Radiological Protection, Environmental Radioactivity and Nuclear Safety

The Department of Radiological Protection and Nuclear Safety (DPRSN) of ITN is the only scientific department in the country with technical capability, experience and know-how in radiological protection. The department ensures the monitoring of radioactive contamination in food and environment, the monitoring of radioactivity in the uranium mining regions, the dosimetry of workers professionally exposed to radiation and radioisotopes, and the safety control of facilities, radiation emitting equipment and radioactive sources.

Research and Development

This Department of ITN primarily carries out research and development in the fields of:

- Environmental radioactivity
- Metrology and dosimetry of ionising radiations
- Nuclear safety and safety of nuclear applications

The activities in the field of *environmental radioactivity* relate with the dispersal, behaviour and fate of radionuclides in the environment. In particular, for the radiological protection of the Portuguese population, the research encompasses the study of radionuclides in terrestrial, aquatic and atmospheric environments, with special emphasis in the food chain transfer.

Metrological activities in the field of ionising radiations are carried out in the Metrological Laboratory of Ionising Radiation and Radioactivity (LMRI). The main objectives of LMRI are the following:

- Keep the national standards and assure their international traceability, under a contract with the Portuguese Institute of Quality;
- Research and development in the field of metrology and dosimetry of ionising radiation;
- Co-operation with EUROMET, AIEA and EA in the field of research, intercomparison of standards and accuracy of measurements;
- Co-operation in research activities of external teams through availability of irradiation beams and dosimetry;
- Technical services of metrological control (calibration, type testing, etc.) of measuring instruments.

The main line of activity in Physical Radiation Dosimetry concerns the evaluation of the radiation dose to the Portuguese population, and conceals both research and service activities.

Biological dosimetry or biodosimetry measures the effects of radiation exposure on biological organisms. The goal of biodosimetry, when the exposure is known is to quantify how an exposure is distributed within an organism, or when the exposure is not known, assess the dose from observation of the organism.

Exposure to ionising radiation may also induce genetic damage and biochemical modifications in tissues and at sub-cellular level.

The Department has not carried out any relevant research activity in the area of *Nuclear Safety and Safety of Nuclear Applications* during 1999. However, several routine activities such as the radiological surveillance of the ITN *campus*, and a number of services related to safety of nuclear applications, transport of radioactive materials, and radioactive waste management, were delivered.

Technical services

The department provides to the country a wide variety of technical services. These services are often performed under contract with companies, under request from other Institutes or Government organisations, and in support to emergency action of Fire Brigades, Civil Protection and Customs.

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Some of the more frequently asked services are:

- radioactivity analysis of water samples
- radioactivity analysis of goods and foodstuffs to export
- dosimetry of radiation workers of hospitals, clinics, industry, etc.
- calibration of equipment used in medicine or industry to measure radiation
- measurement of radon in indoor atmosphere
- safety assessment of facilities and equipment producing or using radiation
- specialised medical control of radiation workers
- training of personnel in radiological protection.

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Environmental radioactivity

Parametrisation / validation of submodels for the transfer of radiocaesium and radiostrontium through the trophic chain under well-defined laboratory conditions*

M.C.V. Carreiro, J.A.G. Corisco, G. C. Ferrador, C.E. Costa

Objectives

The main objective is to obtain experimental data to validate the mechanistic concepts developed by other participants, for the transfer of radionuclides in critical parts of the aquatic food chain, under different environmental conditions. This objective should be carried out in two parts:

Ist part: To study the membrane transport mechanisms for radiocaesium by a representative phytoplankton species, the freshwater green microalgae *Chlamydomonas reinhardtii*.

2nd part: To study the effect of environmental variables on the uptake, accumulation and elimination of radiocaesium and radiostrontium in freshwater plants and fish. The special target is the contamination of common carp, *Cyprinus carpio*, via the hydrophyte *Lemna minor* (duckweed).

In addition some other studies were carried out with *Lemna minor*: effect of different external potassium concentrations and two temperature regimes on radiocaesium accumulation and elimination; caesium kinetics under K^+ sufficiency and deficiency; and radiostrontium accumulation and elimination under K^+ sufficiency and deficiency.

1st part: Study of caesium transport mechanisms by the microalgae *Chlamydomonas* reinhardtii

J.A.G. Corisco's PhD Thesis in progress

Results and discusion

The effect of K_e^+ on the elimination of ^{137}Cs by the algae, can also be expressed by the variation in the biological half-life Tb_i , which is the time needed for a 50% elimination of the radioisotope. For the lowest K_e^+ (<0.1 mM), a single exponential decay in algae radioactivity was observed. The correspondent half-life Tb_1 ranged from 122 h (1µM K_e^+) and 198 h (5 µM K_e^+) to 57 h (10µM K_e^+). Under K_e^+ sufficiency (\geq 0.1 mM), experimental data could be fitted to a sum a two exponential decays. For each phase, fast decay and slow decay, the correspondent half-lives are represented as Tb_1 and Tb_2 . Tb_1 ranged from 7 h (0.1 mM K_e^+) to a stable value of 2 h (K_e^+) 1 mM). Tb_2 ranged from 182 h (0.1 mM K_e^+) to 50-60h (5-10 mM K_e^+).

The effect of external pH in the uptake and accumulation of 137 Cs by Chlamydomonas adapted to either deficiency (def K⁺) or sufficiency (suf K⁺) of external K⁺ is shown in Figures. At very low (1µM) external K⁺, uptake rate (V₀) and concentration factor (CF) reached maximum values (V₀=160 Bq cm⁻³h⁻¹, CF=3900) at pH 7, while for a more acid or a more alkaline treatment, both variables decreased. Under K⁺ sufficiency, variations of external pH in the range 5 to 9 do not cause a significant change in V₀ and either in CF for 137 Cs.

The response of the uptake rate to pH in *Chlamydomonas*, agrees with those quoted for *Riccia fluitans* (Heredia, 1998), in which the threshold for the distinction of two inhibition effects of the carrier mediated cotransport of 137 Cs with protons (H⁺), was pH7.5. For *Chlamydomonas* that threshold is pH7. As in *R.fluitans*, for pH<7 the reduction of uptake is less intense then for pH>7. Under a more acid environment, it is possible that the protonation of the carrier causes a conformational change that reduces its affinity for Cs⁺ and, as a consequence, the transport efficiency. When pH of the external medium is more alkaline than pH7, the inhibition of the uptake rate and, in addition, of CF is probably due to the diminution of the electrochemical gradient for H⁺, the driving force for 137 Cs⁺ transport in K⁺ deficiency.

