Ref. Ares(2022)539186 - 24/01/2022

EUROPEAN COMMISSION
DIRECTORATE-GENERAL FOR ENERGY

DIRECTORATE D – Nuclear energy, safety and ITER
D.3 – Radiation protection and nuclear safety

Verification under the terms of Article 35 of the Euratom Treaty

Technical Report

SPAIN

Galicia and Cantabrian coastal marine environment

18 October 2021

Reference: ES 21-01

VERIFICATIONS UNDER THE TERMS OF ARTICLE 35

OF THE EURATOM TREATY

FACILITIES	Radioactivity monitoring of the Galicia and Cantabrian coastal marine environment in Spain
LOCATIONS	Madrid, Spain
DATES	18 October 2021
REFERENCE	ES 21-01
TEAM MEMBERS	Mr V. Tanner
REPORT DATE	14 December 2021
SIGNATURES	

V. Tanner

TABLE OF CONTENTS

1 INTRODUCTION 7

2 PREPARATION AND CONDUCT OF THE VERIFICATION 7

2.1 PREAMBLE 7

2.2 DOCUMENTS 7

2.3 PROGRAMME OF THE VISIT 7

3 BODIES HAVING COMPETENCE IN MONITORING RADIOACTIVITY IN THE MARINE ENVIRONMENT 9

3.1 MINISTRY FOR THE ECOLOGICAL TRANSITION AND DEMOGRAPHIC CHALLENGE 9

3.2 NUCLEAR SAFETY COUNCIL 9

3.3 CENTER FOR RESEARCH AND EXPERIMENTATION OF CIVIL WORK 9

3.4 CENTRE FOR ENERGY, ENVIRONMENT AND TECHNOLOGICAL RESEARCH 10

4 RADIOACTIVITY MONITORING OF THE GALICIA AND CANTABRIAN COASTAL MARINE ENVIRONMENT 11

4.1 GENERAL 11

4.2 MONITORING PROGRAMMES 11

4.3 MONITORING OF RADIOACTIVITY IN SEAWATER 12

4.4 MONITORING OF RADIOACTIVITY IN SEDIMENTS 13

4.5 MONITORING OF RADIOACTIVITY IN FISH AND AQUATIC BIOTA 13

4.6 INFORMATION TO THE GENERAL PUBLIC 14

4.7 INTERNATIONAL COOPERATION ON MARINE MONITORING 14

5 LABORATORIES 15

5.1 CENTER FOR RESEARCH AND EXPERIMENTATION OF CIVIL WORK 15

5.2 CENTRE FOR ENERGY, ENVIRONMENT AND TECHNOLOGICAL RESEARCH 18

6 VERIFICATIONS 23

6.1 INTRODUCTION 23

6.2 MONITORING PROGRAMME 23

6.3 CEDEX LABORATORY 23

6.3.1 General 23

6.3.2 Sample reception 24

6.3.3 Sample preparation 24

6.3.4 Counting rooms 25

6.3.5 Reporting 25

7 CONCLUSIONS 26

Abbreviations

CEDEX	Center for Research and Experimentation of Civil Work
CIEMAT	Centre for Energy, Environment and Technological Research
CSN	Nuclear Safety Council
EC	European Commission
ENAC	Entidad Nacional de Acreditación (Spanish national standardization agency)
FWHM	Full Width at Half Maximum
HPGe	High-purity Germanium
IAEA	International Atomic Energy Agency
LIMS	Laboratory Information Management System
MDA	Minimum Detectable Activity
MITERD	Ministry for the Ecological Transition and Demographic Challenge
OSPAR	Convention for the Protection of the Marine Environment of the Northeast Atlantic
REM	EC Radioactivity Environment Monitoring database
SASEMAR	Spanish Maritime Safety and Security Society

Annexes

Annex 1	Verification programme
Annex 2	Marine sampling location SMA-01
Annex 3	Marine sampling location MAS-03
Annex 4	Marine sampling location MAS-04

TECHNICAL REPORT

1 INTRODUCTION

Under Article 35 of the Euratom Treaty, all Member States must establish the facilities necessary to carry out continuous monitoring of the levels of radioactivity in air, water and soil and to ensure compliance with basic safety standards[1]. Article 35 also gives the European Commission the right of access to such facilities to verify their operation and efficiency. The radiation protection and nuclear safety unit of the European Commission's Directorate-General for Energy is responsible for undertaking these verifications. The Joint Research Centre Directorate-General provides technical support during the verification visits and in drawing up the reports.

