# **Environmental Radioactivity**

# Maria José Madruga

Under Articles 35 and 36 of the EURATOM Treaty, Portugal has an obligation to conduct yearly a national radiological environmental survey. This survey was established in the Decree-Law 138/2005 of 17th August and its execution legally attributed to ITN. Monitoring The Radiological Environmental the Programmes have been performed by Environmental Radioactivity Group in collaboration with the Measurement Laboratories (LM), the Dosimetry and Radiobiology (GDR) and the Radiological Protection and Radioactive Waste Management (GRRR) Groups. To carry out this programme a considerable effort of the group has been made in terms of human resources (about 75% of the time consumed). This programme involves a financial cost of about 348 000 €.

In the framework of the EURATOM Treaty Article 35 the group has been participating in the elaboration of the European Atlas of Natural Radiation. Our contribution consisted in the treatment of the Portuguese indoor radon data to be included in a grid defined by the EC. The processed data were sent to the JRC and a preliminary European map was already delivered.

Current research activities are ongoing to investigate the levels of radioactive contamination in the atmosphere (aerosols) aquatic and terrestrial environments. In the framework of the FCT Project KADRWaste, coordinated by GRRR, the Group participated in the characterization of geomaterials to radiocesium adsorption.

Improvements have been introduced in the technique for alpha/beta measurements in drinking waters using proportional counters and liquid scintillation counting. Quality control tests of recovery and accuracy were performed.

The group participated in coordination with the UPSR/Measurement Laboratory in three international inter-comparison exercises concerning drinking water samples, organized by the European Commission, Institute for Reference Materials and Measurements (IRMM, EC-Joint Research Centre, Belgium), by the Lab. de Radiactividad Ambiental, Universidad de

# **Research Team**

### Researchers

M. J. MADRUGA, Princ., Group Leader F. P. CARVALHO, Princ. M. J. REIS, Aux. J. A. CORISCO, Aux.

#### Students

A. R. GOMES, FCT grant H. FONSECA, FCT grant J. MELO, FCT grant Extremadura, Cáceres (Spain) and by Institute de Radioprotection et de Sûreté Nucléaire (IRSN), France. The results published in 2008 were in good agreement/compatible with the reference values.

The construction of the infrastructure for housing the Radionuclide Particulate Station (RN53) at S. Miguel, Azores, was concluded last year. The installation of the CINDERELA station and all the related equipment (detector, meteo station, VSAT antenna, etc) was also concluded. A new calibration of the detector with a fresh source was carried out during 2008. A few problems related to the performance of the detector on the low energies region come up resulting in the need of several interventions during 2008. A last intervention is planned for the beginning of the next year and afterwards the station will be ready for certification. This station will be part of the International Monitoring System, established in the framework of the CTBT (Comprehensive Nuclear Test Ban Treaty).

The technical services developed by the group are carried out under contract with companies or, by request from enterprises or Government organizations. Some of these technical services are: the evaluation of the radioactivity levels in public water supplies (Decree-Law n°306/2007) and mineral waters, the radioactivity analyses of foodstuffs and indoor radon measurements. The group income of these technical services was about 31 000  $\boldsymbol{\epsilon}$ .

During the current year the majority of the group members have been actively involved in the improvement of the quality system following the ISO 17025 requirements in order to achieve the laboratories accreditation.

One of the tasks of the group is the education and training of staff and young students. During this year two MSc theses were concluded and one was submitted. A member of the group has finished his dissertation (Ph.D. equivalent) was promoted to Researcher.

### **Technical Personnel**

J. M. OLIVEIRA (1ª) (70%) A. LIBÂNIO A. MOURATO

### Collaborators

I. LOPES M. M. MALTA

# **Radiological Environmental Monitoring Programmes**

M.J. Madruga, F.P. Carvalho, M. Reis, N. Pinhão, J.G. Alves, J.A. Corisco, R. Trindade, J.M. Oliveira, A. Libânio, A. Mourato, G. Silva, L. Portugal, I. Lopes, J. Abrantes, L. Silva, L. Torres, M. Malta, A.R. Gomes, G. Carvalhal, H. Fonseca, J. Melo, M. Pereira, A. Batista

# **Objectives**

The Article 35 of the EURATOM Treaty stipulates that "each Member State shall have the facilities necessary to carry out the continuous monitoring of the levels of radioactivity in the air, water and soil and to ensure the compliance with the Basic Safety Standards". The Article 36 of the same Treaty stipulates that "the appropriate authorities shall periodically communicate information on the checks referred to in Article 35 to the Commission so that it is kept informed of the level of radioactivity to which the public is exposed".

