and mineral waters**

M.J. Madruga, J. Melo, A.R. Gomes, A. Libânio, I. Lopes, F.P. Carvalho, J.M. Oliveira

Regarding the evaluation of the radioactivity levels in drinking waters (Decree-Law n°306/2007) the GRA was requested by Water Suppliers to carry out the determinations of global alpha, global beta, Tritium, ^{238}U , ^{234}Th , ^{226}Ra and ^{210}Po and the Total Indicative Dose parameter in waters. The determination of ^{222}Rn in water samples was also carried out. In order to license mineral waters trade (Decree-Law n°84/90) several enterprises request a radiological study. During 2011, a total of about 300 analyses were performed.

Radioactivity in foodstuff and biological samples

M.J. Madruga, I. Lopes, A. Mourato, F.P. Carvalho, J.M. Oliveira, M. Malta

Analyses of ^{90}Sr in foodstuff and ^{210}Po in biological samples were carried out, under request of external entities, in order to support commercial activities and research projects.

Indoor Radon

M. Reis, E. Andrade, L. Machado

Following the National System of Building Energetic Certification for the Indoor Air Quality (Decree-Law 78/2006, 4th April) public and private enterprises request to GRA the measurements of indoor radon. Besides, since November 2003 a collaborative Protocol was established between UPSR-ITN and DECO to answer the associate's indoor radon requests. In 2011, about 750 measurements were performed.