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^{*} EU Contract FI4-CT95-0018, 2 × 10⁶ PTE, 1997-1999.

2nd part: Effects of environmental variables on uptake, accumulation and elimination of radiocaesium and radiostrontium in aquatic plants and fish

M.C.V. Carreiro, G. C. Ferrador

2.1 Study of Cs⁺ kinetics in *Lemna minor* according to the Michaelis-Menten model for competitive inhibition of Cs⁺ uptake by K⁺

Results and discussion

- a) Results are in agreement with the existence of two different kinds of K⁺ transport systems, as two different Cs⁺ uptake kinetics were found: at low K⁺ concentrations there is an active transport, carrier mediated and kinetics is saturable, being fitted to Michaelis-Menten model; under K⁺ sufficiency Cs⁺ transport is done through K⁺ channels by diffusion and kinetics is almost linear.
- b) No significant differences were observed under K⁺ sufficiency and deficiency.
- c) Both sufficiency and deficiency show a saturation kinetics in the range of 1-80 µM of Cs⁺.
- d) K_m for *Lemna* is 9.90μM, meaning a high affinity kinetics.
- e) V_{max} for Lemna is 27.3 nM g⁻¹ h⁻¹.
- f) Points b), d) and e) seem to suggest that both concentrations are of K⁺ deficiency for *Lemna*, therefore, this hydrophyte probably needs more K⁺ to survive.
- g) Lemna is closer to the rooted plant types (rizophytes).

2.2 Radiostrontium transfer from the hydrophyte *Lemna minor* (duckweed) to the Cyprinid fish *Cyprinus carpio*. Uptake and elimination kinetics

Results and discussion

The TTF evaluated for radiostrontium, through plants pathway, using an empirical model is low, 0.027. It is approximately 10 times lower than we obtained for radiocaesium, what might be explained as fish radioactivity measurements are made in whole body and strontium is preferentially accumulated in bones. No similar experiments were so far found in the literature, so no comparison is possible.

The Biological Half-life was found to be 58 days, which is lower than the majority of the results in bibliography. It may very likely be inputed to the relatively high Ca²⁺ concentration in the water, about 48 mg l⁻¹, which may promote an ionic exchange.

The same modelling approach already referred, the pharmacokinetic model (SAAM II) was used and also a good fit of experimental data was obtained.

The trophic transfer factor evaluated according to this dynamic model is 0.0216. It is again interesting to notice that the previous estimation is not very different of this value.

From these two cases it is suggested that the pharmacokinetic model can describe and predict radiocaesium and radiostrontium accumulation from food and elimination over the time.

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European Research on Radon in Construction Concerted Action (ERRICCA)*

Scivyer, C.¹, Stadtmann, H.², Steinhausler, F.³, Poffyn, A.⁴, Delmotte, C.⁵, Hulka, J.⁶, Andersen, C.⁷, Arvela, H.⁸, Kokotti, H.⁹, Robé, M.¹⁰, Hamel, P.¹¹, Simopoulos, S.¹², Phaff, H.¹³, Dollard, B.¹⁴, Patania, F.¹⁵, Belli, M.¹⁶, Graff, E.¹⁷, Skowronek, J.¹⁸, Ciesla, K.¹⁹, Faisca, M. C., Mocsy, I.²⁰, Jovanovic, P.²¹, Poncela, L.²², Akerblom, G.²³, Piller, G.²⁴, Miles, J.²⁵.

Objectives

Specific objectives were set in order to:

Develop and improve the application of the methods to reduce radon levels in buildings and examine their durability over time;

Consider developments in building design and construction with particular reference to new buildings including the problem of radon emissions from materials;

Consider how legal/buildings codes can be developed to enhance building protection to reduce indoor radon levels;

Improve and apply models for radon entry and transport, on issues related to the basic understanding of the problem in particular improved remedial measures;

Develop new ideas for collaborative research proposals.

Results

Several kinds of usual building materials continued to be analysed in order to evaluate their natural radioactivity levels (232 Th, 226 Ra, 40 K) as well as their radon exhalation rates. From the obtained results it can be conclude that radon exhalation rate can be evaluated through the respective 226 Ra concentration (gamma spectrometry).

Special attention was given to granites and a radon mitigation study was performed.

Phosphogypsum mixtures having a no radioactive compound (different %), were studied in order to find a final phosphogypsum mixture able to substitute natural gypsum. It was verified that mixtures with fhosfhogypsum until 20% in the mixture have a low radon emission.

Several flooring materials were contuined to be tested in order to select the most apropriated that could be used as radon barrier.

Conclusions are presented in "Different Studies on Building Materials", oral comunication at the workshop "Radon in the Living Environment", Athens, 19-23 April, 1999.

Concerning a new house to be built in a granitic and in a radon prone region, a radon prevention study was performed in order to advise the owner about the selection of building materials to be employed. Some other radon preventive modifications were advised as well as the implementation of a tested radon barrier (on the soil beneath the house).

Integrated indoor radon measurements were carried out along the year and a mean value of 85 Bq m⁻³ was obtained.

Under this contract two internatinal calibrations were carried out. An international calibration of radon passive radon detectors, coordinated by the NRPB, beeing the other one a calibration concerning the radon exhalation from a building material, coordinated by the Technical University of Athens.

Further Work

ERRICCA finished at the end of June 1999.

To bring together scientists and technologists working across Europe on radon, attempts are running in order to continue this project as a "ERRICCA 2" (European Radon Research and Industry Collaboration-Concerted Action).

This new project would be set up with the principal aim of transferring existing scientific knowledge to industry, to identify areas where further research is required and to enable science and industry to work in partnership.

EU Contract N°F14PCT 960064. Only travel support. For Partners please see ERRICCA (9. Contracts).

Concerted action to establish and run a user group for the EC computer system PC-CREAM*

R.Trindade, A.M.Brogueira, M.Reis

Objectives

Promote the wider use of PC CREAM in the EU thus contributing to a more harmonised approach in assessing the radiological impact of effluent discharges and to achieve common safety standards.

Identify where further developments of PC CREAM are required to ensure that it remains "state-of-the-art".

Suggest improvements to the default data provided with PC CREAM based on recent experimental research.