Radiological Environmental The Monitoring Programmes planned according to the Articles 35 of the EURATOM Treaty requirements include the Programme at a National Level, established by law in 2005 and the competency of its execution 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 main goals of the radiological environmental survey are to determine the artificial and natural radionuclide levels in environmental compartments (aquatic, terrestrial and atmospheric environments) considered as direct pathways of contamination to man and to evaluate the external radiation levels in order to provide information for the assessment of the potential exposure of the Portuguese population.

The monitoring programme carried out for the ITN *campus* includes measurements of external radiation levels (gamma dose) and measurements of radionuclide activity concentrations in relevant environmental samples (aerosols, rain water and soils). The main objectives of the programme were to verify the compliance with the authorized discharge limits, to check the operation conditions and discharge controls adequacy, to maintain a continuing record on environmental radionuclide levels related to the sources under consideration and to provide information for the public.

The environmental radioactivity monitoring for the regions around old uranium mining sites was implemented in the regions of Viseu and, around the former uranium mining and milling sites of Urgeiriça, Quinta do Bispo, Cunha Baixa, Murtórios, and Barracão. For this purpose three field missions were performed to carry out field measurements and sampling of aerosols, water, soils and plants. Samples were analyzed in the laboratory for the main descendants of uranium by alpha spectrometry. Particular attention was paid to water and agricultural used in human diet in order to assess the exposure of local population to food chain transfer of radionuclides.



# Results

During 2008 about 800 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 2700 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 thermoluminescent dosemeters were also performed. The results show that the Portuguese population was not exposed radioactive to 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 (1) 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).

# **Published work**

M. J. Madruga, F. P. Carvalho, M. Reis, N. Pinhão, J. Alves, J. A. Corisco, R. Trindade, J.M. Oliveira, A. Libânio, A. Mourato, G. Silva, L. Portugal, I. Lopes, L. Silva, J. Abrantes, L. Torres, A.R. Gomes, G. Carvalhal, H. Fonseca, J. Melo, M. Pereira, A. Batista. Programas de Monitorização Radiológica Ambiental (Ano 2007). *Internal Report* DPRSN, Série A, n°32/2008, ISBN 978-972-8660-33-8, Depósito Legal 194022/03, pp. 147.

## Biomass Combustion and Release of Radionuclides into the Atmosphere

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

A collaborative project with University of Aveiro, funded by FCT, was started to evaluate the release into the atmosphere of several contaminants including organic substances, toxic metals and radionuclides. The extension of forest fires in Portugal as well as around the entire Mediterranean basin is a non negligible source of toxic substances and contaminants, including natural radionuclides and artificial radionuclides deposited on surface soils following the Chernobyl accident. Several experimental approaches were assessed including measurements during true forest fires, measurements during small scale programmed fires, and laboratory burning of biomass. A prescribed forest fire in the mountains, near Góis, was jointly organized with Escola Agrária de Coimbra and Serviços Florestais to allow for field measurements.

# Development and Validation of Radioanalytical Methods using Liquid Scintillation Counting (LSC) Technique

# M.J. Madruga, I. Lopes, J. Melo

The Liquid Scintillation Counting (LSC) is a technique widely used for measuring radiation from beta and alpha emitting radionuclides. Following the application of this technique, at the GRA Laboratories, to the determination of <sup>90</sup>Sr in milk based on the use of ion exchange chromatography (Sr resin, Eichrom), validation trials were performed in order to implement the same method to foodstuffs samples. The recovery of the method to these matrices, defined as the ratio of <sup>90</sup>Sr activity measured and <sup>90</sup>Sr activity added, was evaluated and a recovery value of 88% was obtained. This methodology was tested to foodstuffs samples within the radiological environmental monitoring survey. In the framework of the quality control and in order to improve the global alpha and global beta measurements by LSC, calibration and recovery tests in waters were also carried out.

# Transport and Accumulation of <sup>137</sup>Cs and <sup>90</sup>Sr by the Unicellular Microalgae *Chlamydomonas reinhardtii* Dangeard

