

EUROPEAN COMMISSION DIRECTORATE-GENERAL FOR ENERGY

DIRECTORATE D - Nuclear Safety and Fuel Cycle **D.3 Radiation Protection** 

# **TECHNICAL REPORT**

# VERIFICATIONS UNDER THE TERMS OF ARTICLE 35 OF THE EURATOM TREATY

Southern and western Spain – former uranium installations and national monitoring

**SPAIN** 

24-28 September 2012



Reference: ES 12/06

# VERIFICATIONS UNDER THE TERMS OF ARTICLE 35 OF THE EURATOM TREATY

- <u>SITES and FACILITIES</u>: Former uranium mining sites (La Virgen, Valemascaño, 'Quercus'); former uranium production sites at Andújar and Saelices el Chico ('Quercus' and to a certain extent 'Elefante'); national monitoring system for environmental radioactivity in Andújar (Andalusia) and Saelices el Chico (Castile and León); laboratories at Seville University, Granada University, Salamanca University, Saelices el Chico (ENUSA) and Juzbado (ENUSA).
- <u>DATE</u>: 24 to 28 September 2012

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# **TECHNICAL REPORT**

# ABBREVIATIONS AND ACRONYMS

AECID	<i>Agencia Española de Cooperación Internacional para el Desarollo</i> (Agency of International Cooperation for Development)
AEMET	Agencia Estatal de METeorología (National Meteorology Agency, formerly INM)
AVRA	<i>Área de Vigilancia Radiológica Ambiental</i> (Environmental Radiological Surveillance Area; unit at <i>CSN</i> )
a.s.l.	Above sea level
BAC	Balsa Aguas Corta
BOE	Boletín Oficial del Estado (Official Gazette)
BRV	Balsa de Regulación de Vertido
CEDEX	Centro de Estudios y Experimentación de Obras Públicas, Madrid
CIEMAT	<i>Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas</i> (public research institution attached to the Ministry of Science and Education, formerly JEN), Madrid
CITI	Área de Ciencias de la Tierra (Earth Sciences Area; unit at CSN)
CITIUS	Centro de Investigación, Tecnología e Innovación de la Universidad de Sevilla (Research, Technology and Innovation Centre of the University of Seville)
CSN	Consejo de Seguridad Nuclear (Nuclear Safety Council)
DG ENER	Directorate-General for Energy (European Commission)
EC	European Commission; European Community
ENAC	Entidad Nacional de ACreditación (National Accreditation Organisation)
ENRESA	<i>Empresa Nacional de Residuos Radiactivos Sociedad Anónima</i> (Spanish radioactive waste management agency)
ENUSA	ENUSA INDUSTRIAS AVANZADAS, S.A. (former name Empresa Nacional del Uranio Sociedad Anónima, National Uranium Company)
EU	European Union
FEDER	<i>Fondo Europeo de Desarrollo Regional</i> (European Funds for Regional Development)
FUA	Fábrica de Uranio de Andújar (Andujar Uranium Mill)
FWHM	Full Width Half Maximum (resolution measure in spectrometry)
GM	Geiger-Müller (radiation detector)
GEOCISA	GEOtecnia y CImientos S.A., Madrid
HPGe	High Purity Germanium (radiation detector)
IAEA	International Atomic Energy Agency
ICP-MS	Inductively Coupled Plasma – Mass Spectrometry
ICRP	International Commission on Radiological Protection
INM	Instituto Nacional de Meteorología (National Meteorology Institute, now AEMET)
IPPC	Integrated Pollution Prevention and Control
ISO	International Standardization Organization
JEN	Junta de Energía Nuclear (Nuclear Energy Board, now CIEMAT)
LEGe	Low Energy Germanium (radiation detector)

LSC	Liquid Scintillation Counting (radiation measurement)
MITYC	<i>Ministerio de Industria, Turismo y Comercio</i> (Ministry of Industry, Tourism and Trade)
NaI(Tl)	Sodium iodide, thallium activated (radiation detector)
NIM	Nuclear Instrumentation Module
NIST	(US) National Institute of Standards and Technology
NORM	Naturally Occurring Radioactive Material
NPL-UKAS	National Physics Laboratory-UK Accreditation Service
NPP	Nuclear Power Plant
OJ	Official Journal (of the European Institutions)
PIPS	Passivated Implanted Planar Silicon (radiation detector)
PTB	Physikalisch-Technische Prüfanstalt, Braunschweig
PVC	PolyVinyl Chloride
PVM	Plan de Vigilancia y Mantenimiento (Surveillance and Maintenance Plan)
PVRA	<i>Programa de Vigilancia Radiológica Ambiental</i> (Environmental Radiological Monitoring Programme)
PVRAIN	<i>CSN's</i> independent environmental monitoring programme established as a control of the <i>PVRA</i> implemented by licensees
QA/QC/QM	Quality Assurance / Quality Control / Quality Management
RAR	Red de Alerta de la Radiactividad (radioactivity warning network)
REA	<i>Red de Estaciones Automáticas de vigilancia radiológica ambiental del CSN</i> (automatic station network)
REGe	Reverse Electrode coaxial Germanium (radiation detector)
REM	Red de Estaciones de Muestreo (sampling station network)
REVIRA	<i>REd de VIgilancia Radiologica Ambiental</i> (environmental radiological monitoring network – not associated with installations)
RPSRI	<i>Reglamento de Protección Sanitaria contra Radiaciones Ionizantes</i> (regulation for health protection against ionising radiations)
SALEM	Sala de emergencias del CSN (CSN's emergency centre)
SEAC	Sociedad Española de Aplicaciones Ciberneticas, S.A. (Spanish Society of Cybernetic Applications)
SEPI	Sociedad Estatal de Participaciones Industriales
TAC	Tratamiento Aguas Corta
TAD	Tratamiento Aguas Dique
UPS	Uninterruptible Power Supply
UV/VIS	UltraViolet-VIsible Spectroscopy (analytical device)
TEDA	Tri Ethylene Di-Amine
WHO	World Health Organization

# 1 INTRODUCTION

Article 35 of the Euratom Treaty requires that each Member State shall establish facilities necessary to carry out continuous monitoring of the levels of radioactivity in air, water and soil and to ensure compliance with the basic safety standards <sup>(1)</sup>.

Article 35 also gives the European Commission (EC) the right of access to such facilities in order that it may verify their operation and efficiency.

For the EC, the Directorate-General for Energy (DG ENER) and in particular its Radiation Protection Unit is responsible for undertaking these verifications.

From 24 to 28 September 2012, a team of four inspectors from DG ENER D.3 visited former uranium mining and milling sites near Andújar and Saelices el Chico (in particular 'Quercus' and to a certain extent 'Elefante') after having been informed at an opening meeting of the current situation by the site operator, by the regulatory authorities and by other actors. The goal of this verification was to verify a number of monitoring installations involved in the environmental radioactivity monitoring of these sites. The laboratories performing the measurements were also included in this verification. Part of the Spanish network for monitoring environmental radioactivity in the areas visited was covered as well.

This visit included also meetings with the Spanish competent authorities, *Consejo de Seguridad Nuclear*  $(CSN)^{2^{1}}$ , the Ministry of Industry, Madrid, the Regional Governments (Andalusia, Castile and León), the operator *Enusa Industrias Avanzadas S.A.* (former name *ENUSA*, *Empresa Nacional del Uranio Sociedad Anónima*, National Uranium Company), the *Empresa Nacional de Residuos Radiactivos (ENRESA*; Spanish radioactive waste management agency) and *SEAC (Sociedad Española de Aplicaciones Ciberneticas, S.A.*, the firm in charge of the automatic radiological monitoring devices), as well as with representatives of the analytical laboratories performing the measurements. The verifications were carried out in accordance with the programme in Appendix 1.

The purpose of the review was to acquire full information both from the operator and from the regulator concerning the environmental radiological monitoring at and around former uranium mining and milling sites in Spain and to get state of the art information on the remedial measures put in place till now.

The present report contains the results of the discussions with the site operators, other actors and the Spanish competent authority *CSN*, as well as results of the verification team's review of some aspects of the environmental radioactivity surveillance at and around these former uranium mining and milling sites.

With regard to general radiological and environmental radioactivity and discharge monitoring aspects the present report is also based on information collected during the past verifications of the NPPs Trillo, Cofrentes and Ascó as well as during the verification of the Huelva (NORM industry) site in Spain.

# 2 PREPARATION AND CONDUCT OF THE VERIFICATION

## 2.1 INTRODUCTION

The Commission's request to conduct an Article 35 verification was notified to the Spanish authorities on 2 February 2012 (letter reference ENER/D3/CG/es Ares (2012) addressed to the Spanish Permanent Representation to the European Union).

All practical arrangements for the implementation of this mission were made with the persons designated at *the Consejo de Seguridad Nuclear*, in particular with Mrs Lucila Ramos Salvador, Deputy Director for Environmental Radiological Protection.

<sup>&</sup>lt;sup>1</sup> Council Directive 96/29/Euratom of 13 May 1996 laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation (OJ L-159 of 29/06/1996, page 1).

<sup>&</sup>lt;sup>2</sup> Consejo de Seguridad Nuclear / Nuclear Safety Council, Justo Dorado 11, 28040 Madrid.

# 2.2 DOCUMENTATION

In order to facilitate the work of the verification team, a package of information was supplied in advance by *CSN*. Additional documentation was provided during and after the visit. All documentation received is listed in Appendix 2 to this report. The information thus provided has been extensively used for drawing up the descriptive sections of this report.

# 2.3 **Representatives of the competent authorities, the site operators and other organisations involved in environmental radioactivity monitoring**

During the visit the following representatives of the national authorities, the operator and other parties involved were met:

## Nuclear Safety Council (CSN)

Lucila RAMOS SALVADOR	Deputy Director for Environmental Radiological
	Protection
Rosario SALAS COLLANTES	Head of Environmental Radiological Surveillance
	Area (AVRA)
Carmen REY DEL CASTILLO	Scientist, AVRA
Inmaculada MARUGAN TOVAR	Scientist, AVRA
Sofía LUQUE HEREDIA	Scientist, AVRA
Antonio VELA GUZMAN	Scientist, CITI

#### Ministry of Industry, Tourism and Trade; Directorate General for Energy Policy and Mines

Jaime DE PONGA DEL POZO	Head of branch in the Nuclear Energy Deputy
	General Directorate

#### **Regional Governments (Andalusia, Castile and León)**

Francisco JIMENEZ ALMANSA	Head of the Department of Industry, Energy and Mines of the Regional Government of Andalusia
Julio RODRIGUEZ SORIA	Head of the Territorial Department of Industry, Trade and Tourism of the Regional Government of Castile and Leon

#### Enusa Industrias Avanzadas S.A.

Javier Ruiz SANCHEZ-PORRO	Head Manager (Saelices el Chico)
Agustin PEREZ FONSECA	Head of Radiological Protection Department
	(Juzbado)
Maria Luisa BORDONABA PEREZ	Head of Radiological Protection Department
	(Saelices el Chico)
Olegario TEIJON ESCUDERO	Responsible Laboratory Technician (Juzbado)
Francisco Javier CHAMIZO REBOLLEDO	Laboratory Analyst (Saelices el Chico)
Ignacio SANCHEZ RODRIGUEZ	Laboratory Analyst (Saelices el Chico)
Beatriz CURROS GONTAD	Responsible Quality Control (Saelices el Chico)
Javier BIELA JAVIERRE	Responsible for Valdemascaño Project

## ENRESA

Maria Teresa LOPEZ FERNANDEZ	
Fernando BELTRÁN DE HEREDIA	ENRESA's Partner from Westinghouse

## SEAC

Mario RUIZ LLATA	Head of Maintenance Department

## University of Granada

M <sup>a</sup> Angeles FERRO GARCÍA	Professor and Head of Research Laboratory
Francisco PIÑERO GARCÍA	Laboratory technician

#### University of Seville

Rafael GARCÍA-TENORIO GARCÍA-BALMASEDA	Professor; Scientific Advisor at CITIUS, Head of
	Research Group "Applied Nuclear Physics"
José Luis MAS BALBUENA	Scientist, CITIUS
Santiago HURTADO BERMÚDEZ	Scientist, CITIUS

#### University of Salamanca

Begona QUINTANA ARNES	Head of research group and Project manager
Felipe LORENZO	Quality manager
Teresa MARCOS	Laboratory technician
Juan CARLOS LOZANO	Project manager

# **3 COMPETENT AUTHORITIES & LEGAL BACKGROUND**

## 3.1 INTRODUCTION

In Spain, generally, facilities liable to generate radioactive effluents and/or waste must have proper control storage, treatment and removal systems. Radiological monitoring programmes must be based on site and discharge characteristics. For installations of the nuclear fuel cycle site-related environmental radiological monitoring programmes have to be implemented by the operators. Site-specific control programmes are implemented by the *Consejo de Seguridad Nuclear (CSN)*. Nationwide monitoring networks for environmental radioactivity are set up and managed by *CSN*.