Results

As a result of the model tests performed by the different PC CREAM model users some suggestions were made to the developers during the group meetings.

The treatment of radionuclide progeny is not consistent throughout the PC CREAM suite. It was suggested to include at least one radioactive daughter for all nuclides that are part of a decay chain and consolidate the treatment of progeny between the different components of PC CREAM.

The default list of radionuclides included in the models package has some omissions. Additional radionuclides should be included for any future development of PC CREAM, for example radionuclides commonly used by hospitals and research establishments.

A greater number of stacks, stack heights and receptors for atmospheric assessments should be possible in the future.

RESUS (resuspension model) could be altered to allow the user to change the resuspension factor, and expanded to include a simple dust loading model to account for man made and mechanically generated resuspension over short time periods.

In many regions of Europe, including Portugal, the irrigation of crops with river water is a common practice, and as a result is a possible public exposure pathway. Such an addition to the river module would require integration of the river and FARMLAND (radionuclide transfer in the foodchain model) models, and perhaps some additional investigation into the applicability of some model parameters in irrigation situations.

Atmospheric dispersion model should allow greater flexibility in the definition of the default weather category wind speeds and increase the range of roughness lengths in atmospheric dispersion modelling.

It is desirable to develop the ability of PC CREAM to interact with GIS systems.

Further work

As well as improving the models currently included in the package it might be useful to include new models.

It is intended to draw up a comprehensive questionnaire on future development priorities for PC CREAM which will circulate through all group members.

^{*} EC Contract F14P-CT0074. Only travel support.

Aguifers and Surface-Waters in the Chernobyl Area- Observations and Predictive Evaluation*

J. Smith¹, A. Konoplov², R. Comans³, A. Kudelsky⁴, G. Laptev⁵, M.J. Madruga⁶, G. Zibold⁷

Objectives

The global objectives of this project are: to investigate long-term radiocaesium and radiostrontium dynamics in rivers, lakes and groundwaters; to develop simple, predictive, physically based and empirical models for prediction of long-term radiocaesium dynamics in these systems; to investigate the long-term dynamics of radiocaesium solid/water distribution in suspensions of soils, sediments and clay minerals as a function of the water phase chemical composition.

Results

In order to accomplish the project a set of laboratory experiments of sediments characterization was carried out at the DPRSN, for sediments from the Chernobyl area and Tejo river (Portugal). The characterization consisted mainly on the determination of the frayed edge sites (FES) capacity and radiocaesium interception potential (RIP). The methods used are described in Madruga (PhD thesis, 1993). However, when these methods were applied for organic soils and bottom sediments we faced with same problems. For instance: the plateau on sorption isotherm could not be reached; the position of maximum was observed at different caesium concentrations and depend on substrate. So, another approach was used to calculate the FES. It is known that sorption isotherm with saturation is described by Langmuir Isotherms. The intercept of the dependence inverse concentrations, corresponds to the inverse value of FES capacity. Moreover taking the initial range of the isotherm, at low caesium concentrations, the capacity of high affinity sites (HAS) can be calculated. The results for a set of sediments are presented in the next table.

of a set of sediments are presented in the next table.							
Sediments	(Madruga, 1993)	Langmuir Isotherm		RIP (K)	$RIP_{ex}(K)$	RIP	RIP_{ex}
						(NH_4)	(NH_4)
	FES	FES	HAS				
	meq kg ⁻¹	meq kg ⁻¹	Meq kg ⁻¹	meq g ⁻¹	meq g ⁻¹	meq g ⁻¹	meq g ⁻¹
T1	4.1±0.7	3.7	0.29	0.91	0.33	0.2	0.12
			(7.9%)				
A	9.9±2.7	8.6	0.63	-	-	-	-
			(7.4%)				
S	10.6±1.8	9.7	0.44	6.4	2.2	0.9	0.5
			(4.5%)				
SV5	-	0.14	0.03	0.09	0.03	0.02	0.007
			(21.4%)				
SV2	5.5±0.7	6.6	1.0	-	-	-	-
			(15.2%)				
SV(C2)	2.0±0.1	2.3	0.5	-	-	-	-
			(21.7%)				

It can be seen that FES values obtained from the Langmuir linearization are similar to those calculated from the radiocaesium isotherm plateau. This will be due to the low organic matter content of the sediment samples. The HAS presented values which correspond to about 5 to 20% of the FES. The exchangeable RIP values using ammonium acetate extraction atr lower than those corresponding to the RIP.

Further work

The characterization of sediment samples from the Chernobyl area will be continued. Radiocaesium extraction from sediments using two different protocols and long-term measurements of rates of radiocaesium and radiostrontium sorption to sediment samples will be carried out.

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^{*} Funding: EC Contract ERB IC15-CT98-0205 (240.000 ECU), 2.2 × 10⁶ PTE, for ITN during 1998-2001.

¹IFE (United Kingdom),

²SPA "Typhoon" (Russia),

³ECN (Netherlands),

⁴IGS (Belarus),

⁵UHMI (Ukraine),

⁶ITN/DPRSN (Portugal),

⁷Fachhochschule-Weingarten (Germany)

Environmental Impact of the Uranium Mill Tailings*

M.J. Madruga, A. Brogueira, I. Faria¹, C. Pires², C. Costa

Objectives

The main objectives of this project are: to evaluate the environmental impact of the natural radionuclides ²²⁶Ra and ²¹⁰Pb from *Urgeiriça* uranium mill tailings in what concerns its dispersion in the environment and transfer to plants, which are growing in the tailings; to investigate the radium speciation in the solid wastes.