# J.A. Corisco, J. Fernandez<sup>1</sup>

The present state of the art in the field of ion transport supports the hypothesis that  ${}^{137}Cs^+$  and  ${}^{90}Sr^{2+}$  free ions in water are accumulated by the unicellular microalgae C. reinhardtii, as a result of a transmembrane transport mediated by the mechanisms involved in the transport of  $K^+$  and  $Ca^{2+}$  ions through the plasma membrane. Experimental objectives were (i) to verify the effects of external concentrations of  $K^+$ ,  $Ca^{2+}$ ,  $Na^+$  and  $H^+$  on the transport and accumulation of  ${}^{137}Cs^+$  and  ${}^{90}Sr^{2+}$ ; (ii) to characterize the uptake kinetics of  $Cs^+$  and  $Sr^{2+}$  under deficiency and sufficiency of  $K^+$ ; (iii) to verify the effect of  $K^+$  and  $Ca^{2+}$  channel blockage with specific inhibitors, namely tetraethilamonyum (TEA<sup>+</sup>) and  $La^{3+}$ ; and (iv) to verify the effect produced by the inhibition of plasma membrane H<sup>+</sup>-ATPase with cyanide (CN<sup>-</sup>). The obtained results reveal the presence of two different mechanisms mediating the transport of <sup>137</sup>Cs<sup>+</sup>. An active uptake mechanism with Michaelis-Menten type kinetics and a relevant sensibility for pH is induced by  $K^+$  deficiency, whereas under  $K^+$  sufficiency  ${}^{137}Cs^+$  undergoes a passive transport through ion channels. Such observations support the hypothesis that  $K^+/H^+$  co-transporters and  $K^+$  channels are mediating the transport of <sup>137</sup>Cs<sup>+</sup> in *C. reinhardtii*, according to the external K<sup>+</sup> regime. <sup>137</sup>Cs<sup>+</sup> concentration factor (CF) reached the value 48388 in K<sup>+</sup> deficiency and 4568 in K<sup>+</sup> sufficiency. Uptake of  ${}^{90}$ Sr<sup>2+</sup> is likely to be mediated by  $Ca^{2+}$  channels, with more intense fluxes through channels activated by hyperpolarization of the cell membrane, when external  $K^+$  concentration is within the micromolar range. The observed  ${}^{90}\text{Sr}^{2+}$  CF for cells in K<sup>+</sup> deficiency reached the maximum value of 10755 in an alkaline medium of pH=8.3. Cells under K<sup>+</sup> sufficiency showed maximum  ${}^{90}\text{Sr}^{2+}$  accumulation (FC=2309) in a medium with physiological pH=7.3. It is likely that changes in pH to more acidic and alkaline values intensify the extrusion of  ${}^{90}$ Sr<sup>2+</sup> via the Ca<sup>2+</sup>-ATPase, thus reducing the value of FC.

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### Marine Radioactivity F.P. Carvalho, J. M. Oliveira, M. Malta

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. Monitoring of radionuclides along the Portuguese coast was performed using mussels as bioindicator organisms collected at various sites on the open coast and in the main harbours. Determination of radionuclides in mussels, fish and marine mammals (dolphins) was used to compute radiation doses to biota and it is part of a Master Thesis (M Malta).

### **Indoor Radon Risk Mapping**

M.J. Reis, H. Fonseca

A new approach is being implemented in order to obtain indoor radon risk maps by using geostatistic simulation techniques. Besides the radon data already available for Portugal, new data were processed. Indoor radon distribution maps were developed for Portugal, namely prediction and probability maps (for example the probability map of exceeding 400 Bq m<sup>-3</sup>). After data processing several methods will be investigated for the establishment of risks categories and radon mapping. Geographical Information Systems (GIS) techniques are being used to integrate radon concentrations, geological and administrative maps, buildings locations and other useful information. In collaboration with the REM (Radioactivity Environmental Monitoring) group of the JRC (Joint Research Centre) a European Atlas of Natural Radiation, including radon mapping is being developed. The Portuguese indoor radon data was processed and transformed in order to fit the European grid defined by the JRC. A preliminary European indoor radon map was constructed and presented in the *9th International Workshop on the Geological Aspects of Radon Mapping* (as part of the 33<sup>rd</sup> International Geological Congress), held in Oslo in August 2008.

### SERVICES

### 1. 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 UPSR was requested by Water Suppliers to carry out the determinations of global alpha, global beta, Tritium, <sup>238</sup>U, <sup>234</sup>Th, <sup>226</sup>Ra and <sup>210</sup>Po and the Total Indicative Dose parameter in waters. The determination of Radon in same water samples was also carried out. To license the mineral waters trade an evaluation of its radioactive levels should be performed (Decree-Law n°84/90). The radiological study included analyses of <sup>226</sup>Ra and global beta. Several enterprises often request this radiological study.

During 2008, a total of about 450 analyses were performed.

### 2. Radioactivity in Surface and Ground Waters

*M.J. Madruga, J. Melo, A.R. Gomes, A. Libânio* The alpha/beta global activities and the tritium levels in water samples were determined within FCT-KADRWaste project in collaboration with GRRR group.

### 2. Indoor Radon

### M.J. Reis, H. Fonseca

By request of public and private enterprises indoor radon measurements were performed in buildings. Since November 2003 a collaborative Protocol was established between UPSR-ITN and DECO to answer the associate's indoor radon requests. A total of 120 measurements were performed during this year.