For NORM industries decisions with regard to any environmental radioactivity monitoring programme have to be made, case by case, by the relevant competent authority, based on advice by *CSN*.

The nationwide radiological monitoring network established and managed by *CSN* is operational since 1992 (except for rivers, which are surveyed since 1984). It is independent from the networks associated with nuclear facilities. It includes an automatic station network (*REA*) for real-time measurement of ambient gamma dose rate and atmospheric radioactivity and a sampling station network (*REM*) for sampling and analysis programmes for air, soil, rivers, coastal water, drinking water, milk and mixed diet.

## **3.2** COMPETENT SPANISH AUTHORITIES AND OTHER ACTORS

# 3.2.1 *Consejo de Seguridad Nuclear* (Nuclear Safety Council)

The *Consejo de Seguridad Nuclear (CSN)*, established in 1980, is the Spanish organisation responsible for nuclear safety and radiological protection. It is independent from the government and reports to the Spanish Parliament. *CSN* issues reports with binding content prior to the awarding of authorisations to regulated facilities (either "nuclear" and/or "radioactive") by the Ministry of Industry, Tourism and Trade and proposes regulations on nuclear safety and radiation protection nationwide.

*CSN* is an associated body formed by five members (a president/chairman and four commissioners) proposed by the Government and endorsed by the Congress of Deputies. Under the overall responsibility of the Secretary General, *CSN* is organised in two Technical Directorates: Nuclear Safety and Radiation

Protection. The latter includes three Deputy Directorates: Emergencies, Operational Protection and Environmental Radiological Protection.

*CSN* maintains a control and monitoring programme for nuclear installations and facilities related to medical, industrial or research activities that are using radioactive substances. It has to provide mandatory and binding documents for any modifications of such installations and facilities. On average, *CSN* carries out around 200 control inspections per year in nuclear power plants operating in Spain. It is also responsible for proposing regulations to the minister responsible for industry concerning radiological protection of workers and members of the public and safety criteria for waste management.

With respect to the environment, *CSN* has the following regulatory functions:

- To control the radiological impact of nuclear and radioactive installations on the environment, especially concerning radioactive discharges (aerial/liquid) into the environment, their accumulation in the surroundings of such installations and the evaluation of the resulting radiological impact.
- To run its own programmes of environmental radiological surveillance (both around nuclear installations and at national level) and to supervise all environmental radiological protection activities conducted by nuclear installations and by facilities using radioactive substances. In this regard *CSN* has established and manages the national environmental radiological monitoring network, which includes, among others, the sampling of foodstuffs (milk and mixed diet). The design and development of this network follows EC recommendations.

*CSN* also promotes research programmes in matters related to its field of activity. It proposes regulations and informs the public through direct contact with the media, diffusion of publications, an internet web page and an information centre. *CSN*'s annual report to Spain's Congress and Senate provides information on the results of the monitoring programmes; more detailed information about these results is published in a specific annual report. A summary of the results is posted on *CSN*'s internet site to provide information to the public.

The competent authority advised by *CSN*, will require the owners of work activities within which there is an increased exposure to natural radiation sources, to carry out the necessary studies, in order to assess the existence of a significant increase in the exposure of workers or of members of the public which cannot be disregarded from the radiation protection point of view. The studies are to be performed according to recommendations from *CSN*. With the study results, *CSN* identifies those work activities that must be subjected to control and defines those activities that need to be provided with adequate exposure monitoring devices; establishing, if the case arises, the need of corrective actions or radiation protection measures. *CSN* informs the competent authorities about the conclusions and measures needed, to be required of the party responsible for the work place.

# 3.2.2 Ministry of Industry, Tourism and Trade

The Nuclear Energy Act defines radioactive waste as any residual material (i.e. for which no use is foreseen) that contains radioactivity above certain levels. These levels have to be established by the ministry responsible for industry (at the time of the verification the *Ministerio de Industria, Turismo Y Comercio, MITYC*; Ministry of Industry, Tourism and Trade). A report of *CSN* serves as a binding basis. Within *MITYC*, the Directorate General for Energy Policy and Mines is in charge of such issues.

## 3.2.3 Autonomous government of Andalusia "Junta de Andalucía"

With regard to effects of discharges from NORM as well as of uranium mining and milling activities, the regional government is the competent authority at regional level. In the case of the Andújar sites, this is the Department of the Environment of the *Junta de Andalucía*. The competence is related to the implementation of EU Directive 96/61/EC (the 'IPPC Directive').

# **3.2.4** Ministry of Health and Consumer Affairs

The body responsible for the radiological monitoring of foodstuffs is the Ministry of Health and Consumer Affairs. Through the *Centro Nacional de Sanidad Ambiental* (national centre for environmental health) the ministry analyses the radioactivity content of imported food products and issues export certificates.

# 3.2.5 ENUSA Industrias Avanzadas, S.A.

*ENUSA Industrias Avanzadas, S.A.* was constituted in 1972. Its former name was *ENUSA (Empresa Nacional del Uranio Sociedad Anónima* – national uranium company). This company belongs to the "Sociedad Estatal de Participaciones Industriales" (60%), which itself is owned by the Ministerio de Hacienda y Administraciones Públicas (Ministry of Finances and Public Administation), and to the Centro de Investigationes Energéticas Medioambientales y Tecnologicas (CIEMAT, 40%), which itself belongs to the Ministerio de Economía y Competitividad (Ministry of Economy and Competitiveness). This means that ENUSA belongs to and is controlled by two Spanish ministries. ENUSA is located in Madrid (head offices), Juzbado (nuclear fuel factory) and Saelices el Chico (former uranium mining and milling sites). It employs 610 staff in these locations.

Among other activities *ENUSA* is in charge of the environmental radiological monitoring and remediation of the former uranium mining and milling sites in the Ciudad Rodrigo area (Saelices el Chico with the *'Elefante'* uranium production plant, the *'mina Fe'* and 'D' mines and the 'Quercus' mill, as well as other former mines in the surroundings, such as Valdemascaño and Casillas de Flores).

# 3.2.6 ENRESA

*ENRESA* (*Empresa Nacional de Residuos Radiactivos*) is the public company in charge of the safe management, storage and disposal of the radioactive wastes produced in Spain. *ENRESA*'s capital is 100% public. The company belongs to *CIEMAT* (80%) and to *SEPI* (*Sociedad Estatal de Participaciones Industriales*, 20%).

*ENRESA* is also responsible for the dismantling of nuclear power plants when their service lifetime has come to an end and for the environmental restoration of disused uranium mines and facilities.

## 3.3 LEGAL PROVISIONS FOR ENVIRONMENTAL RADIOACTIVITY MONITORING

With regard to legal provisions reference is made to the reports of the Article 35 Euratom verifications to the NPPs at Trillo (2004), Cofrentes (2007) and Ascó (2008), as well as to the ones at Huelva (2009) and Palomares (2010).

# **3.3.1** Legislative acts regulating environmental radioactivity monitoring (general)

- Law 25/1964 of 29 April 1964, on nuclear energy. Published in the *Boletín Oficial del Estado (BOE*, Official Gazette) nº 107, of 4 May 1964.
- Law 15/1980 of 22 April 1980, on the creation of *CSN*. Published in *BOE* n° 100 of 25 April 1980; partially amended by Law 14/1999 and Law 33/2007 of 7 November 2007.
- Law 14/1999 of 4 May 1999, on rates and public charges for services provided by the Nuclear Safety Council. Published in *BOE* n° 107 of 5 May 1999 and amended in *BOE* n° 131 of 2 June 1999.
- Royal Decree 783/2001 of 6 July 2001, adopting the regulations on health protection against ionising radiations. Published in BOE n° 178, of 26 July 2001. This includes national responsibilities in relation to the protection against natural radiation exposure.
- Royal Decree 1836/1999 of 3 December 1999, adopting the regulations on nuclear and radiological installations. Published in *BOE* n° 313, of 31 December 1999. This regulation was modified by the *Royal Decree* 35/2008 of 18 January 2008.

# **3.3.2** Legislative acts regulating environmental radioactivity monitoring (uranium mining and milling)

• Nuclear Energy Act (Law 25/1964) developed, among others, by Royal Decree 2869/1972, Regulation on Nuclear and Radioactive Facilities: Facilities producing uranium, thorium and their compounds are considered first category radioactive facilities which require prior construction and operation permits. Preoperational and operational monitoring programmes are required in construction and operation permits. This Royal Decree was repealed by Royal Decree 1836/1999.

Royal Decree 1836/1999 (Regulation on Nuclear and Radioactive Facilities), modified by Royal Decree 35/2008: Facilities producing uranium, thorium and their compounds are considered nuclear fuel cycle radioactive facilities, which are required to have prior construction, operation and dismantling authorisations and a decommissioning statement.

Construction, operation and dismantling permits must be supported by a preoperational, operational and dismantling monitoring programme.

The operation permits and the execution of restoration plans of uranium mines will require, prior to their concession, a compulsory and binding report of the *CSN* concerning radiation protection matters.

• Law 15/1980 creating the Nuclear Safety Council (amended by Law 14/99 and Law 33/2007): *CSN* shall issue reports to the Ministry of Industry, Energy and Tourism on nuclear safety, radiological protection and physical protection issues prior to the award of authorisations for nuclear and radioactive facilities as well as for the exploitation, restoration and closure of uranium mines.

CSN shall assess the environmental radiological impact of nuclear and radioactive installations.

*CSN* shall control and monitor the radiological quality of the environment throughout the national territory and cooperate with the competent authorities in matters related to environmental radiological supervision.

• Law 6/2010 on environmental impact assessment of projects.

## **3.3.3** Legislative acts regulating the radiological surveillance of foodstuffs

- Royal Decree 140/2003, of 7 February 2003, establishing the health criteria for water quality for human consumption, published in *BOE* n° 45 of 21 February 2003.
- Royal Decree 1744/2003, amending Royal Decree 1074/2002, regulating the procedure for the preparation, transport and sale of bottled drinking water.

Radiological monitoring of foodstuffs in areas around installations which discharge radioactivity is required of the owners of such installations in specific regulations and directives.

## **3.3.4** International legislative and guidance documents

- ICRP Publication 60. Recommendations of the International Commission on Radiological Protection, 1990.
- IAEA International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources. Safety Series N° 115, 1996.
- Council Directive 96/29/Euratom of 13 May 1996 laying down basic safety standards for the health protection of the general public and workers against the dangers of ionizing radiation.

- Commission Recommendation 2004/473/Euratom of 8 June 2000 on the application of Article 36 of the Euratom Treaty concerning the monitoring of the levels of radioactivity in the environment for the purpose of assessing the exposure of the population as a whole.
- Council Regulation (EC) 737/90 of 22 March 1990, on the conditions governing imports of agricultural products originating in third countries following the accident at the Chernobyl nuclear power station.
- WHO Codex Alimentarius Commission Guideline 5-1989, Guideline levels for radionuclides in foods following accidental nuclear contamination for use in international trade.

# 4 FORMER URANIUM MINING AND MILLING SITES IN SPAIN

## 4.1 INTRODUCTION

At the opening meeting and during the course of the verification, the team of Commission inspectors received extensive information concerning former uranium mining and milling sites in Spain. This information is detailed below.