The main purpose of the verifications under Article 35 of the Euratom Treaty is to provide an independent assessment of the efficiency and adequacy of monitoring facilities for:

- liquid and airborne discharges of radioactivity from a site into the environment;
- levels of environmental radioactivity at the site perimeter and in the marine, terrestrial and aquatic environment around the site, for all relevant pathways;
- levels of environmental radioactivity on the territory of the Member State.

Taking into account previous bilateral protocols, a Commission Communication[2] describing practical arrangements for Article 35 verification visits in Member States was published in the *Official Journal of the European Union* on 4 July 2006.

2 PREPARATION AND CONDUCT OF THE VERIFICATION

2.1 PREAMBLE

The Commission notified Spain of its decision to conduct an Article 35 verification in a letter addressed to the Spain Permanent Representation to the European Union. The Spanish Nuclear Safety Council (CSN) was designated to lead the preparations for the visit.

2.2 DOCUMENTS

To assist the verification team in its work, the national authorities supplied an information package in advance[3]. Additional documentation was provided during and after the verification visit. The information provided was used as a source during drawing up the descriptive sections of the current report.

2.3 PROGRAMME OF THE VISIT

The Commission and the CSN discussed and agreed on a programme of verification activities in line with the Commission Communication of 4 July 2006. It was agreed to carry out the verifications on the structure of the monitoring programme and on the CEDEX laboratory, which is in charge of most of the

[1] Council Directive 2013/59/Euratom of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, and repealing Directives 89/618/Euratom, 90/641/Euratom, 96/29/Euratom, 97/43/Euratom and 2003/122/Euratom (OJ L 13, 17.1.2014)

[2] Commission Communication *Verification of environmental radioactivity monitoring facilities under the terms of Article 35 of the Euratom Treaty — Practical arrangements for the conduct of verification visits in Member States* (OJ C 155, 4.7.2006)

[3] Replies to the preliminary information questionnaire addressed to the national competent authority, received on 22 September 2021

analytical work related to marine samples. The facilities of the other involved laboratory (CIEMAT) have been verified during earlier Art. 35 verifications in Spain[4].

The verification team met the following representatives of the national authorities and other parties involved:

Nuclear Safety Council (CSN)

- Ms Inmaculada Simón Cirujano. Technical Coordinator of Radiological Protection of the Public and Environmental Radiological Monitoring
- Ms Carmen Rey del Castillo. Head of Environmental Radiological Monitoring Department
- Mr Pablo Martínez Vivas. Deputy Direction for Environmental Radiological Protection, Expert
- Mr José Antonio Trinidad Ruiz. Deputy Direction for Environmental Radiological Protection, Expert
- Ms Sofia Luque Haredia. Deputy Direction for Environmental Radiological Protection, Expert
- Mr Manuel Aparicio Peña. International Department

Center for Research and Experimentation of Civil Work (CEDEX)

- Mr Javier Cachón de Mesa. Director of the Center for Studies of Applied Techniques
- Mr Javier Rodriguez Arévalo. Head of the Isotopic Applications Area
- Mr Luis Pujol Teres. Scientific Technical Program Coordinator

Junta de Galicia

- Mr Dionisio Garcia Pomar. CSN inspector in the autonomous community of Galicia

[4] Verification ES 19-01 (Environmental radioactivity monitoring in Palomares and Foodstuffs radioactivity monitoring in Palomares) and verification ES 08-09 (Huelva sites: Phosphogypsum piles and CRI-9)

3 BODIES HAVING COMPETENCE IN MONITORING RADIOACTIVITY IN THE MARINE ENVIRONMENT

3.1 Ministry for the Ecological Transition and Demographic Challenge

With respect to the protection against the presence of radioactivity in the environment, the Ministry for the Ecological Transition and Demographic Challenge (MITERD) has the following regulatory functions:

- Implementation of the Government's policy in the fight against climate change, prevention of pollution, protection of natural heritage, biodiversity, forests, sea, water and energy for the transition to a greener productive and social model.
- Development of state legislation on water and coasts, climate change, protection of biodiversity, environment, mountains, meteorology and climatology.
- Management of the public hydraulic domain of intercommunity basins and the maritime-terrestrial public domain.