Results

Solid wastes, soils and plants (*Eucalyptus globolus, Pinus pinea* and *Cytisus s.p.*) samples were collected at the *Urgeiriça* mill tailings and surrounding area. ²²⁶Ra and ²¹⁰Pb activities of solid wastes, soils and plants (aerial part and roots) were determined by gamma spectrometry. Solid wastes and soils were characterized in terms of cation exchange capacity (CEC) and the exchangeable cations determined by ionic chromatography [1]. The pH, organic matter content and exchangeable radium were also determined for solid wastes and soil samples. Results show different behaviour concerning the ²²⁶Ra uptake by the plants. For Eucalyptus globolus and *Pinus pinea* (trees) ²²⁶Ra concentration ratios decrease at low radium concentrations in the solid wastes and appear relatively constant at higher radium concentrations. For Cytisus s.p (shrubs) concentration ratios increase at higher solid waste concentrations approaching a saturation value. The exchangeable radium soil/plant tansfer factors are one order of magnitude higher than those calculated for total radium. For the plants studied the soil/roots transfer factors were higher than those obtained to the aerial parts [2] [3]. The ²¹⁰Pb soil/plant transfer factors are, in general, lower than those obtained for radium [3]. Comparative studies between the exchangeable radium transfer factors on plants groups and sites seem to indicate no significant differences among the different dams studied [2]. It is verified that transfer factors increase with pH, decrease with organic matter content and have two different behaviours with CEC, i.e., decrease for lower CEC values and remain constant for higher CEC values [3]. Results concerning the sequential radium extraction from solid wastes indicate that the larger percentage of radium is bounded on a non-exchangeable form and so no easily available for plant sorption [3]. Studies of ²²⁶Ra and ²¹⁰Pb vertical distribution at two dams show similar behaviour to these radionuclides, being however, slightly higher the²¹⁰Pb activity at the upper layer.

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Further work

The study of 210 Pb soil/plant transfer factor and radium speciation in solid wastes will be continued. Exchangeable and total cations (K⁺, Ca²⁺ and Mg²⁺) will be determined in solid waste and plants samples by ionic chromatography.

Geochemical characterisation and contamination levels on the major and trace elements in particular, heavy metals associated with this mineralization area will be carried out at the Chemistry Department of the ITN (responsible M.F. Araújo). This study aim at the evaluation of the dispersal pathways and transfer mechanisms on the soil/plant system.

^{*} Collaborative project with ITN, Chemistry Department.

¹ ITN graduate student.

² ITN MSc student.

Study of Aerosol Deposition Processes using Natural Radionuclides as Tracers*

M.J. Reis, R.N.Rosa¹, A.O. Bettencourt, A.L. Brogueira

Objectives

The main purpose of this project is to study aerosol deposition processes (dry and wet) using natural radionuclides as tracers and its dynamics through the determination of size distributions, deposition velocities, scavenging coefficients and mean residence times that could be used in simulation models.

Results

Natural radionuclides such as ⁷Be, ²¹⁰Pb, ²¹⁴Pb and ²¹²Pb participate in the formation and growth of the accumulation mode aerosol. While associated to the atmospheric aerosols, the radionuclides behaviour will become that of the carriers aerosols, allowing their use as natural tracers for aerosol transport and deposition processes.

Activity size distributions of short-lived radon decay products (reported as ²¹⁴Pb and ²¹²Pb) and cosmogenic ⁷Be in atmospheric aerosol particles were measured using a five-stage high volume cascade impactor.

The activity distributions of 214 Pb, 212 Pb and 7 Be, measured by γ spectrometry (HPGe detectors), were found to be associated with submicron aerosols in the accumulation mode. The activity median aerodynamic diameter (AMAD) ranged from 0.02 to 0.30 μ m (mean 0.19 μ m) for 214 Pb, from 0.07 to 0.32 μ m (mean 0.19 μ m) for 212 Pb and from 0.37 to 0.55 μ m (mean 0.46 μ m) for 7 Be. The total suspended particles (TSP) concentration during the sampling periods ranged from 24 to 95 μ g.m⁻³ and the mass median aerodynamic diameter (MMAD) from 0.35 to 2.38 μ m (mean 1.15 μ m).

A clear difference was observed between ⁷Be and radon short-lived decay products AMAD's, which is probably due to the longer half-life of ⁷Be (53 days) compared to the half-lives of ²¹⁴Pb and ²¹²Pb (26.8 min and 10.6 h, respectively). The longer half-life of ⁷Be, relative to particle attachment and coagulation time, might favour the presence of larger aerosol associations of this radionuclide.

The 214 Pb shift to larger aerosol sizes, in relation to the 212 Pb size distribution, reported by some authors and hypothetically due to the α -recoil of 218 Po, were not observed in our data

Mean residence times, calculated using the ⁷Be activity median aerodynamic diameters (as a result of the activity size distributions) ranged from 3 to 5.6 days. This results seems to indicate that no stratospheric aerosol contribution occurs, at least for the time period for which the size distributions were obtained (summer values), once residence times of about 7 days are considered to be valid, by several authors, for tropospheric aerosols at all levels of the troposphere.

References

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Further work

Determinations of dry deposition velocities and scavenging ratios for different wet deposition mechanisms will be carried out in order to evaluate the relative importance and efficiency of those processes.

Further determinations of the activity aerodynamic size distributions for different periods of the year will be needed in order to assess the occurrence of stratospheric-tropospheric aerosol transfers and its implications in terms of atmosphere dynamic.

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^{*} This is the subject of a PhD Thesis currently in preparation, to be presented to Évora University.

¹ Physics Department of Évora University

Field study for site selection of a permanent station to monitor radioactivity in the atmosphere*

F.P. Carvalho, E. Amaral, J.M. Oliveira, A.Brogueira, M.C. Reis

Objective

The Comprehensive Nuclear Test-Ban Treaty (CTBT) signed up by Portugal foresees the installation of a radionuclide monitoring station (RN 53) as part of an International Monitoring System for detection of nuclear weapon tests.

The ITN/DPRSN was contracted by the CTBTO Secretariat to carry out a field study in S. Miguel in order to select the best site for the station.

Results

A pre-site survey study allowed for screening and compiling the information available about the geology and climate of Azores.

A field survey took place in S. Miguel island during the period 6 to 12 December 1999. Several potential candidate sites were investigated for the actual levels of radioactivity in surface air, and the external dose rate of gamma radiation was measured with portable equipment. Samples of surface soil layers were collected at the same places and brought back to the Laboratory for further analyses.

References

1. Fernando P. Carvalho, Preliminary survey of the site foreseen for installation of Station RN53, Azores, Portugal. Contract Report, Contract N°. CTBTO/99/30/6034. ITN, 10 November 1999.

Further work

Another field trip will take place in early 2000 to complete the survey. The outcome of the project will be the installation of the radionuclide monitoring system.

^{*} Contract Nr. CTBTO/99/30/6034.

Radiological Surveillance of the Rivers Águeda and Mondego in Relationship with Uranium Mining Activities

J.M. Oliveira, M.M. Sequeira, A. Pereira, M.A. Tavares

Objectives

The effluents of uranium mines and facilities of uranium milling are directly discharged into the Rivers Mondego and Águeda. The investigation of enhanced radioactivity in these rivers was started in 1991. The eventual radioactive contamination of the rivers may occur through accumulation of uranium and thorium, which are the most significant radionuclides in the uranium extraction.