Uranium mining sites:

- Saelices el Chico (Salamanca/Castilla y León) was in operation from 1974 to 2000. A total of 81 Mt of rock was mined to extract 12 Mt to produce 5750 t of U<sub>3</sub>O<sub>8</sub>. After closure of the mine, the mining area was restored from 2004 to 2008. After this, the compliance period started<sup>3</sup>. A monitoring programme for this site is under assessment.
- Valdemascaño (Salamanca/Castilla y León) was an investigation site, first (1957 to 1973) by the *Junta de Energía Nuclear (JEN)* and afterwards by *ENUSA*, until 2000. Site restoration took place from 2006 to 2007 and in 2008 the ongoing compliance period started.
- The La Virgen mine was in operation from 1954 to 1967. Restoration started in 1999 and was finalised in 2000.
- Apart from the uranium mines mentioned above, several other small mines were productive for some time (e.g. Casillas de Flores/Salamanca in the sixties and La Haba/Badajoz open pit mine, operational from 1983 to 1990).

Uranium milling sites:

- 'Quercus' (at Saelices el Chico) was in operation from 1993 to 2002. During this time 2322 tons of  $U_3O_8$  were produced. The activities were terminated in 2003. The dismantling and licensing processes were suspended from 2004 to 2012.
- *FUA* (*Fábrica de Uranio de Andújar*, at Andújar) was in operation from 1959 to 1981. During this time 1354 tons of U<sub>3</sub>O<sub>8</sub> were produced. Dismantling of the plant took place from 1991 to 1995. After this, the compliance period started. A radiological surveillance programme was started in 1995 and is ongoing.
- *'Elefante'* (at Saelices el Chico) was in operation from 1978 to 1993. During this time 3430 tons of  $U_3O_8$  were produced. Dismantling of the plant took place from 2001 to 2004. After this, the compliance period started. A surveillance programme was started in 2006 and is ongoing.

<sup>&</sup>lt;sup>3</sup> Compliance period is the period that comes after the completion of the dismantling of the facility. Its objective is to verify that the parameters included in the surveillance and maintenance plan are set up, and to check the suitability of the restoration and conditioning activities performed, and that they are accomplished as foreseen.

• The 'Lobo-G' site at La Haba/Badajoz is closed and remediated. The site was in operation from 1977 until 1990, and in dismantling phase between 1995 and 1997. After this a five year compliance period was approved with its corresponding surveillance and control programme. Finally, the Order issued by the Ministry of Industry, Tourism and Trade on August 2nd 2004 declared the decommissioning of the restored site of the facility and established an ongoing programme of long-term surveillance and control of the tailings dyke.

Table 1 lists the laboratories that are involved in the environmental radiological monitoring programmes for the areas visited, both with regard to site specific tasks (*PVRA*, QC, *PVRAIN*) and with regard to tasks within the national monitoring network (*REM*). Stations of the automatic network (*REA*) are indicated as well.

 Table 1:
 Laboratories and stations involved in the radiological monitoring programmes (site specific and nationwide) in the areas visited

Installation	Programme	Laboratory
		ENUSA Juzbado
	Operator's PVRA	CIEMAT*)
		Cantabria University**)
	Operator's quality control programme	CIEMAT*)
		GEOCISA*)
Andújar (FUA)	CSN complementary programme <i>PVRAIN</i>	Sevilla University
		Sevilla University
	<i>REM</i> (atmospheric and terrestrial environment)	Málaga University
		Granada University
	REM (aquatic)	CEDEX**)
	REA	CSN station Andújar
		ENUSA Saelices
	Operator's PVRA	ENUSA Juzbado
		Cantabria University**)
	Operator's quality control programme	CIEMAT*)
'Quercus'	CSN complementary programme PVRAIN	Salamanca University
	<i>REM</i> (atmospheric and terrestrial environment)	Salamanca University
	REM (aquatic)	CEDEX**)
	REA	CSN station Saelices el Chico
Operator's PVM		ENUSA Saelices
Valdemascaño		ENUSA Madrid
		ENUSA Juzbado
		Cantabria University**)

\*) Not part of this verification; was verified in an earlier mission \*\*) Not part of this verification

Highlighted in grey: part of national environmental radiological monitoring

## 4.2 **RESTORATION OF FORMER URANIUM SITES**

In December 1993, *ENRESA* submitted to the ministry responsible for energy its proposal for a restoration plan for former uranium mines. This proposal included all actions necessary to achieve the restoration of the mines formerly exploited by the *Junta de Energía Nuclear (JEN)* in Andalusia and Extremadura. The plan dealt with twenty six mining sites, most of which with underground works.

From 1997 to 2000, nineteen mining sites were environmentally restored. Nine mining sites did not need to undergo restoration since they were minor sites with very low extraction works performed for exploration purposes.

The actions included restoration of the morphology of the surface and of the landscape. All work was designed to minimise dispersion of radioactive mining tailings and other materials and to limit as far as possible public access to the underground mines. The principal goal of these works was reduction of the radion exhalation and reduction of the radiation levels in the environment to background levels at these sites. As far as ground water and superficial waters were concerned, the contact of these waters with waste rock was minimised.

#### 4.3 URANIUM MINING AND MILLING SITES IN ANDALUSIA

The restored mining sites in Andalusia are those of Trapero, Cano and San Valentín, situated in the natural park of Cardeña–Montoro (Province of Córdoba) as well as the mines of La Virgen, Montealegre and Navalasno, located in the natural park of Andújar (Province of Jaén).

In this area all former uranium mines were remediated (as far as deemed necessary).

## 4.3.1 La Virgen mining site – description and verification

The verification team verified the environmental radiological monitoring of the former mining site of "*La Virgen*". Originally this site started operation as a copper mine. Aerial and geological prospection of the site demonstrated that in addition to copper it also contained uranium.

The team noted that the area was fenced and locked to prevent public access.

A concrete slab blocks the former main shaft, which had a depth of ca. 120 m. The lowest level galleries of the mine were used for copper mining, whereas in the upper parts uranium mining took place. Currently the mine is flooded and the water level is at about 25 m from the soil surface.

In large parts of the former mining area mats have been placed on the soil surface in order to prevent erosion.

The area is very dry; currently there are no springs. Generally in the area springs originate only in top soil and not from the granite bed rock. The hills are mostly covered by low vegetation and trees.

The site is currently owned by a real estate company, which is also responsible for the nearby "*La Virgen*" sanctuary. The site is used as a hunting ground (deer); the park is known for its lynx population.

Several concrete ditches coming from the top of the reshaped slope are designed to prevent any heavy rainfall eroding the site. Six landmarks that are placed to control any surface movements were seen.

The verification team was informed that the objectives of restoration have been fully achieved and the site does not demand further monitoring since it is very dry.

The verifications performed do not give rise to any suggestions.

# 4.3.2 Andújar uranium mill – description and verification

The uranium production plant of the *Fábrica de Uranio de Andujar (FUA)*, situated in the province of Jaén, started its activities in November 1959 and was in operation until July 1981. The former *Junta de Energía Nuclear (JEN)* was the owner and treated there 1 218 098 t of uranium ore with an average uranium content of 1.23‰, producing a total of 1354 t of uranium concentrate.

The rather low grade ores from various mines situated in different geographical locations of Spain were shipped to FUA. The solid residues of the production accumulated in tailings covering an area of 9.4 ha, housing a volume of about one million m<sup>3</sup>. The original tailings reached a height of 10 to 20 metres and had slopes between 25 and 30 degrees.

*FUA* ceased activities by order of the ministry responsible for industry (at that time the *Ministerio de Industria y Energía*) of 30 June 1981. Further to this, by its order of 1 February 1991, the ministry attributed to *ENRESA* the authorisation to conduct dismantling and restoration activities at the site. The restoration plan included stabilisation and restoration of the tailings pile, which accumulates all residues of the production process. Production installations and buildings were dismantled and demolished and the residues incorporated into the above mentioned tailings pile, together with the contaminated soils. The overall goal of this design was confining all existing materials in a remodelled tailing pile with a slope of 5(H):1(V) achieving a stable configuration, and placing a final cover with a multilayer system.

The objective of this restoration was to prevent any dispersion of the contaminated residues and materials through a multilayer cover guaranteeing a designed durability of a minimum of 200 years with a goal of 1000 years.

Fundamental roles of the multilayer cover are (1) watertightness to reduce infiltration, (2) drainage to enhance evaporation and (3) erosion protection. Waterproofing is achieved with clay barriers with very low permeability in order to prevent that infiltration reaches the tailings and to avoid dispersion of contamination into underground waters. Also this layer acts like a barrier to prevent radon exhalation.

By its resolution of 17 March 1995 the Directorate-General for Energy within the ministry declared the period of execution of dismantling and restoration work of *FUA* finalised. By its decree of 1 February 1991, the ministry responsible for energy had foreseen after the finalisation of these activities a compliance period of at least ten years, which had to comply with the limits and conditions of nuclear safety and radiological protection laid down in an annex of this decree. The content of the programme to carry out during this period is included in another annex. The conditions include the implementation of an environmental radiological surveillance programme, whose results are evaluated by *CSN*.

After ten years none of the values forecast has been reached because the evolution of the controlled parameters was slower than initially foreseen. The restored site still remains in the above mentioned compliance period and stays within the responsibilities of *ENRESA*. According to this responsibility an environmental radiological surveillance and maintenance plan has been established for the duration of the compliance period; revisions are performed when needed. This maintenance plan includes the following areas: 1) Final site conditions reached after the decommissioning and restoration activities, 2) Final documentation "as built", 3) Monitoring of structures, 4) Monitoring of radon emissions, 5) Monitoring of ground and surface waters and foodstuffs irrigated with waters from surveilled wells, 6) Scheduled and extraordinary inspections, 7) Maintenance repairs and corrective actions.

*ENRESA* and *CSN*, regularly (at a minimum yearly) perform inspections to verify the general conditions of the environmental radiological monitoring of the site, hydrogeological parameters, the evolution and any changes of the cover of the restored tailings heap (by intrusion of animals and/or plants), stability of the pile, erosive processes, etc.

The original tailings heap was some 20 m high. All material including building material was moved to form the inside structure of a new heap (with a softer slope). *ENRESA* continuously studies the behaviour of the heap base and cares for erosion protection. (Initially a single layer cover was applied, but then a multilayer redundant cover was installed, different for slopes and the upper part.) Compared to the initial surveillance programme (from 1995), the current programme contains an increased number of wells, in

situ measurements, and every 2 years determination of chemical parameters. A study is planned to evaluate the possible changes of the flow behaviour of the nearby river Guadalquivir.

The verification team visited the site and verified monitoring structures and parameters. The total area is protected from public access by a locked fence; warning signs are in place.

Groundwater samples are taken quarterly in 24 boreholes and wells (7 inside the site and 17 outside, in the influence area). Also 4 superficial water points are sampled. The team verified the sampling points PC-2 and PC-3 inside the installation.

The team was informed that there are surveillance points to control levelling, eventual movements of the clay layer and to check the layer's integrity. There are six points in the surroundings of the heap and 10 land marks on the pile.

The team noted the presence of several iron plates (some 50 by 50 cm) that formerly were used for checking any movements of the pile using aerial photographic analysis.

*ENRESA* takes care of the vegetation; currently there are no trees on the pile. The heap vegetation is usually native with no deep rooting to prevent any changes of the (artificial) shape of the heap. The team witnessed that there are rabbits (trying to dig holes) and also other animals on the site.

At the site entrance and over the heap, *ENRESA* has installed two small monoliths with specific information of what is stored on the site (meant as information for future generations).

With regard to the control of any movement of the site, the team witnessed land mark 'H-3' on the top of the tailings heap.

ENRESA controls also any changes of the meanders of Guadalquivir River.

The verifications performed pointed out that there might be an issue of animals (rabbits) damaging the slopes of the site. The team suggests that CSN should follow up this issue during its inspections<sup>4</sup>.

## 4.4 CIUDAD RODRIGO AREA

*ENUSA* is in charge of environmental radiological monitoring and remediation of the former uranium mining and milling sites in the Ciudad Rodrigo area, in particular the large Saelices el Chico site, as well as other former mines in the surroundings such as Valdemascaño and Casillas de Flores.