3.2 Nuclear Safety Council

The Consejo de Seguridad Nuclear (Nuclear Safety Council, CSN), established in 1980, is the Spanish organisation competent in nuclear safety and radiological protection. It is independent from the Government and reports to the Parliament of Spain. CSN is a collegiate 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 General Secretary, 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. With respect to the protection against the presence of radioactivity in the environment, CSN has the following regulatory functions:

- Propose the necessary regulations on radiological protection;
- Issue mandatory and binding reports to MITERD prior the approval of matters related to authorisations of facilities and activities;
- 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;
- Control and monitor radiological quality of the environment throughout the national territory, in compliance with the international obligations. CSN runs its own programmes of environmental radiological monitoring (both around nuclear installations and at national level) and supervises all environmental radiological protection activities conducted by nuclear installations and by facilities using radioactive substances.

CSN is also responsible for proposing regulations to the MITERD concerning radiological protection of workers and members of the public.

3.3 Center for Research and Experimentation of Civil Work

The Center for Research and Experimentation of Civil Work (CEDEX) is a public research institution, organically attached to the Ministry of Transport, Mobility and Urban Agenda and functionally to the Ministry and to the Ministry for Ecological Transition and Demographic Challenge. It is responsible for the determination of the radiological quality of the Spanish continental and coastal waters. The CEDEX lines of activity in matters of radiological quality of water include:

- Scientific-technical support to the regulation of environmental radiological protection of the aquatic environment
- Radiological analysis of continental and coastal waters
- Characterization of the reference radiological background in the aquatic environment
- Origin and dispersion of natural and artificial radionuclides in water
- Radon in groundwater
- Innovation and development of analytical capabilities and test methods
- Development of quality standards for radiological analysis of water
- Management of databases on environmental radiological surveillance

3.4 Centre for Energy, Environment and Technological Research

The Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Centre for Energy, Environment and Technological Research CIEMAT), former Junta de Energía Nuclear (JEN), which the Spanish Atomic Energy Commission founded in 1951, is a public research organization assigned to the Ministry of Science, Innovation and Universities under the General Secretariat for Scientific Policy Coordination. It is a public centre for research in the fields of energy, environment and technology. CIEMAT is responsible for the analysis of Cantabrian Sea biota samples taken triennially by the CSN in the framework of the OSPAR co-operation.

4 RADIOACTIVITY MONITORING OF THE GALICIA AND CANTABRIAN COASTAL MARINE ENVIRONMENT

4.1 GENERAL

There are no nuclear facilities discharging radioactive material on the north coast of Spain, or on the Spanish rivers flowing to the Cantabrian Sea. Possible sources of artificial radioactivity in the marine environment are for example radioactive discharges originating from other European countries and historical sources on the seabed, most notably the radioactive waste containers (steel and concrete drums), which were disposed of on the seabed on the so-called Atlantic Trench some 40-60 years ago. In addition, there is one sunken nuclear submarine (Soviet Union K-8, sank in 1970) also on the seabed near the Bay of Biscay. Technical condition of these objects is not known. The locations and approximate depths of the waste drums are known (Fig. 1); they are on international waters several hundred kilometres from the Spanish coast. There are no regular radioactivity monitoring programmes of the marine environment in the historical radioactive waste disposal areas.