In 1999 samples of water, suspended matter, sediments and fish were collected every trimester, in several sampling points in both rivers. The concentrations of ²²⁶Ra and uranium were determined by alpha spectrometry in all samples, but in sediments ²²⁶Ra was determined by gamma spectrometry.

In the Águeda river, the analysis of uranium in water was always 4-5 times higher than in the water of the river Coa, which was selected as a reference environment.

So far, in the river Mondego there has not been recorded any increment of the natural ²²⁶Ra and uranium levels which could be attributed to the uranium mining.

References

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Further work

The radiological surveillance of these rivers will be continued.

Artificial Radioactivity in the Tejo and Zêzere Rivers

M.C.V. Carreiro, M.M.A. Sequeira, A.L.M. Brogueira, M.A. Pereira

Objective

To perform a survey of the artificial and also natural radioactivity in Tejo River, as some nuclear installations exist upstream in Spain, and in Zêzere River, which doesn't suffer such influence and serves as a background to Tejo River.

Results

Monthly sampling of water, sediments, hydrophytes and fish are carried out at three stations in Tejo River (Vila Velha de Rodão, Fratel dam and Valada do Ribatejo). Monthly sampling of water is also made at Castelo do Bode in Zêzere River. At Fratel rain water is also sampled.

¹³⁷Cs is usually detected in all compartments of the river ecosystems; ⁹⁰Sr and ³H are only measured in river and rain waters.

Natural radioactivity (²²⁶Ra, ²²⁸Ra, ²³⁵U) is measured in sediments, hydrophytes and fish.

Radiochemical analyses of river water are carried out in 40 liter of 0.45 μ m filtered samples. Methods for 137 Cs, 90 Sr and 3 H radiochemical analyses have been described (Carreiro & Sequeira, 1987, 1993, and Carreiro et al., 1991). River bank sediments, after drying, are subjected to quantitative gamma-spectrometry on the whole sediment and on the fraction $\leq 212 \mu$ m and $\leq 75 \mu$ m. Fish and hydrophytes from the most common species at the sampling stations, are dry ashed and gamma analysed.

Beta radioactivity measurements are performed in a low-background gas flow counter. Tritium is measured by liquid scintillation. Gamma-spectrometry is carried out in a Ge detector linked to a 8000 channel analyser.

Further work

This radiological survey (iniciated in1976 in Tejo River and in1990 in Zêzere River) needs to be carried out every year.

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Radiological study of Tagus River through Extremadura (Spain) and Alentejo (Portugal)*

A. Baeza¹, E. Garcia¹, C. Miró¹, A.M. Brogueira, C.V. Carreiro, M.M.A. Sequeira, M.M.R. Teixeira

Objective

To carry on a collaboration between the Departamento de Física da Faculdade de Veterinária (Universidade de Extremadura) and the ITN / DPRSN, that focuses on research of radionuclide transport mechanisms, concerning Arrocampo (cooling pond of Almaraz NPP), and the dams of Arrocampo, Torrejon, Alcantara and Cedillo in the Spanish part of Tejo river and Vila Velha de Rodão and Fratel dam in the Portuguese part.

Intercomparison of methods and results.

Results

Two intercomparisons were accomplished and two meetings (in Cáceres and Sacavém) were held for discussions.

Two joint papers were presented to the XXVII Reunión Bienal de la Real Sociedad Española de Física, one is in press in *Water Research*, and another one is in preparation.

Further Work

The work is foreseen until 2001.

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^{*} This work is beeing carried out in the frame of the Regional Programme of Research and Technological Development of Extremadura Commission (Spain).

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Radioactivity levels of alpha-emitters in Portuguese and Iberian rivers

J.M. Oliveira, F.P. Carvalho

Objectives

Rivers may have enhanced concentrations of radionuclides from the uranium series due to uranium mining phosphate on processing and other human activities.

For many years the DPRSN has investigated several watersheds which justified in depth studies, such as the Zêzere, Tagus estuary, the Mondego river and the Águeda river. The results intend to allow for an assessment of the radiation exposure of the population and for any remedial measures if needed.

Results

An outstanding 4 year survey of the river Águeda, which receives run off and waste from U- mines in Spain, was completed. The radioactivity levels of uranium isotopes, radium and thorium in the Águeda were not significantly different of those measures in the river Côa and in the river Douro. The differences observed could be explained by the geological composition of the river beds.

Results on the river Mondego are preliminary. Due to the existence of old uranium mining in the region, a contamination of this watershed is foreseen, although at low levels.

The water of the hydrographic bassin of Zêzere and Tagus rivers shows baseline levels of the U-series radionuclides, with the exception of some areas, especially around the Barreiro peninsula. Here, the concentrations in sediments are elevated due to past phosphoric-acid production activities and releases of phosphate wastes into the estuary.

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Further work

A continuation of the radionuclide survey in Iberian rivers is planned. It will focus especially in the radioactivity in rivers used to supply population with drinking water.

Contamination of coastal lagoons in tropical regions*

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

Objectives

This project aims at assessing the contamination of tropical coastal lagoons by agrochemical residues from the surrounding fields. The water mixing and the sediment accumulation rate in the lagoons will be determinated through the measurement of radionuclides from the uranium natural series.

Results

The project was aproved last year. Two study areas are foreseen: one, Laguna de Términos in Campeche, Mexico, and the other Bahia de Todos os Santos, Salvador, Brasil. In both areas preliminary work was started by local partners in order to select the sampling sites and to install the equipment needed for the project.

A first field trip had been planned to Laguna de Términos last October, but it was cancelled due to the huge rain storms which flooded the area. A new date has been set up and it is expected that most of the work will be carried out during this year.

Preliminary work on the cycling of persistent pesticides and ²¹⁰Po was carried out in comparable environments.

References

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Further work

Field missions will take place to both sites in order to allow for collection of samples. Samples of soils, sediments, biota and water will be analysed for pesticide residues as well as for naturally-occurring radionuclides such as ²²⁶Ra , ²¹⁰Pb and ²¹⁰Po. Especially attention will be paid to the transfer of pesticides and radionuclides (²¹⁰Po) in this environment.