## 4.4.1 Saelices el Chico site – description and verification

The team visited the Saelices el Chico site with the Saelices laboratory, the dismantled '*Elefante*' uranium concentrates manufacturing plant, the '*Fe*' and '*D*' mines and the '*Quercus*' mill. It witnessed that the entire site is fenced and all entries are controlled. Figure 1 provides an aerial view of the Saelices el Chico site.

During the verification of the site, which hosts also the local *ENUSA* headquarters, the team noticed that – before arriving at the entrance control point – at the right side of the road is a fenced pond (*BAC*) with a maximum capacity of  $400\ 000\ m^3$  and a red colour stemming from pyrite. There is also a second storage pond (*BRV*) both for the storage of water stemming from former restored open pits and waste dumps (prior to chemical treatment). At the moment of the verification, due to a long very dry period (dry season), there was no water in these ponds.

Altogether the site comprises about 1600 ha; half of it affected by mining and milling activities, including construction works. Most of it is already remediated.

<sup>&</sup>lt;sup>4</sup> After the verification visit, this issue has been taken up within the scope of the inspections of the Geosciences Branch (*CITI*), unit of the *CSN*.

A dam ('Dique Majuelos') was built to stop water from the Arroyo de Majuelos entering the 'Quercus' tailings dam area and later to the restored 'Elefante' site. Main materials (sandy clay, 'arcosas') used for mining restoration came from different quarries near the site. The most important is situated at the left side of the entrance road near the site entrance gate. This quarry was used for the construction of the tailings dam and for capping the tailings heaps.

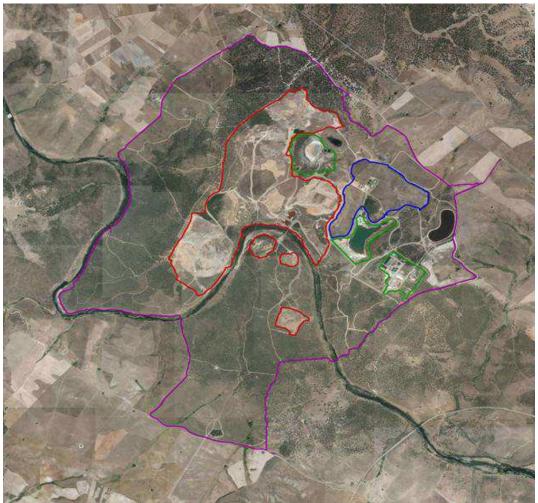


Figure 1: Aerial view of the Saelices el Chico uranium mining and production site [magenta: *ENUSA*'s farm, green: '*Quercus*' facilities, red: mine facilities, blue: '*Elefante*' facilities]; source: *ENUSA* 

## 4.4.1.1 '*Elefante*' plant

The '*Elefante*' plant was active from 1976 to 1993 and produced 3430 t of  $U_3O_8$  (with an average of ~200 t per year) using the heap leaching process (mineral stockpiles). The plant was dismantled from 2001 to 2004. Since 2006 a compliance period environmental monitoring and control programme is in place. Till now no significant events have been reported.

Dismantling activities (2001-2004) consisted of:

- In situ stabilisation of former heap leaching stockpiles: 7.2 Mt, final slope < 20%, surface 68 ha.
- Dismantling of industrial plants, storing wastes in a containment enclosure under depleted mineral and capping.

- Construction of protection layers (capping):
  - 0.9 m of clay and arkoses (to avoid water infiltration, radon exhalation and radiation);
  - 0.9 m of selected slate from waste dumps (to avoid erosion);
  - $\circ$  0.5 m of top soil (to improve the re-vegetation of the environment).
- Technical and radiological controls.

The heap leaching site is covered with clay and high density polyethylene (PE) layers. Studies are being performed to identify possibilities for improved remediation.

The 'Elefante' plant did not form a specific part of the verification.

#### 4.4.1.2 Mining exploitations and restorations at the 'Quercus' site

The most common uranium mineralisations on the 'Quercus' site were:

- pitchblende and black oxides (with pyrite and carbonates), filling fractures in slates and schists;
- uranium sulphates and others.

From 1974 to 2000, 81 Mt of rocks were mined in this area containing 12 Mt of uranium ores (average grade  $U_3O_8$  650 ppm, with an economical cut-off at 200-325 ppm). From this ore, 5750 t  $U_3O_8$  were produced with a maximum of 300 t  $U_3O_8$  per year.

In 2000 production was ceased because of mining difficulties and worldwide low uranium prices. The site comprised four pits of 15 Mm<sup>3</sup>, a surface of 75 ha and eight waste dumps of 35 Mm<sup>3</sup> with a surface of 175 ha.

Decommissioning and restoration of the area started in 2001 and is still on-going. Open pits of 15  $\text{Mm}^3$  (with important volumes of acid waters), waste dumps of 35  $\text{Mm}^3$ , mineral stock piles of more than 4  $\text{Mm}^3$ , tailings dumps of more than 1  $\text{Mm}^3$ , as well as the metallurgical facilities of the '*Elefante*' and '*Quercus*' plants had to be dealt with in this context.

The goal of the dismantling and restoration activities of the area was to retrieve similar environmental conditions to the preoperational ones (background radiation levels).

The current status is that the site has been restored (2004 to 2008). A monitoring and control programme has been proposed. The restoration consisted of:

- Geomorphological restoration: filling of mining pits with waste from dumps and stabilisation of the rest of waste dumps by levelling (final slope <20%);
- Hydraulic actions: protection for gullies, new canals (10 km), new ponds and dams (0.2 Mm<sup>3</sup>);
- Revegetation: 250 ha;
- Water management: collection, treatment, drainage, discharge, seepages and leachate control;
- Others: infrastructure, buildings, roads...

#### 4.4.1.3 Treatment of contaminated waters from the 'Quercus' site

Contaminated water (surface-, seepage-, mining-, groundwater) stems from the acidic lixiviation of uranium and other metals from the soil in the mining areas (former pits, waste dumps and other non-restored areas). Currently, per year ~  $500\ 000\ m^3$  of water are collected from contaminated sectors (former big open pits, former heap leaching piles, former waste-dumps, etc.).

One month before the visit the water treatment plant stopped operation because due to the extremely dry season there was no water to be treated.

It is still necessary to treat waters because they are very acidic (pyrite occurrence in ore body rocks), even with mining restoration done. Although the volume has been significantly reduced, approximately 500 000 m<sup>3</sup> of mine water are treated per year before discharging to the river Águeda. In the treatment process radium is precipitated using BaCl<sub>2</sub>. By raising the pH to a high value (~ 10), manganese is co-precipitated. After precipitation the pH is brought back to 7. The remaining solid phase is treated with a thickener to form 'pies' (ca. 0.5 m in diameter). After treatment and monitoring, the 'clean' water flows to the nearby river. The solid phase ('pies') is shipped to the tailings dam ('*Dique de Estériles*') and also to the '*Quercus'* heap leaching stockpile.

The current treatment is expensive and would have to be performed for a long time. Other approaches are currently discussed with companies such as *Rio Tinto, Billiton* and *AREVA*. A US company working for the US Dept. of Energy has recommended actions to reduce volume and costs (e.g. by using special sprinklers). Such an approach is used in the USA to enhance evaporation; it will be tested at the site in 2013. At the time of the verification evaporation was very high due to extremely dry weather. In normal times collected water first runs by gravity to a pit situated close to the nearby river Águeda. From there it is pumped to ponds feeding the treatment plant. The team verified the site and noted the dark brown colour of the liquid to be treated.

## 4.4.1.4 'Quercus' plant

The 'Quercus' uranium concentrate production plant was active from 1993 to 2000 and produced 2240 t of  $U_3O_8$  (average of 300 t per year). The plant used the dynamic heap leaching process of crushed material with sulphuric acid. In 2001 and 2002 there was a small residual production. Exploitation was definitively stopped in 2003. Currently the plant is still on standby. The verification team was informed that a final decision about project dismantling should be taken before the end of 2012. The reason is that originally plans of using (parts of) the site in collaboration with the Australian company *Berkeley Resources Limited* were investigated. However, recently the plans were abandoned.

Restarting dismantling needs a new application. According to *CSN* in this case within three months a planning programme for updating the dismantling plan has to be established. Within one year the full new plan must be submitted. Once dismantling activities have finished, adjusted monitoring would have to take place during the compliance period.

Currently contaminated waters are treated by chemical neutralisation. An environmental surveillance and control programme is in place. Till now no significant events have been reported.

## 4.4.1.5 Site visit

The verification team, after having received information at an opening meeting, visited the site. Currently 735 000  $\text{m}^3$  of solid tailings (from sedimentation) are stored in former open mine pits (the total capacity is 2.1 billion  $\text{m}^3$ ). One pit was not totally filled and is used for rain water collection.

The dam forming the barrier to the restored '*Elefante*' site is seismic proof (7 - 8 Richter) although the site is not at seismic risk. The team witnessed monitoring point PMM-3 (*Punto de mira móvil*) for control of dam movements situated close to the road.

Further on, the team noted that the former heap leaching area, near the pit 'FE3', is filled with the filter cakes – 'pies' – from the treatment plant.

Altogether there are some 250 ha of the area already restored and revegetated. The team was told that generally, trees in the area are shallow rooting. For the capping of all the restored structures deep rooting trees are not desirable, thus local bushes are used instead. Some 25 years ago fast growing pines were planted. Until the time of the visit, *ENUSA* and *ENRESA* have spent about 90 M $\in$  on restoration. More than 22 Mm<sup>3</sup> of soil and tailings have been moved.

The Águeda River that meanders through the site flows to Portugal. *ENRESA* has an information exchange with Portugal concerning monitoring programmes of this river.

The team visited the installations for water collection and treatment. There is an intermediary pond before the treatment installations with a pumping station (not far from the river). Four special pumps (adapted to acidic water of a pH about 3) pump water from the pond to the treatment station when needed.

This pond is divided by the road into two parts, which. are linked by tubes that can be closed by valves.

Furtheron, the team visited one of the water treatment plants (*TAC*). The other one (*TAD*) is situated in the West part of the *ENUSA*'s property.

At *TAC* feed water enters the plant and serves an installation consisting of  $2 \times 2$  reactors. At the time of the visit, the reactors were open for cleaning of lime incrustations. When operating, mine water is treated by the addition of lime (the amount being regulated based on acidity measured by pH meters and using pumps with controlled flow) and of a BaCl<sub>2</sub> solution. *'Nalco'* is used for improving flocculation in a later step to remove solids from liquid solution ("clean water").

The resulting slurry is pumped to a nearby cabin housing a filter press with a series of chambers (thickness ca. 2 cm each). The press applies some 200 bar pressure and produces solid filter 'cakes' ('pies'), which are deposed in a special area of the site. TLDs and Rn dosimeters that were mounted in the cabin and form part of the occupational control system.

After flocculation, 'clean' water overflows from the thickener and the filter-press; at this point it has a pH of 10. By addition of hydrochloric acid it is brought to pH 7 and released to a pond in which its pH is checked once more. From there the water is piped through a discharge channel to the river. (Water from the filter press is recirculated to the pH control point.)

An automatic sampling point is installed in the discharge channel. Every 30 minutes a sample is taken. This requirement was established when the uranium production plant was in operation, and it is still applied.

At the sampling point two pipes (PVC) arrive from the *TAC* and *TAD* ponds. Water is collected in one concrete basin (common metal pipe). From there automatic sampling with two devices takes place (in parallel with one being a backup); two 301 containers ('RVM3(1)' and 'RVM3(2)') are used. From there two pumps discharge water through a pipe to the river.

Measurements of conductivity and pH are performed in a continuous manner. The outflow rate is measured by two redundant methods (ultrasonic discharge meter in bypass and an old mechanical registering device, *SEBA Hydrometric*, Kaufbeuren, Germany).

With regard to on-site environmental monitoring the team verified the presence of the thermoluminescence dosimeter (TLD) TE-8 and the ground water sampling point ES09.

The team was shown the 'Quercus' extraction building where organic solutions with kerosene had been used. All equipment from the mechanical ore process (crushing, wet grinding and mineral stock) was dismantled in 2005. The whole 'Quercus' plant was left in case of an agreement with Berkeley Resources Ltd. for re-use.