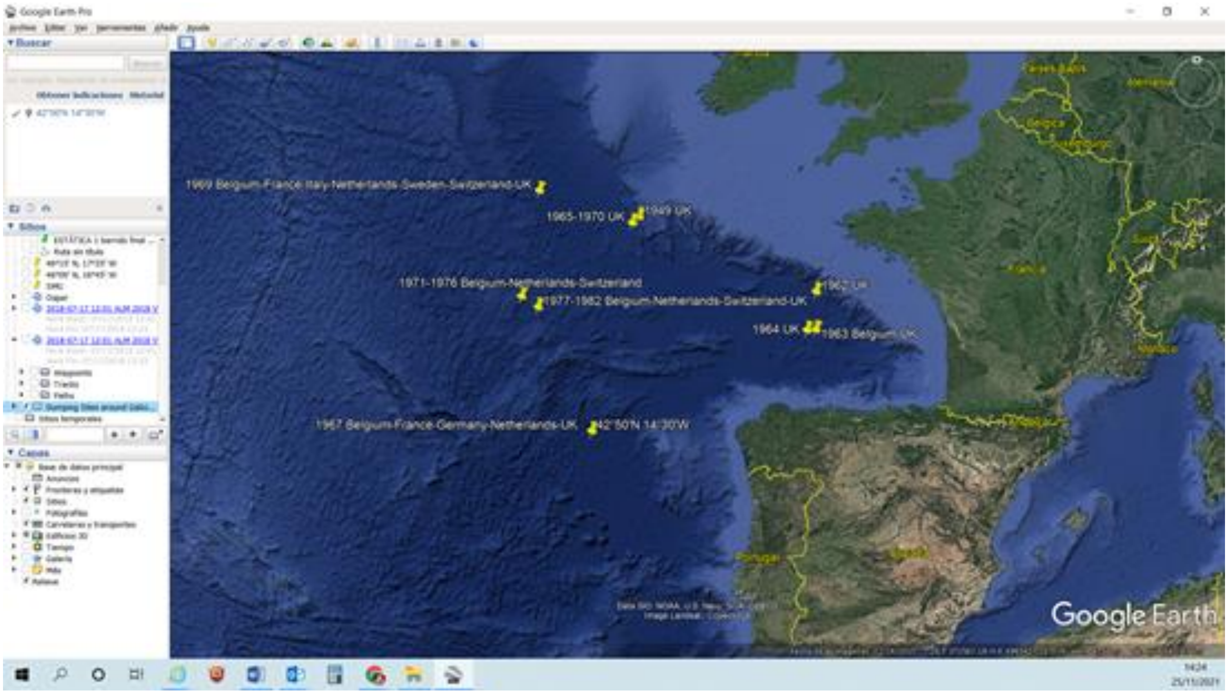


Figure 1. Locations of the dumping sites in the North-East Atlantic close to Galicia and Cantabrian coasts[5]

4.2 MONITORING PROGRAMMES

CSN carries out control and surveillance of the environmental radiological impact of nuclear and radioactive facilities and monitors the radiological quality of the environment throughout the Spanish national territory, in compliance with the obligations of the Spanish State in this matter. Radiological surveillance of the environment is carried out through a network made up of a surveillance network in the surroundings of nuclear facilities and a national network. The National Environmental Radiological Surveillance Network, not associated with facilities, (REVIRA) is distributed throughout the national territory and is managed by the CSN. REVIRA provides radiological information on radioactivity in the

[5] IAEA-TECDOC-588 "Inventory of radioactive material entering the marine environment: Sea disposal of radioactive waste" https://www-pub.iaea.org/MTCD/Publications/PDF/te_588_web.pdf

atmosphere, soil, water (drinking, continental and marine water) and food, with results available since 1993.

The sampling and radiological analysis programs follow the recommendations established by the EU in order to facilitate compliance by the Member States with Articles 35 and 36 of the Euratom Treaty. REVIRA includes a Sampling Stations Network (REM) and an Automatic Stations Network (REA). REA provides real time information about radioactivity in air, while REM is made up of two programmes:

- Earth and atmosphere programme, which samples and analyses air, soil, drinking water, milk, and mixed diet. This programme is divided in two networks: dense network, characterized by having numerous points throughout the entire Spanish territory; and sparse network, characterized by having fewer points but with a higher sensitivity, which enables to detect activity of the radionuclides of interest.
- Continental and coastal water programme. This programme is divided into the same two networks (dense and sparse), both carried out by CEDEX, under agreement with the CSN, made up of 102 points and 4 points, respectively, of the main rivers and coastal waters. The national aquatic environment surveillance network includes the rivers of the main hydrographic basins and the waters of the Spanish coastal perimeter.

In order to monitor the quality of the water of the Spanish coast from the radiological point of view, a network of stations was established in 1987, under agreement between CSN and CEDEX, and expanded in 1993 and 2004 with new stations and analyses. The stations were selected in such a way that due to their location and characteristics they could be representative of the Spanish coast (main capes, ports and beaches subject to marine currents or located in river mouths). Figure 2 presents the monitoring stations at the Cantabrian Sea from both programmes (earth and atmosphere as well as continental river and coastal waters).