^{*} EU Contract IC-18-CT98-0264, 6×10^6 PTE.

Study of the actinide polyoxometalate systems, mainly the study of the interaction of the lacunary form P₂W₁₇ with the actinide di-oxo cation uranyl UO₂²⁺

M.I. Paiva¹

Objectives

The main objective of this Postdoctoral program is the developmento knowledge concerning the speciation of actinides in the environment. and refers to the period July-December, 1999.

Research program

Studies of actinide behaviour in neutral and basic solutions have been trying to provide answers on the fate of these elements in potential nuclear waste disposal sites and in the environment. The actinide concentration levels in the environment and the mobility in aquifers would be related to the complexes formed with inorganic anions and organic complexing agents in the water, and to some extent limited by the low solubility of certain compounds. Therefore, it is important to characterise the chemical behaviour of the actinides (complex formation and speciation, solubility, etc.) in neutral and slightly basic solutions and the influence of complexing agents that may be present in concentration levels representative of environmental waters. Also the discovery of colloidal forms of actinides, mainly plutonium, has been a major concern related to the underground storage and/or disposal of radionuclides. The majority of the colloids that exist in nature are heterogeneous and difficult to reproduce in lab conditions. Therefore, one of the main goals of the research related to this field is to find analogue compounds that can behave as reproducible pseudo-colloids and clay models and study their ability to complex with metal cations in solution. Among the possible compounds are the large aggregate metal oxides called polyoxometalates or POM. POM's can exhibit two basic structures: plenary P2W18 and lacunary P₂W₁₇. Concerning the specific case of the uranyl ion, Saito and Choppin [J. of Alloys and Compounds 271-273 (1998) 751-755], have studied the interaction of the uranyl ion, UO_2^{2+} , with the plenary form P_2W_{18} and determine the stability constants for the metal-polyoxometalate systems using solvent extraction techniques. However, no data has been reported so far concerning the lacunary P₂W₁₇, mainly, whether the defect site on the lacunary form, which binds the trivalent cations stronger than does the plenary P₂W₁₈ interaction, have a similar stronger binding for the linear dioxo cations.

Experimental Work

- The plenary potassium octadecatungstodiphosphate, $K_6[\alpha-P_2W_{18}O_{62}].XH_2O$ and the lacunary potassium heptadecatungstodiphosphate $K_{10}[\alpha-P_2W_{17}O_{61}].XH_2O$ have been synthethised and characterized by ^{31}P and ^{183}W NMR techniques. Characterization by elemental analysis is under progress (waters of crystallization by thermogravimetry and potassium and tungsten by ICP-MS analysis).
- Preliminary experiments to study the solution behavior of the complex uranyl ion and lacunary P₂W₁₇ by UV-VIS (Cary-14, absorption spectra at 370 nm) have been carried out. It is planned to use a competitive reaction between the metal and a chromophoric indicator for this ligand.
- The study of the application of the solvent extraction to determine the stability constant of the complex uranyl ion and lacunary P₂W₁₇ has started. Preliminary results indicate the following extraction conditions: extractant=HDEHP in toluene (4x10⁻⁵-6x10⁻⁵ M); prH=4.62 (0.01 acetate as buffer); ionic strength=1 molar NaClO₄; uranium concentration=²³³U carrier free solution, total activity=20000 cpm/liquid scintillation vial.

Experimental Work not Related to the Main Project

- Lifetime measurements of Eu(III) in CHCl₃ and CDCl₃ in order to identify the hydratation number of Eu(b-diketone)3 phenanthroline and the decay constants for Eu(III) in the phenanthroline adduct of the PTA chelate in CHCl₃ and CDCl₃. Solvent extraction samples were analysed by Laser Induced Fluorescence using a Spectra-Physics DCR-2 10Hz pulsed Nd-YAG Pumped Dye Laser. Data was treated by Sigmaplot using the Marquardt-Levenberg algorithm (work done in collaboration with Prof.Yuko Hasegawa of the Science University of Tokyo).
- Separation and purification of ²⁴²Pu from a batch of liquid wastes by co-precipitation with bismuth phosphate (work done in collaboration with Dr. Dean Peterman, FSU).

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¹ Post doctoral position at the Department of Chemistry, Florida State University.

Study of Lead Concentrations Effects on Freshwater Fish

M.C. Carreiro, M.L. Pedro, M.A. Gameiro, M.A. Ramos¹

Objective

Lead is an environmental toxic spread with a large industrial application. Many industries reject liquid effluents without treatment into the rivers, resulting in a significative aquatic environmental pollution.

It is well known that the heme biosynthetic system is a target of the toxic action of lead. Inhibition of an erythrocytic enzym (the delta-aminolevulinic acid dehydratase - ALAD) lead induced is well documented.

The purpose of this project is to study the induced effects by several lead concentrations on freshwater fish, namely, *Cyprinus carpio*. Lead interference on the activity of the fishes ALAD is evaluated. A spectrophotometric method is performed, as Directive du Conseil 82/605/CEE selected and recommended as standard.

Results

The determination of ALAD activity and the hematocrit in a healthy population of 43 freshwater fishes with no lead exposure was performed. The studied fishes weight range from 20 - 1300 g and his length range from 20 - 40 cm. The fishes lives in the aquariums with 3000 liters the capacity, in the IPIMAR instalations.

The mean value found for the fish population ALAD activity is around 0,099 U/ml, which corresponds to 0,099 delta-aminolevulinic acid (ALA) μ mol/min/ml RBC.

The mean value found for the fish population hematocrit is around 28%.

¹ Instituto Português de Investigação das Pescas e do Mar (IPIMAR) - Lisboa.

Metrology and radiation dosimetry

Procedures for the routine individual dose assessment of external radiation within EU countries and Switzerland

P.Ambrosi¹, D.T.Bartlett², A.F.Carvalho, A.Delgado³, E.Fanttuzzi⁴ and L.Lindborg⁵

Objective

The European Union legal requirements for individual monitoring are given in "Council Directive 96/29 EURATOM of 13 May 1996 laying down basic safety standards for the protection of public and workers. The dose quantities to be estimated for the purposes of dose control and dose limitation are effective and equivalent doses and the Directive states that 'operational quantities for external radiation are used for individual monitoring for radiation protection purposes'.