The verification team would like to point out that any restarting of operations could lead to significant changes of the monitoring programme. With regard to future decommissioning the issue of long-term preservation of knowledge about the site (e.g. developing methods not depending on sophisticated means such as electronic information storage devices) and securing resources for appropriate monitoring should be considered.

# 4.4.2 Valdemascaño uranium mine – description and verification

The verification team visited the former Valdemascaño uranium mine, located north of Saelices el Chico, near Lumbrales.

The Valdemascaño mine was a small underground uranium mine exploited in the 1970s. It is located in a granitic area. The mine was about 200 m deep and had two mine shafts (linked with tunnels) and two chimneys for ventilation. The extracted ore was leached in the adjacent ore preparation area.

All administration and laboratory buildings have been dismantled and the shafts closed. Resulting material was moved to one place (shaft 2 area) which was also surrounded by a drainage area. This area (ca. 610 m a.s.l.) and the first shaft (ca. 605 m a.s.l.) have been covered with three material layers: clay, gravel (40 to 80 mm diameter) and a cap of soil for vegetation.

The rest of the site was evened out and covered with one soil layer, smoothed out and reshaped. Currently the site can hardly be distinguished from the neighbouring non-mining areas.

Remediation works were carried out in 2007, followed by a compliance period. A monitoring programme started that year and is still ongoing. Surface water is sampled twice a year (April/May and November/December) in three locations on site and in three off site. Radon exhalation is monitored by the University of Cantabria at 25 points once a year (April/May). At each sampling point five capsules with dosimetric film are exposed. At the beginning the university used CR39 film material, now they are using their own specific material. Exposure is for 24 hours, then the films are track etched and the tracks analysed. Also gamma radiation is monitored by scanning at more than 150 points, and vegetation samples are taken yearly.

The site is fenced with barbed wire. All relevant places are marked as topographic points for identification. The team verified some of these points -'pozo1', 'pozo2', 'chimenea1', 'chimenea2' - marked in orange. The site also has drainage ditches which were originally planned to be lined with concrete, but the owner found that they would be better done without concrete. The team noted erosion effects in these ditches.

The verification team suggests considering concrete protection for the drainage ditches if meteorological conditions so require.

# 4.4.3 General situation of uranium mines in the area

At a meeting with representatives of *CSN*, *ENUSA* and the regional authority of Castile and León the verification team discussed the situation of possible uranium mining in the area in the future and the issue of keeping knowledge about the sites for long times.

The team was informed that there are six other former uranium mines in this area. Three of them have been restored, but for three there is still discussion between the autonomous government of the region and the State about who shall be responsible. (The team was told that an agreement was near.) Verification of these sites was not included in the agenda, but the team was informed that some mine shafts are covered and some places are fenced to prevent access.

With regard to any future U mining there are State reserves (due to the national interest), for which mining rights are with the State. Others (e.g. the Australian company *Berkeley Resources Ltd.*) have received mining rights from the respective autonomous community. According to the *Berkeley Resources* website at the time of the verification no definite decision about starting uranium mining had been made.

The verification team recommends clarifying responsibilities for the remaining former uranium mines and implementing restoration and monitoring programmes.

## 5 SPANISH NATIONAL ENVIRONMENTAL MONITORING PROGRAMMES

## 5.1 INTRODUCTION

*CSN* runs a nationwide radiological monitoring network, the *Red de Vigilancia Radiológica Ambiental no asociada a instalaciones (REVIRA*; Environmental Radiological Monitoring Network Not Associated With Installations), which is distributed throughout the national territory. It is formed from a network of sampling stations (*Red de Estaciones de Muestreo, REM*) and a network of continuously measuring automatic stations (*Red de Estaciones Automáticas, REA*). *REVIRA* provides nationwide radiological information on the radioactivity in the air, soil, water (drinking water, fresh water and sea water) and of foodstuffs. This national monitoring network is independent from the networks associated with nuclear facilities. It includes almost all autonomous regions and accounts for features such as coastal lines when establishing the number and characteristics of the sampling points.

The autonomous administrations of Valencia, Catalunya, País Vasco and Extremadura run their own regional automatic monitoring networks. In addition, the Ministry of the Interior has a radioactivity warning network (*Red de Alerta de la Radiactividad, RAR*) composed of 900 gamma dose rate measurement points distributed all over Spain. In the *CSN* emergencies room (*Sala de emergencias, SALEM*) there is a computer connected to a *RAR* control centre which provides real-time information.

#### 5.2 THE NATIONAL MONITORING NETWORK *REVIRA*

*REVIRA* is composed of:

- The network of sampling stations (*REM*). Monitoring is achieved through sampling and analysis programmes carried out by different laboratories. *REM* consists of:
  - The atmospheric and terrestrial environment monitoring programme (air, soil, drinking water, milk, mixed diet; see table 2).
  - The aquatic environment (inland and coastal waters) monitoring programme.
- The network of automatic stations (*REA*), enlarged with the connection to the autonomous administrations' automatic networks of Valencia, Catalunya, País Vasco and Extremadura under specific agreements. *REA* carries out continuous measurement and provides information in real time on radioactivity concentrations in the air and in water and on ambient dose rate levels in different areas of the country.

The *REVIRA* network has been operational since 1992 (river monitoring since 1984) with the following functions:

- Ascertain the distribution and evolution of radioisotopes present in the environment and the levels of environmental radiation;
- Provide an environmental database to be able to obtain reference levels at any time;
- Provide experimental data for estimating the potential radiological impact on the population as a result of possible radioactive contamination of the environment;
- Provide data for reporting to the Congress, Senate and to the public on the radiological quality of the environment in Spain.

Type of sample	Frequency of sampling	Dense network		Sparse network	
		Type of analysis	Frequency of analysis	Type of analysis	Frequency of analysis
Aerosols	Continuous sampling Weekly change of filter	Total alpha Total beta Gamma spectr. Sr-90	Weekly Weekly Monthly Quarterly	Cs-137 (gamma spectr.) Be-7 (gamma spectr.)	Weekly Weekly
Radioiodine	Continuous sampling Weekly change of activated carbon cartridge	I-131	Weekly		
Soil (Total deposition)	Annual	Total beta Gamma spectr. Sr-90	Annual Annual Annual		
Drinking water	Monthly	Total alpha Total beta Gamma spectr. Sr-90	Monthly Monthly Monthly Quarterly	Total alpha Total beta Residual beta H-3 Sr-90 Cs-137 Natural isotopes	Monthly Monthly Monthly Monthly Monthly Biennial
Milk	Monthly	Gamma spectr. Sr-90	Monthly Monthly	Sr-90 Cs-137 (gamma spectr.)	Monthly Monthly
Mixed diet	Quarterly			Sr-90 Cs-137 (gamma spectr.)	Quarterly Quarterly

Table 2: REM - Programme of sampling and analysis of air and the terrestrial environment

The implementation of Article 35 of the Euratom Treaty regarding nationwide radioactivity monitoring in Spain is done by setting-up (i.e. logically defining) a 'Dense' and a 'Sparse' Network as follows:

- The *Dense Network* is composed of numerous sampling points all over the territory. The required detection levels of that network are of low sensitivity. This network is currently composed of 25 REA points, 18 air sampling points, some 85 inland surface water points, 15 seawater points along the Spanish coast, 15 drinking water points, 18 soil sampling stations and 15 mixed diet sampling locations. The analysis of most milk samples is also implemented within this network.
- The *Sparse Network* is composed of very few sampling points with a very high sensitivity of detection. This network currently has 5 high-volume air sampling stations, 5 drinking water and mixed diet sampling points and 4 milk sampling points.

## 5.2.1 Network of automatic *REA* stations

The automatic *REA* station network has been established for performing real-time monitoring of atmospheric radioactivity. The network is made up of 25 stations spread throughout Spain and one in Portugal (Penhas Douradas). In addition, under specific agreements with the autonomous administrations responsible for these networks, *CSN* has integrated stations in the Valencia, Catalunia, País Vasco and Extremadura networks into the *REA* management and operation system.

The CSN-managed network stations (with exception of the one in Madrid, located at CIEMAT, and the one in Penhas Douradas, Portugal) are located in measurement stations of the National Meteorology Agency (Agencia Estatal de Meteorología, AEMET, formerly INM). They share the communications system with the meteorological network. Table 3 shows the parameters monitored by the automatic stations.

Radiological Data	Meteorological Data
Gross alpha activity Gross beta activity I-131 Rn-222 Gamma dose rate	Wind velocity Wind direction Air temperature Air humidity Rain intensity Air pressure

The stations take measurements continuously. The monitors installed in each station are incorporated into a single piece of equipment, which has its own electronics for communication and data management. Each station is connected by switched telephone network, regular subscriber's line, to a central computer installed in the emergencies co-ordination centre *SALEM*, in Madrid (verified during the Trillo verification of 2004). *SALEM* has the appropriate software for archiving and processing the data transmitted by the stations and acting as communications manager. There is an alarm function which alerts in the event of the detection of anomalous measurements.

*CSN*'s web site provides information on the daily average dose rate value and the average dose rate value for the previous thirty days at each *REA* station, as well as a historical file of the same data.

The verification encompassed the *REA* stations in Andújar and in Saelices el Chico. *CSN* plans to renew all stations.

## 5.2.1.1 REA Andújar station - verification

The *REA* station in Andújar is well located in a very wide valley on a flat terrain with gravel cover, near to the former Andújar uranium mill within the fenced area of the local meteorological station of the National Meteorological Agency. At about ten metres there are a one storey building and a few very small trees.

The measurement of <u>ambient gamma dose rate in air</u> is obtained using double chamber Geiger-Müller probes (measuring range  $10^{-2}$  to  $10^7 \,\mu$ Sv/h). Two devices are mounted on the meteorological mast of the station at an effective height of about two metres from the ground: A new probe (Ser.Nr. 6204, Rev00) is currently used to measure ambient gamma dose rate; An old probe with 2 built in GM tubes (for low and high dose rate), is still used as a backup. Both devices are from *Berthold*, Germany. Communication to the electronics of the system in the nearby building is done by cable (to data logger *SEAC* with modem). Data transmission to Madrid takes place every hour. The *SEAC* company (*Sociedad Española de Aplicaciones Ciberneticas, S.A.*) is responsible for servicing the devices all over Spain, including the air monitors.

For the measurement of <u>aerosol activity</u> the installed equipment consists of a *Berthold BAI 9100-D* step filter band device (advances by 40 mm/hr) with a ZnS(Ag) scintillation detector which, through a system of pseudo-coincidence, permits artificial radioactive aerosols to be measured in the presence of natural radioactivity. Filter material is glass fibre. The device provides information on  $\alpha$ ,  $\beta$  and radon activity. The measuring range for alpha and beta activity is 0.2 to  $0.5 \times 10^7$  Bq/m<sup>3</sup>. The nominal air flow rate is about 6 m<sup>3</sup>/hr; at the time of the visit the flow rate was 5 m<sup>3</sup>/hr.

Measurement of <u>radioiodine in air</u> uses a *Berthold BAI-9103-1* monitor. Gaseous iodine is adsorbed on an activated carbon filter. Gamma emitters are analysed by a NaI(Tl) detector, with a single channel, fixed at the energy of I-131, symmetrically paired with a second channel which permits a dynamic background subtraction. The iodine monitor is heated to 40°C. The iodine cartridge (TEDA impregnated activated carbon from *Hi-Q Environmental products Co.Inc.*, San Diego, USA) is changed once per month. Used cartridges are stored at *SEAC*. The measuring range is 0.5 to  $10^7$  Bq/m<sup>3</sup>.

The team was informed that *CSN* staff visits the station three times a year. Maintenance is performed twice a year and there is one yearly check by *SEAC* (evaluation, calibration using Am-241 and Cl-36) based on a service contract with *CSN*.

The team noted the equipment production date 6/91; no calibration or check labels were on the devices. Staff from *SEAC* was present for explanations: service logs are done on a notebook computer that also has data connection to *CSN* (by GSM).

All the above described systems are designed to work in the presence of high radiation fields. The detectors of the air monitoring system are protected by lead shields of up to 5 cm.

The verification team suggests marking control and calibration dates on the respective devices in order to allow quick and efficient control of such tasks by CSN.

## 5.2.1.2 REA Saelices el Chico station - verification

The *CSN* measurement equipment at Saelices el Chico is located in a container at the meteorological station of the National Meteorological Institute in an isolated fenced area some 200 m from the mining facility.

The temperature-controlled container houses a *Berthold BAI 9100-1* continuous mixed nuclide monitoring system for airborne iodine, alpha/beta and radon. In addition there are monitoring devices for dose rate (Geiger-Müller), temperature, humidity and rain intensity.

Air alpha/beta is monitored with an on-line sensor a with a step filter band system. The air flow is about 6  $m^3/h$ . The filter paper roll is changed every 4 months. There is also a carbon filter cartridge for iodine measurement; this cartridge is changed monthly.

Radiological and meteorological data are transferred to *CSN* via a modem connection every hour. In the event of a power failure the station is able to transfer dose rate data autonomously for eight hours.

The meteorological site has also one of the site TLDs (No. 17), two radon detectors (track-etch) and a low volume air filter station (*ENUSA*).

Verification does not give rise to recommendations.

# 5.2.2 Network of *REM* sampling stations

The sampling station network *REM* includes monitoring of the aquatic medium, the atmosphere, soil, drinking water and foodstuffs. Apart from some aspects that were dealt with during the visits of the laboratories it was not particularly included in the verification.

To execute the national programmes *CSN* has entered into specific collaboration agreements with the following organisations:

- The civil works studies and experimentation centre (*CEDEX*) of the Ministry of Public Works (*Ministerio de Fomento*) which has been carrying out radiological monitoring of Spain's major rivers since 1978. *CEDEX* also provides *CSN* with the results of radiological controls of drinking water in different areas in the country.
- 19 university laboratories, which conduct the sampling programmes on university campuses;
- CIEMAT.

The collection of samples is the responsibility of the laboratories, which have the necessary equipment. Sampling is carried out in accordance with written procedures which must be adjusted to the requirements established in the agreements.

The analyses are also carried out in accordance with written procedures which guarantee the detection levels required for these programmes.

In the case of gamma spectrometric measurements, the isotopes on which information must be supplied are, as a minimum:

- Natural isotopes: Be-7, K-40, Tl-208, Pb-212, Bi-214 and Pb-214.
- Artificial isotopes: Cr-51, Mn-54, Co-58, Co-60, Fe-59, Zn-65, Nb-95, Zr-95, Ru-103, Ru-106, I-131, Cs-134, Cs-137, Ba-140, La-140 and Ce-144.

The laboratory must also provide information on other isotopes if above the detection limit.

## 5.2.2.1 Air samplers

The equipment used in the **dense network** for the collection of samples of particulate material operates at aspiration rates between 1.8 and 3 m<sup>3</sup>/hour (low volume), with weekly gross alpha and gross beta analysis on each filter, monthly gamma spectrometry analysis and quarterly strontium-90 analysis on all the filters accumulated in each sampling station. Cellulose and glass fibre filters are used.

The **sparse network** aerosol samples are collected on a weekly basis using equipment with a flow rate of approximately  $900 \text{ m}^3$ /hour (high volume). The filters are analysed by gamma spectrometry with extended measuring times of some 72 hours. The filters used are of polypropylene.

## 5.2.2.2 Surface water sampling programme

*CSN* has signed a specific agreement with the *Centro de Estudios y Experimentación de Obras Públicas* (*CEDEX*) to participate in the programme for the radiological monitoring of the waters of all the basins of Spanish rivers. The programme was established by *CEDEX* and has been operational since 1978; monitoring of coastal waters was added in 1992. Organisationally *CEDEX* belongs to the *Ministerio del Fomento* (Ministry of Public Works and Transport), functionally to both the *Ministerio del Fomento* and the *Ministerio de Agricultura, Alimentación y Medio Ambiente* (Ministry of Agriculture, Foodstuffs and the Environment),

The sampling stations for inland waters are located along the rivers of the various hydrographic basins, both in zones potentially affected by the nuclear and fuel cycle installations and in areas at a distance from these. There are more than eighty sampling points. *CEDEX* undertakes the sampling, with the collaboration of the Hydrographic Confederation's personnel, and analyses the samples.

The collection of the samples is generally manual. Frequency of collection and analysis at each station can be quarterly, monthly or fortnightly. In the latter case continuous proportional collection equipment is used.

The determinations made in respect of the samples collected are:

- Gross alpha activity
- Gross beta activity
- Residual beta activity
- Tritium
- Gamma spectrometry

The coastal waters monitoring programme currently includes 15 sampling stations with quarterly sample collection and with the same analytical determinations being made as for the fresh water samples.

## 5.2.2.3 Drinking water sampling programme

Tap water samples are collected monthly, about 5 l in the dense network and up to 1000 l in the sparse network in order to lower the gamma spectrometry detection limits. The determinations made are shown in table 2.

## 5.2.2.4 Soil sampling programme

Five soil samples of 5 cm depth (at each corner and in the middle of a square) are taken in an area of one square meter. These five samples are pooled in a plastic bag and provided to the laboratory for analyses. If the soil surface does not permit the previously described sampling method, the samples can also be taken in one row. Soil sampling at *REM* sites is performed once a year in accordance with the procedure described in a published regulation.

## 5.2.2.5 Foodstuffs sampling programme

## <u>Milk</u>

Milk sampling is performed under the *REM* scheme in the provinces or autonomous regions of Cantabria, Léon, Cataluña, Sevilla, La Coruña and Oviedo.

## <u>Mixed diet</u>

Standard diet samples are collected in the canteens of universities or institutions charged with the programme and consist of the complete diet of one person over five consecutive days. The sample is prepared using solely the edible parts.

## 5.2.2.6 Quality assurance arrangements

*CSN* requires technical documentation to be provided by each of the laboratories participating in the sampling station network (*REM*). This documentation must include the following:

- A description of sampling, detection and measurement equipment;
- Sampling, analysis and measurement procedures used by the laboratory;
- Quality assurance programme for the measurements made;
- Results of participation in analytical inter-comparison exercises organised by *CSN*.

Quality systems which integrate the organisation's structure, responsibilities, procedures, processes and resources required for suitably managing quality have been implemented. In 1997 *CSN* requested also that participants in *REM* develop quality manuals and programmes for establishing and implementing them.

In order to verify that the established quality assurance programmes are properly enforced, internal controls are introduced into the organisations. Proper external actions are taken, such as comparative inter-laboratory studies and audits. Since 1992 *CSN*, in collaboration with *CIEMAT*, has been undertaking annual analytical inter-comparison campaigns among Spanish laboratories measuring environmental radioactivity, using samples similar to those analysed in the environmental radiological monitoring programmes.

# 5.2.3 Transmission of monitoring data and records

Monitoring data from the automatic network (*REA*) are continuously transmitted to *CSN* and made available in the emergency centre *SALEM*. Data from the *REM* system are transmitted by email, diskette or telematic system and by biannual reports to *CSN*. Additionally, data from the sparse network are transmitted by email as soon as they are available. All of them are stored in a database ('KEEPER') together with data from environmental radioactivity measurements.

## **5.3** MOBILE MEASUREMENT SYSTEMS

*CSN* has mobile environmental radiological monitoring units for use in cases of an emergency, through agreements signed with *CIEMAT* and with the *Junta* and the University of Extremadura.

These mobile environmental radiological monitoring units were not included in the present verification.

## 5.4 *RAR* NETWORK

The radioactivity warning network *RAR* (*Red de Alerta de la Radiactividad*) is an emergency network operated by the Ministry of the Interior. It was not included in the present verification.

## **6** VERIFICATION OF LABORATORIES

#### 6.1 INTRODUCTION

The following chapters describe the verification activities performed in the laboratories involved in environmental radioactivity monitoring (studies and routine programmes).

*CSN* does not have an own environmental laboratory or sampling capabilities. Therefore the Spanish authority's independent national environmental radioactivity monitoring programme is outsourced to regional laboratories in the universities. Currently there are 20 regional laboratories participating in the programme. Seven of these laboratories are accredited according to ISO 17025 and some 5 more are in process; most of them are also ISO 9001 certified. All of the laboratories also participate in International inter-laboratory comparisons (EC-REM, IAEA, etc.). *CSN* performs the supervision and control of the quality of these laboratories through the assessment of the results obtained by them and the performance of technical visits on site. Furthermore, *CSN* fosters and coordinates the development of procedures on sampling, conservation and preparation of samples, analytical methods and measuring equipment, and requires the laboratories to apply them.

This arrangement maintains local expertise and education in radiological monitoring, but at the same time it may lack efficiency: some of the participating laboratories analyse fairly small numbers of samples and do not necessarily have the resources to maintain qualified staff and modern analytical capabilities. Although *CSN* provides guidance documents the arrangement may also lead to small differences in analysis methods and calibrations.

## 6.2 UNIVERSITY OF SEVILLE

# 6.2.1 *CITIUS*

*CITIUS* (*Centro de Investigación, Tecnología e Innovación de la Universidad de Sevilla*) is part of the University of Seville. General tasks of *CITIUS* were already verified in 2009 (Huelva verification); thus part of this verification report relies on that visit. *CITIUS* has collaboration with *CSN* regarding NORM industries in general, environmental radiological monitoring of former uranium mining and milling sites and with regard to the national environmental radioactivity monitoring network.

At the *Servicio de Radioisótopos* the verification team received detailed explanations from the scientific advisor (who is also professor and head of the research group "applied nuclear physics" of the University of Seville).

In the institute, three permanent positions are provided with persons employed full time, additionally the laboratory maintains collaboration with PhD students (requirement established in the quality manual by CSN).

The team was told that some 30-50% of the *CITIUS* premises (including the laboratory equipment) were financed by the European FEDER programme.

The *Servicio de Radioisótopos* of the University of Sevilla has an agreement with *CSN* by which the laboratory is charged to perform once a year an independent environmental radiological monitoring programme of the former *Fábrica de Uranio de Andújar (FUA)*, in addition to the owner's programme. The institute performs sampling itself. Sampling points are decided by the university based on the annual programme established by *CSN* (which is 'linked' to the owner's programme). Sampling includes ground water from wells and surface water from the Guadalquivir River. *FUA* is informed about the date of sampling, but not about details. Generally, sampling takes place in spring and samples are transferred directly to Seville.

The team received a list of samples. It was told that for the QC programme samples are split thus the lab has the 'same' sample as the operator.

*CITIUS* works also on a commercial basis for industry, receiving samples as "black boxes" (without information e.g. about the origin) and returning analysis results.

The service has started preparations to obtain ISO 17025 accreditation for its measuring tasks. *CITIUS* as a whole is ISO 9001 certified.

#### Radiochemistry laboratory

Sample registration is done in the radiochemistry laboratory. Normally the client's sample code is kept, with an internal number added. The registry is on paper using measurement specific sheets with the measuring tasks to be performed added to the sample description. The sample code consists of a running number and the year of sampling. Each sample is accompanied by a detailed sampling sheet (no signature of sampling person, since it is always the same). Additionally an analysis sheet is generated in which remarks are entered by hand during work; later on these data are transferred to a computer (final result sheets are signed and stored on paper and electronically).

The radiochemistry laboratory is also responsible for the preparation of the tritium samples (including tritium enrichment by electrolysis). Control of the electrolysis process is done using tritium spiked water. As background water for tritium determinations *Solan de Cabras* mineral water is used. (The owner of the water source does tritium measurements within his water quality control programme.) The overall limit of detection for H-3 achieved at the *servicio de radioisótopos* is some 0.2 Bq/l.

The team witnessed also the presence of a *Carbolite* furnace.

#### Gamma spectrometry

The laboratory houses a special Anti-Compton gamma spectrometry system with a well type HPGe detector (*Canberra*, horizontal cryostat), a large, horizontal, cylindrical lead shield, and a 5" well type NaI(Tl) detector with six photomultiplier tubes for anti-coincidence counting. It uses *Canberra* electronic NIM devices and *Canberra Genie 2000* as data acquisition and analysis software. The system is mainly used for determination of Pb-210 for dating. Since the visit in 2009 the system was moved to the basement in order to benefit from the lower background radiation in that place.