The principal objectives of the project were the consolidation within the EU of the quality of individual monitoring using personal dosemeters and to facilitate harmonised procedures. For these purpose it was revised dosimetric performance requirements. Others two EURADOS groups of experts prepared a catalogue of body dosemeters suitable for estimating $H_p(10$ and $H_p(0.07)$ from all forms of external radiation and carry out performance tests for whole body and extremity personal dosemeters broadly representative of those in use in the EU.

Results

This report has given details of approval procedures, dosimetric requirements, and performance tests for dosemeters and dosimetric services of all Member States and Switzerland and of the international recommendations from which, in general they are derived. There are widely differing national technical and legal requirements and the information contained herein may lead to some convergent evolution of procedures and a greater degree of harmonisation.

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Study of LiF:Mg,Cu,P and LiF:Mg,Ti Detectors on the Low Gamma Dose Range: Thermal Stability and Limits of Detection*

J.G. Alves, A. Delgado¹, M. Secca², E.M. Amaral, J.L. Muñiz¹, J.M. Gómez Ros¹

Objectives

This work aims at the study of two of the most important characteristics of LiF:Mg,Ti (TLD-100) and LiF:Mg,Cu,P (GR-200) when used in the fields of Environmental and Personal Dosimetries, the low gamma dose range.

It aims at the characterisation of the thermal stability of GR-200 compared to the classical TLD-100, taken as reference. And it also aims at the experimental evaluation of the limits of detection and of determination of the dosimetry system based on these two materials. In the course of this work, computerised glow curve analysis methods (GCA) and conventional methods were used.

Results

LiF:Mg,Ti (TLD-100) and LiF:Mg,Cu,P (GR-200) are two well known thermoluminescent materials used in the Dosimetry of ionising radiation. TLD-100 as it is one of the most widely used materials and GR-200 for its attractive properties: hypersensitivity, glow curve simplicity and the same matrix as TLD-100.

Thermal stability was studied by storing samples of both materials, both irradiated and unirradiated, in laboratory controlled conditions at 40°C and 70°C, for storage periods that varied from 6 hours to 24 days [1,2]. These two temperatures were considered as representative of the temperatures endured during environmental exposures. All curves were analysed using computerised methods allowing the deconvolution of the glow curve on its individual peaks, so that the evolution of each peak could be followed. In this way the temperature induced effects on filled and empty traps (irradiated and unirradiated detectors) could be followed. The results show that GR-200 is far more stable than TLD-100 for the two temperatures studied, and that the migration and aggregation of the impurities based defects structure altering the trap system, are the main cause of the sensitivity changes observed. Thermal fading, understood as the spontaneous release of the trapped charges, does not influence the behaviour of the main peaks, in both materials [1,2].

The limits of detection and of determination were studied starting from Currie and Hirning's well known expressions, originally deduced for the conventional analysis of the glow curves. All curves were analysed with the conventional and the simplified analysis method (SGCA). This method, without attempting to resolve the individual peaks, identifies specific points of the curve, determines the background signal and provides a net result based on the analysis of the glow curve shape. The simplification of the procedures inherent to the use of SGCA was introduced on the expressions for the limits and new expressions were proposed. An experiment was set up to test the proposed expressions and to compare the results of the two methods [3]. The results show that SGCA always provides better results than the conventional method, and allow the estimate of a detection threshold for the detectors and for the measurement system used. $10 \,\mu$ Gy and $1 \,\mu$ Gy were estimated as the detection threshold, for TLD-100 and GR-200, respectively, analysed with SGCA [3].

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Further work

This work is presently completed. A PhD Thesis has been prepared and presented at the "Fac. de Ciências e Tecnologia (Univ. Nova de Lisboa)" for discussion.

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PhD Thesis presented at the New University of Lisbon, Co-funded by PRAXIS XXI, BD/13522/97.

CIEMAT – Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas, Madrid, Spain.

² Dep. Física, Fac. de Ciências e Tecnologia da Univ. Nova de Lisboa (PhD supervisor along with A. Delgado).

Computer methods in radiation protection. Computational dosimetry for photon energies in the range 10-150 keV.*

A. D. Oliveira, J. J. Pedroso de Lima¹

Objectives

The aim of this work is the computer implementation of well-known computer routines for Monte Carlo simulation of the interaction of photons in matter, resulting in several home made codes. Nevertheless the first goal of the work was the application to the description of the photon scattering in the energy range 10-150 keV. This energy range is used mainly in medicine in radiodiagnostic. A short description of the code was presented in a previous report (annual report 1998).

Results

Since last year the progress of the work are mainly in the next topics:

- a) The entropy concept was applied successfully to the description of the energy degradation in the scatter of a photon beam. One important result is the connection found between the entropy of the photon energy degradation and the behaviour of the effective quality factor. The range 10-150 keV can be divided in three zones in accord with the entropy value.
- b) We have developed and applied a new method, designed dynamic analysis of data, for the definition of the radiation beam boundary, which allows fresh insights in the spatial structure of the radiation scatter.

These two topics are new approaches to radiation physics, allowed only by the data obtained with the computer simulation. As the time goes by they have become more important in all work.

Other topics developed are:

- a) Determination and study of backscattered, transmitted and imparted energy from photon beams incident in water slabs. This study are made for three water slabs with dimensions of 1, 5 and 30 cm and for the photon energy 20, 40, 60, 80, 100 and 150 keV. These results are applied to the optimisation of the radiodiagnostic practice from a physics point of view.
- b) In the study of the computer simulation results, we found several types of convergence. With a careful analysis of the convergence type we can decrease the uncertainty associated with the result of the simulation. For example, in the build-up determination we can go from B=1.888±0.001 to B=1.8659±0.0001 with the same data and a judicious procedure.
- c) We found in the literature several methods for the computer determination of the backscatter factor. We present also a new procedure for this determination and compare with others.

References

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Further work

All the topics presented are under development but there is much yet to do in the traditional concept: the dose calculation.

We intent to finish the thesis before the end of the year 2000.

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^{*} This is the subject of a PhD these currently in preparation.

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Biological Dosimetry

J. H. Pereira Luis

Objectives

Study the dose-effect relationship for induction of chromosomal aberrations with different types (alpha, beta, gamma, X and neutrons) and qualities of radiation, as a tool for dose and risk assessment of human exposure to ionising radiation.

Research the low-level effect of radiation on chromosomal peripheral blood lymphocytes of individuals professionally exposed to ionising radiation.