The verification team observed also a mobile gamma spectrometry system based on a *Canberra Inspector* 2000 device. The whole system that contains a notebook PC for the spectrometry and data management programmes is installed on a motor driven 4-wheel-frame. All devices, including the lead shield consisting of individual rings, can be dismounted.

With a view to avoid any disturbances in the measurements all PCs for handling the systems including gamma spectrometry software have been set up in the anteroom (with a large connecting window to the measuring room for easy observation).

#### <u>Alpha/beta laboratory</u>

The the laboratory houses two LSC devices of the type *Quantulus 1220*, an old one (supplied by *LKB Wallac*) and a newer one (supplied by *Perkin Elmer Wallac*). Staff remarked that the new device has better operating characteristics.

For Ra analysis the lab uses *Perkin Elmer HiSafe*® *3* as liquid scintillation cocktail (9 ml sample, 11 ml cocktail); measurement is after 4 weeks (to reach equilibrium). Quench corrections are applied. Calibration is done using certified standards.

For alpha spectrometry, a *Canberra Alpha Analyst* device with 12 chambers is used (6 devoted to U, 4 to Th and 2 to Po-210). The laboratory uses U-232, Th-229 and Po-209 for spiking. The chambers house 450 mm<sup>2</sup> PIPS detectors. A low noise *Edwards* vacuum pump is used for building up the necessary vacuum.

For total alpha and total beta measurements as well as for the determination of Sr-90 in radiochemically prepared samples a gas proportional counter type *Berthold LB770 10 channel low level counter* with a PC interface *Berthold low radioactivity data system LB 530 PC* is used. Calibration is done with Pu-239 and Am-241 for alpha measurements and with Sr-90 for beta measurements. The laboratory uses argon methane as counting gas, supplied by *Alphagas* (subsidiary of *Air Liquide*).

## <u>ICP-MS</u>

For the analysis of stable heavy metal isotopes, uranium and thorium in water (after chemical treatment) and in biological samples an *Agilent 7500 Series* single quadrupole ICP-MS device is available, allowing a detection limit of some 100 ppt (parts per trillion). A sample changer for up to 50 samples is included. Main area is the determination of isotopic ratios for lead and mercury. An expansion of the range of analysis to include determination of Tc-99 is foreseen. For this device *CITIUS* has a service contract with *Agilent*, Barcelona. The team was told that the lab applied for a new device at the Ministry of Economy and Competitiveness.

## <u>Clean cell lab</u>

Sample preparation for ICP-MS is done in the "clean cell laboratory". This room also houses the cooled device that contains eight cells for electrolytically enriching tritium, a *Cecil CE4004* spectro photometer for potassium measurement in water (for the determination of the K-40 content) and various sample preparation devices (furnaces *J.P. Selecta*, centrifuge *Hettich Universal 32*).

#### Other rooms

The verification team was shown a series of rooms belonging to the *servicio de radioisótopos* such as a special storage room for acids and chemicals, a balance room (the balance available is not yet calibrated but will be for accreditation), and a locked room for radioactive sources. Secondary standards are stored in a cooled environment (5°C); samples are stored cooled for 1 month, then destroyed.

#### <u>Tracing</u>

The team performed a tracing of sample 028-2011 and found full agreement of the data at the various levels of data handling.

#### Data handling, reporting

For measuring results computer printouts are produced and electronically sent to the environmental data base at *CSN* (data upload with checks of data and auxiliary information by the installed system).

Gamma spectra are kept on the analysis PC. Each apparatus has its log book; all systems are password protected. As a consequence of the verification in 2009, every week backups of the spectra are performed on the server. All results are calculated in the system (e.g. using *Excel*) on a PC. The team received a demonstration of the calculation tool for alpha spectrometry.

#### <u>QM/QC</u>

The institute has now ISO 9001 and ISO 14001 certifications for its measurement work and standard operating procedures. Annually, it participates in inter-comparison exercises organised by *CSN*. It also takes part in international inter-comparisons, e.g. for U-238, U-234 and Ra-226 by the IAEA.

The verification team was shown the procedures for measuring uranium, thorium, polonium, Pb-210 and Ra-226. As general practise, a copy of the relevant procedure is used as a template for the measurement log; the individual sample data are added.

Certified radioactive sources are from PTB, NIST, NPL-UKAS and CIEMAT-CEM.

Verification does not give rise to recommendations.

# 6.2.2 Nuclear physics department

The nuclear physics department was not part of the present verification. However, for completeness, we include the description of the verification in 2009:

"The *Departamento fisica atomica molecular y nuclear*" of the Physics faculty of the University of Seville, through the research group "Applied Nuclear Physics group", participates in the national environmental radioactivity monitoring programme set up by *CSN*. It also does annual sampling in relation to the radioactive waste disposal site El Cabril and the *FUA* uranium processing plant. This work is performed on the basis of an internal quality control programme developed by *CSN* with a view to controlling another laboratory that performs measurements on a monthly sampling basis.

[In 2009] The verification team received detailed explanations of the work of the department by the head of the "Applied Nuclear Physics Group" of the University (who is also scientific advisor at *CITIUS*). Three persons of this group are permanently employed, one works on basis of a scholarship. If needed, students are assigned to specific tasks. They receive according training and are supplied with the necessary documentation of procedures.

## <u>Roof area</u>

On the roof of the Physics faculty building several devices are installed, belonging to the national radioactivity monitoring programme.

A high volume aerosol sampler type ASS 500 (serial number 4/00) has been purchased from CLOR, Warsaw, Poland, via Canberra, Spain. The air flow measuring device is type DPFA95, serial number 95021, from Physik Technik Innovation, Erlangen, Germany, giving data on air flow rate, flow since last start, integrated flow, and operating hours. The flow rate is checked every two years by the supplier. Weekly air flow is 40 000 to 90 000 m<sup>3</sup>. Similar devices are set up in Bilbao, Barcelona, Madrid and Cáceres. Above the filter holder a heater is installed to avoid clogging of the filter in winter. The heating device consists of a quartz tube heater, triggered during night time by an electronic clock. Filters are 44 cm x 44 cm; they are changed each Monday.

A low volume air sampler with *Millipore* paper filter and charcoal cartridge (weekly filter change) is operated as well. The paper filters are used for total alpha and total beta activity determinations in air and – three months composite samples – for Sr-90 analysis. The device is based on a pump from *Busch*, Chevenex, Jura, CH, and a gas counter from *Kronschroeder SA*, Barcelona. The flow rate is controlled every year.

[In 2009] The verification team noted also a precipitation collector with a sampling area of  $1 \text{ m}^2$ . Until 2000 it was part of the national system.

# <u>Radiochemistry lab</u>

[In 2009] The team was informed that laboratory staff prepare samples for gross alpha, gross beta analysis and for determination of Sr-90 and actinides. However, sample measurement is performed at *CITIUS*. About ten samples are prepared per month.

The team observed preparation of a sample for Sr-90 analysis. It was shown the corresponding protocol with all relevant data being introduced (check marks are used for control). Sample preparation sheets were shown for Sr-90 determination in air, mixed diet and milk. Checks are performed by the technician and the head of the research group, however these are not signed off on the sample preparation sheets.

#### Spectrometry lab

[In 2009] The verification team visited the spectrometry lab that houses both, the alpha and the gamma spectrometry systems.

For alpha spectrometry a *Canberra Alpha Analyst* device with eight chambers is available: four are reserved for uranium, two for thorium and two for polonium determinations. A *Canberra DSA 1000* device interfaces to a PC.

For gamma spectrometry three detectors are available:

- Detector 'A', the 'extra system', a *Canberra* HPGe of 30% relative efficiency, with L-shaped cryostat, a shield made of "very old" lead (lead from the inner part of an old palace roof having a low content in Pb-210), and a *Canberra Bicron* active shield;
- Detector 'B', a *Canberra* REGe (reverse electrode germanium) detector in a Pb and Cu shield, with *Canberra* NIM equipment;
- Detector 'C', a *Canberra* LEGe (low energy germanium) detector used for Pb-210 and Am-241 determinations.

For data acquisition and evaluation *Canberra Genie* is available. However, this system is usually not used for evaluations in the automatic mode; the interactive peak fit contained in *Genie* is preferred, in particular for research tasks with low sample numbers.

Two PCs are used for controlling the devices.

Air filters, after changing, are folded in cling film to approximately 10 cm x 10 cm and pressed at 2 tons. Then, "immediately" a first gamma spectrometric measurement is performed to guarantee detection of any elevated gamma activities. A second long term gamma spectrometric analysis is done after two to three days to allow to a certain extent the decay of radon daughter products.

The devices are calibrated when purchased using mixed radionuclide sources. Energy checks are done with the peaks in the samples ("self-calibration", in particular for the LEGe detector). Efficiency controls are performed once every two months using an IAEA reference sample. Calibration of the REGe detector is based on modelling using a Monte Carlo method, verified by measurements.

Background is measured once per month (minimum two days, usually during weekends or holidays); generally the last measurement is taken as being representative.

Fine tuning of the gamma spectrometry system is done using an oscilloscope.

Several UPS's with holding times of approximately ½ hour are in operation. Electric power generation by a diesel generator is available at the faculty, but not available for this lab.

Liquid nitrogen for cooling the detectors is filled into the dewars each Monday or Tuesday. A large  $LN_2$  storage tank is installed at the faculty premises.

## <u>QM/QC</u>

[In 2009] The verification team was shown the quality manual for the Nuclear Physics department and for CITIUS (defining among others the responsibilities) with regard to the tasks for the national monitoring systems: Manual de la calidad de laboratorio de radiactividad ambiental (Programa REVIRA REM y PVRAIN) - Departamento de Física Aplicada. The team was also shown various written procedures for sample preparation."

#### 6.3 GRANADA UNIVERSITY LABORATORY

The Inorganic Chemistry department of the Granada University has a contract with *CSN* to work on radiochemistry and environmental radioactivity (*REM* programme), including radioactivity monitoring in the Cordoba region.

#### Radiochemistry laboratory

The radiochemistry laboratory, part of the Inorganic Chemistry Department and the Radioactive Facility of the Faculty of Science of the University of Granada is involved in the *CSN*-coordinated national environmental monitoring programme and in a special programme on monitoring around the El Cabril low/medium activity waste disposal facility. The laboratory participates in inter-comparison exercises organised by *CSN* and the IAEA. In addition the laboratory has carried out a project supported by the Spanish Agency of International Cooperation for Development (*AECID*), during the years 2007, 2008, 2011 and 2012 to establish environmental radiological monitoring in Morocco. On the other hand, the

laboratory carries out research on radiochemistry and in several areas related to environmental radiation monitoring.

Laboratory staff carry out the sample collection. Typical samples analysed in the laboratory are potable water (tap), food samples (from the university canteen) soil (30x30 cm top soil from Quéntar National Park 15 km from the laboratory) and atmospheric aerosols. For sample preparation water samples are evaporated from 10 to 1 litre, food samples are ashed. Generally, all the samples analysed are prepared according to *CSN* procedures.

The laboratory has a low-background proportional counter *Berthold LB770* for alpha measurements and a *Canberra* low-background HPGe gamma spectrometry system (20% relative efficiency). The gamma spectrometry system is calibrated for Marinelli (water, 0.2 litres and 1 litre capacity), a hermetic vessel (water, 60 millilitres capacity) and air filter geometries using standards provided by *CIEMAT*. Combined Monday-Friday mixed diet samples are prepared by ashing.

Monitoring results are reported to *CSN* on a regular basis. If radioiodine is detected, the laboratory has to inform *CSN* immediately. According to the *CSN* instruction the non-water samples are stored for 2 years, before being destroyed. Sample storage is located in the counting room.

The gamma spectrometry system is calibrated for energy and efficiency, but there is no procedure for controlling the system resolution (FWHM).

The laboratory quality manual and individual work instructions were presented to the verification team. The laboratory does not perform commercial analysis and is not accredited.

The laboratory facility is adequate, but cramped and slightly messy. Laboratory staff consist of only 2 persons.

#### Air sampling system

The medium flow rate air sampler is placed on the roof of the University building. It is part of the *CSN* controlled national monitoring network consisting of 20 stations. The sampler has both particulate and iodine filter (activated charcoal). A flow counter is used to determine the total sampling volume; typically the sampler operates on a flow rate of 75-100 litres/min. (There are seasonal variations in the flow due to dust clogging of the filter.) The filter is changed every Wednesday for total alpha/beta measurement. In addition, a monthly gamma analysis is carried out on a composite sample of one month's filters. For Sr-90 analysis a composite sample of three months' filters is used.

Power back-up for the sampler is provided by the building diesel generator; there is no battery back-up.

Filters are analysed by the university radiochemistry laboratory. According to Spanish practise there is no centralised analysis laboratory. This approach may create slightly varying laboratory practises affecting comparability of the results, but on the other hand it maintains local knowledge and education in environmental radioactivity.

The verification team was presented with the measurement procedure document in place for the air sampler.

The verification team recommends that, as a matter of good laboratory practise, the laboratory includes regular control of HPGe system resolution (FWHM of the Co-60 peak at 1.332 MeV) in the calibration control procedures. In addition, in order to reduce background, the verification team suggests removing the sample storage cabinet from the counting room. In the long term the verification team recommends staff rotation arrangements and providing more working space for the laboratory.

## 6.4 SALAMANCA UNIVERSITY LABORATORY

The verification team visited the radiological laboratory of the Salamanca University which is involved in the *CSN*-coordinated national environmental monitoring programme (*REM*) and in the independent *CSN* monitoring programme (*PVRAIN*) around the '*Quercus*' mine and the Juzbado nuclear fuel fabrication sites. In addition, the laboratory carries out research on radiochemistry and in several areas related to environmental radiation monitoring. The laboratory does not perform commercial analyses and is not accredited.

The *REM* programme includes drinking water, air particulates, soil (alpha/beta, gamma and Sr-90 measurements) and mixed diet (gamma).

The *PVRAIN* programme includes alpha, gamma, thorium, radon and uranium measurements of soil and biota samples.

Laboratory staff carry out the sample collection themselves. A sampling log sheet was presented to the verification team.

Samples are prepared in a sample preparation room housing scales, ovens for drying and furnaces for ashing samples.

The laboratory counting room houses two HPGe low-energy gamma spectroscopy systems equipped with background-reducing nitrogen ventilation. Typically counting times are several days. Every five years the gamma spectroscopy systems are calibrated for each sample geometry managed in the laboratory using commercial standards. In addition there is a regular control programme for HPGe-detectors (efficiency and energy). Long background measurements (15 days) are carried out twice a year. System resolution (FWHM) is not controlled on a regular basis.

Measurement data are stored on a central server and on paper. Results are reported to *CSN* twice a year for the national programme and once for the '*Quercus*' and the Juzbado programmes. Abnormally high values are reported immediately. Samples are stored at the laboratory for five years.

The laboratory participates in inter-comparison exercises organised by *CSN* and the IAEA. It has a staff of five, with possibility to have additional staff from other university departments.

The laboratory is well organised and has sufficient working facilities.

#### Air sampling system

The air sampler at the Salamanca University is located in a fenced area next to the chemistry laboratory building. The system consists of two individual particulate filters and one iodine filter. The total filter flow volume is calculated by multiplying the average flow rate with operation time. This method is rather inaccurate, since it does not take into account filter clogging by dust etc. The verification team was informed that a total flow meter has been ordered for the system.

The verification team recommends that, as a matter of good laboratory practise, the laboratory includes regular control of HPGe system resolution (FWHM of the Co-60 peak at 1.332 MeV) at the calibration control procedures. The verification team supports the work towards improving the accuracy of air sampler total flow volume determination.

## 6.5 ENUSA SAELICES LABORATORY

The *ENUSA* Saelices laboratory is responsible for the environmental monitoring of the '*Quercus*' site and the surrounding area. The laboratory is not accredited, but it has a quality system based on ISO 17025 and it participates in inter-comparison exercises organised by *CSN*. In addition to environmental radioactivity the monitoring includes also other monitoring areas (metals concentration, bacteria etc.).

Site staff collect the samples themselves. The number of environmental samples is some 1500 per year. There are in total about 35 persons working on the site (*ENUSA* staff 23 persons); the laboratory has a staff of two. The laboratory carries out analysis of soil, sediments and water. Biological samples (plants,

fish and foodstuffs) are prepared at the *ENUSA* Juzbado laboratory. Air radon measurements have been outsourced to the University of Cantabria.

The laboratory manages also the site TLD programme. Some 30 TLDs are placed on site and in the surrounding areas for long-term radiological monitoring. TLDs are collected quarterly and measured at the Juzbado laboratory.

Uranium analysis of high concentration samples (>30 ppm U) is carried out using a *Perkin Elmer Lambda 25* UV/VIS spectrometer. Low concentration uranium samples (typically environmental, around 2 ppb) are analysed with a *JASCO FP-8300* spectrofluorimeter.

The laboratory chemistry room is equipped to carry our radiochemical separations. A *Berthold LB* 770-1 and a *Berthold LB*-770PC low level counters are used for alpha and beta counting.

Laboratory measurement devices are calibrated annually by an external calibration service. Calibration certificates were shown to the verification team.

Samples are stored for two years in a separate sample storage room. Data are stored on a central server with back-up arrangements. *CSN* receives a regular monitoring report and the measurement data for compiling the national environment monitoring report of the old uranium mining sites. *ENUSA* uses the data also in its own annual environment report.

The laboratory is well organised and has plenty of working space.

Verification does not give rise to recommendations.

#### 6.6 ENUSA JUZBADO LABORATORY

The *ENUSA* Juzbado nuclear fuel fabrication plant produces fuel assemblies for all Spanish NPPs and several other NPPs in Europe. Part of the factory is an environmental laboratory, which analyses water, soil, biota and sediment samples. It has a staff of 2 analysts and 2 technicians. The water, soil and biota analyses of the laboratory are accredited to ISO 17025 by *ENAC*.

The environment laboratory is located in a separate building outside the main plant. Samples come to the laboratory with a sampling sheet containing information on sample type, sampling location and date. Each sample is numbered and recorded in the laboratory data system. During analysis a sample log is kept on PC and paper. The sample preparation room has furnaces for ashing, ovens for drying samples and a freezer for foodstuff samples. After analysis the samples are stored for one year in a separate sample storage room.

The radiochemistry room of the laboratory houses equipment for radiochemical separations. The counting room has two gamma spectroscopy systems (*Canberra* HPGe 20%) and an alpha spectroscopy system (*Canberra*). Typical counting times range from 24 hours to three days, which obviously limits the laboratory throughput.

The calibration system for gamma spectroscopy includes calibration for energy, efficiency and peak width (resolution). Calibration standards are provided by *CIEMAT*; activity certificates were presented to the verification team.

Measurement data are kept on the laboratory central server with automatic back-up. Data are copied to DVDs on regular intervals. The laboratory UPS system provides power autonomy for one hour.

The laboratory participates in inter-comparison exercises organised by the IAEA and CSN.

The laboratory is well organised and has sufficient working facilities.

Verification does not give rise to recommendations.

# 7 CONCLUSIONS

As part of a specific task within the programme of verifications according to Article 35 of the Euratom Treaty, a team of four inspectors of DG ENER D.3 visited former uranium production related sites (mines and mills) in Andalusia and in Castile and León. The goal of this verification was to obtain complete information and to verify a number of monitoring installations as part of the environmental radioactivity monitoring of the sites. The laboratories performing the measurements were also included in this verification. The verification did not touch any non-radiological aspects of the sites.

All verifications that had been planned by the verification team were completed successfully. In this regard, the information supplied in advance of the visit, as well as the additional documentation received before the start and during the verification, was useful.

The information provided and the outcome of the verification activities led to the following observations:

- (1) The information provided and the verification activities that were performed demonstrated that the radiological studies and surveillance programmes established for the concerned areas are appropriate and efficient.
- (2) With regard to some former uranium mines that still are not yet completely decommissioned (and that were not part of the verification) the team recommends urgently finding administrative solutions and starting an appropriate remediation and monitoring programme.
- (3) Concerning any uranium production re-use of sites in the future the team hopes that decisions can soon be found in order to achieve permanent solutions for remediation and surveillance.
- (4) A few topical suggestions are formulated. These aim at improving some aspects of the environmental radiological surveillance of the sites. They do not discredit the fact that environmental radioactivity monitoring of the uranium sites visited is in conformity with the provisions laid down under Article 35 of the Euratom Treaty.
- (5) The Commission Services ask the Spanish competent authority to inform them of any implementation achievements with regard to the situation at the time of the verification.
- (6) The verification team acknowledges the excellent co-operation it received from all persons involved in the activities it performed.

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## **APPENDIX 1**

Verification p	programme
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	Team 1	Team 2
Mon, 24 Sep	Opening Meeting in Granada	
	Verification of restored milling plant Andújar and CSN station. Verification of mine La Virgen	Verification of Granada University laboratory
Tue, 25 Sep	Verification of University Sevilla laboratory	Verification of Salamanca University laboratory
Wed, 26 Sep	Meeting at Lumbrales Verification of Valemascaño mine Verification of Quercus site (Saelices el Chico)	Verification of ENUSA Saelices (Quercus) laboratory Verification of CSN station Saelices el Chico
Thu, 27 Sep	Verification of Quercus site (cont.)	Verification of ENUSA Juzbado laboratory
Fri, 28 Sep	Closing Meeting (CSN, Madrid)	

Team 1: C. Gitzinger, E. Henrich

Team-2: V. Tanner, E. Hrnecek

## **APPENDIX 2**

<b>References and documentation</b>		
Documentation:		
CSN	Facilities for monitoring radioactivity related to former uranium mining and milling operations / Parts of the national monitoring system for radioactivity in the surroundings of the mentioned sites [Verificacion Art35.Mills-mines.CSN.ppt]	
CSN	Sample types, sample numbers and frequency of sampling for FUA [PVRAIN FUA.pdf]	
ENUSA	Restauración de la mina Casillas de Flores (Salamanca) – programa de vigilancia y mantenimiento [calendario 2010 casillas.pdf]	
ENUSA	Restauración de la mina Valdemascaño (Salamanca) – programa de vigilancia y mantenimiento [calendario 2010 valdemascaño.pdf]	
ENUSA	FORMER URANIUM ACTIVITIES IN CIUDAD RODRIGO AREA [PRESENTACION EURATOM GRANADA (24-09-12).pdf]	
ENUSA	Fábricas de concentrados de uranio – planta Quercus y planta Elefante y explotaciones mineras de Saelices el Chico (Salamanca) – Propuesta del programa de vigiláncia radiológica ambiental – Año 2012 [PVRA QUERCUS.pdf]	
ENRESA	Programa de vigilancia y mantenimiento / Programa de control de calidad del programa de vigilancia y mantenimiento / de la FUA [PVRA FUA.pdf]	
ENRESA	PRESENTATION TO VERIFICATION OF ENVIRONMENTAL RADIOACTIVITY ARTICLE 35 DECOMMISSIONING ANDUJAR URANIUM MILL FACILITIES [Presentación FUA-ART-35-Rev 0.pdf]	
ENRESA	PRESENTATION TO VERIFICATION OF ENVIRONMENTAL RADIOACTIVITY ARTICLE 35 ENVIRONMENTAL URANIUM MINES RESTAURATION IN SPAIN. LA VIRGEN URANIUM MINE [Presentación LA VIRGEN-ART-35-Rev 0.pdf]	
ENRESA	FÁBRICA DE URANIO DE ANDUJAR / RESTAURACIÓN DE ANTIGUAS MINAS DE URANIO ABANDONADAS EXPLOTADAS POR LA JUNTA DE ENERGÍA NUCLEAR [RGM Verificación euratom art 35 FUA.pdf]	
JUNTA DE ANDALUCIA; CONSEJERIA DE ECONOMIA, INNOVACIÓN, CIENCIA Y EMPLEO	MINING ACTIVITY (radioactive minerals) [URANIUM MINES.ppt]	

# Web sites consulted:

CSN	http://www.csn.es/
CIEMAT	http://www.ciemat.es/
ENUSA	http://www.enusa.es/
ENRESA	http://www.enresa.es/
Berkeley Resources Ltd.	http://www.berkeleyresources.com.au/projects-overview