Maintenance of operational and standardised biological dosimetry methods, to be used in cases of radiological accident situations or in overexposures to ionising radiation, in order to estimate the dose and the risk to humans.

Results

During last year our work were distributed by the followings tasks: Chromosomal aberration analysis in peripheral lymphocytes of radiation workers, Biological Dosimetry on Alqueva accident and Retrospective biodosimetry on a case of accident 22 years ago.

The radiation-induced damage on cellular chromosomal DNA was measured by cytogenetic methods on the Biological Dosimetry Laboratory. The cytogenetic methods used were chromosomal aberrations sister-chromatid exchanges and micronucleus. All the studies were done *in vitro* using chromosomal peripheral blood lymphocytes.

Uranium miners

It has been done an analysis of the chromosome aberrations of peripheral blood lymphocytes in a group of 15 miners and in a control group. It was observed an increased frequency of chromosomal damage on peripheral blood lymphocytes of miners, relatively to the control group, which can mean an increased health risk for these workers. The analysis of chromosomal aberrations distribution on the cells, of one thousand of metafases, on cases that the miners' lymphocytes exhibit dicentric chromosomal aberration, shows an overdispersion of damage on cells, that agrees with an exposure to high Let radiation from radon. For those miners, there are a linear relationship between total chromosomal damage and working years on mines.

<u> Alqueva Accident</u>

In the sequence of an Alqueva accident on 6-4-99, with an industrial radiation Iridium 192 source, 5 radiation workers, which were not using any physical dosimeter, were exposed to gamma radiation. On this case, the only method to know the real absorbed dose was by Biological Dosimetry. After the analysis of chromosomal aberration on one thousand peripheral blood lymphocytes of each worker, and using our calibration curve for gamma radiation, we conclude that total dose received by the worker more exposed, was less than 0,1 Gy.

Retrospective biodosimetry

Radiation dosimetry of human radiation exposure is currently based on scoring of dicentric chromosomes in human blood lymphocyte metaphase preparations. However dicentrics are lost with successive cell divisions, and so this technique can lead to an underestimation of the dose for past exposures. For retrospective dosimetry stable chromosomal aberrations, like translocations, must be used. The technique of fluorescence *in situ* hybridization (FISH), using the labelling of all centromeres and whole chromosome painting is the suitable one for chromosomal translocation analysis. In this study, human blood samples, of 2 radiation workers accidentally exposed, 22 years ago, to a gamma radiation, whole body doses, 15 - 25 rem, were collected and lymphocyte metaphase preparations are made. We did also, lymphocyte metaphase preparations after irradiation *in vitro*, of control human blood samples, with known cobalt gamma doses. The aberrations were been detected by means of FISH with a cocktail of DNA probes specific for whole chromosomes 1, 4 and 12. The analysis of chromosomal aberrations is in progress.

References

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Further work

Study the dose-effect relationship for induction of chromosomal aberrations with X-rays and neutrons and conclude the work of Retrospective biodosimetry by using fluorescence *in situ* hybridization analysis.

Study of genetic and biochemical effects of ¹³¹I treatment in thyroid cancer patients^{*}

Octávia Monteiro Gil, Nuno Oliveira^{1,2}, António Rodrigues¹, António Laires¹, Teresa Ferreira³, Edward Limbert³, José Rueff¹

Objectives

This study aims to evaluate the DNA damage produced by radioactive iodine (¹³¹I) treatment in patients with thyroid cancer, using cytogenetic and biochemical parameters. The cytogenetic parameters evaluated were the frequencies of chromosomal aberrations and micronuclei in peripheral blood lymphocytes, whereas the biochemical parameters were indicators of oxidative stress, such as thiobarbituric reactive products (prTBA), plasma uric acid and total antioxidant status.

Results

We studied 19 thyroid cancer patients, from the Nuclear Medicine Department of the Portuguese Oncology Institute of Lisbon, immediately before treatment, and 1 and 6 months after treatment with therapeutic ¹³¹I, after surgical ablation of the thyroid gland. All patients - who were non-smokers except one- received a dose of 70 mCi. All but 3 were female. Considering the pooled data of the nineteen ¹³¹I treated patients the frequencies of micronuclei in cytochalasin-blocked human lymphocytes (MNCB (‰)) increased from 5.2 to 9.7 at one month after treatment (p<0.01) and to 8.4 at six months after treatment (p<0.05). The chromosomal aberrations (CA (%)) increased from 1.7 to 3.5 at one month after treatment (p<0.01) and to 4.0 at six months after treatment (p<0.01). Furthermore, when studying the prTBA present in whole blood expressed as MDA, the uric acid and the total antioxidant status present in plasma, we found some slight differences in these parameters among the three periods studied. These changes were generally not significant except for the decrease of prTBA from one month to six months and the decrease in plasmatic uric acid concentration observed one month after therapy. In conclusion this study pointed out to a mild but significant and persistent increase of DNA damage as assessed by both cytogenetic assays (1,2).

Considering the conflicting results in the literature about the presence of chromosomal instability in this kind of patients before any therapy, we have also studied its possible existence through the study of spontaneous CA's and MNCB in peripheral blood lymphocytes. We studied these indicators in a group of 22 thyroid cancer patients and compared them with a population of 24 persons who do not have thyroid cancer. Our results demonstrated that there was no significant alteration in the spontaneous cytogenetic damage pattern of these cancer patients. (This report will soon be submitted to a scientific journal).

Additionally, we have tried to perform a similar study on patients with hyperthyroidism. In spite of many efforts we couldn't continue the study because the great majority of the patients refused to participate (in a five-month period only one patient agreed to participate in the study).

Presently we are studying the same group of thyroid cancer patients two years after iodine exposure, excluding those who received additional treatments, to look at the long-term effects of radiation in terms of chromosomal damage. For this purpose CA's and MNCB assays are being used as indicators of DNA damage.

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Further work

Furthermore we have initiate the study of the induction of an adaptive response in peripheral blood lymphocytes of thyroid cancer patients after treatment with ¹³¹I, by assessing the induction of MNCB after a challenge *in vitro* with a radiomimetic genotoxic agent. Preliminary results show that there is indeed a reduction of MNCB in lymphocytes induced by the genotoxic agent, after treatment with ¹³¹I *in vivo*, but this reduction is transient, not observable 6 months after the treatment.

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^{*} This is the subject of a PhD thesis currently in preparation.

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² Faculty of Pharmacy, University of Lisbon.

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