Figure 2. Radioactivity monitoring locations on the Cantabrian Sea and coastal areas in the northern Spain

4.3 MONITORING OF RADIOACTIVITY IN SEAWATER

Radioactivity in seawater on the Galicia and Cantabrian coastal marine environment is monitored by taking samples from four sampling sites, shown on Table I and Fig. 3 below. Annexes 2-4 present the detailed maps of the sampling sites. Samples are taken on the surface on a quarterly basis at a distance of 10 miles from the coast, except in seaports, where the samples are taken at the mouth. SASEMAR (Spanish Maritime Safety and Security Society) collects the water samples (10 litres) manually and sends them to the CEDEX laboratory for analysis together with a filled-in sampling sheet. Sampling follows a protocol valid for all surface waters under surveillance in Spain. The frequency of the sampling is defined with the aim of obtaining the best representative information about a specific radiological situation.

In addition to the quarterly sampling, the CSN collects triennial seawater samples from the Ajo Cape as a part of the OSPAR co-operation. For these samples, the CSN uses four containers of 5 L with a sealed lid to avoid losing sample. Samples are taken from the seashore, avoiding taking sand as much as possible. All recipients are introduced in portable fridges with cold accumulators to ensure sample conservation. The following analysis are carried out (see sections 5.1 and 5.2):

- Seawater sampled by CEDEX: Gross alpha activity, gross beta activity, residual beta activity, tritium activity, gamma spectrometry (artificial radionuclides[6]).
- Seawater sampled by CSN (analysis by CIEMAT): Gamma emitters[7] and radiochemical analysis of Pb-210, Po-210, Ra-224, Ra-226, Pu-238 and Pu-239,240.

Table I. Seawater sampling programme

Code	Sampling Location	Type	Samples/ year	Frequency	Coordinates	
					LONGITUDE	LATITUDE
SMA01	AJO CAPE – CANTABRIAN SEA	Seawater	4	Quarterly	03°34’00”W	43°32’07”N
SMA01*	AJO CAPE – CANTABRIAN SEA	Seawater	1	Triennial	03°34’00”W	43°32’07”N
MAS03	ORTEGAL CAPE – ATLANTIC	Seawater	4	Quarterly	07°4800"W	43°52'00"N
MAS04	VILLANO CAPE – ATLANTIC	Seawater	4	Quarterly	09°22'00"W	43°00'00"N
MAS05	SILLEIRO CAPE – ATLANTIC	Seawater	4	Quarterly	08°54'00"W	42°15'00"N

*Triennial sample taken by CSN.

Figure 3. Seawater sampling locations at the Cantabrian Sea and Galicia

4.4 MONITORING OF RADIOACTIVITY IN SEDIMENTS

The current marine monitoring programme does not include monitoring of radioactivity in sediments. This type of samples are not required in Euratom Article 36 recommendations, nor in the OSPAR cooperation monitoring programme.

4.5 MONITORING OF RADIOACTIVITY IN FISH AND AQUATIC BIOTA

Within the framework of the OSPAR Convention (Convention for the Protection of the Marine Environment of the Northeast Atlantic), every three years samples of seaweed, molluscs, fish and

[6] Cr-51, Mn-54, Co- 58, Fe-59, Co-60, Zn-65, Nb-95, Zr-95, Ru-103, Ru-106, Cs-134, Cs-137, Ba-140, La-140 and Ce-144

[7] Be-7, K-40, Cr-51, Mn-54, Co-58, Fe-59, Co-60, Zn-65, Nb-95, Zr-95, Ru-103, Ru-106, Cs-134, Cs-137, La-140, Ba-140, Ce-144, Tl-208, Pb-210, Pb-212, Bi-214, Pb-214, Ra-228 and Am-241

seawater are collected by the CSN at the sampling point of Cape Ajo (Table II), belonging to the Cantabrian Sea.

Table II. Fish and aquatic biota sampling programme

Code	Sampling Location	Type	Samples/ year	Frequency	Coordinates	
					LONGITUDE	LATITUDE
SMA01	AJO CAPE – CANTABRIAN SEA	Seaweed Fish Molluscs	1	Triennial	03°34’00”W	43°32’07”N

CSN uses the following biota sampling procedures:

- Seaweed: 20-30 L resistant bags. Sample is taken from the seashore and cleaned with seawater, avoiding taking sand as much as possible.
- Fish and Molluscs: Bought in fish market, making sure that the origin is the Cantabrian Sea (at least 3.5 kg).

All samples are transported to CIEMAT in portable fridges with cold accumulators to ensure sample conservation. Gamma emitters[8] are analysed by gamma spectroscopy. Radiochemical analysis of Pb-210, Po-210, Ra-224, Ra-226, Pu-238 and Pu-239,240 is also carried out (see section 5.2).

Additionally, within the dense and sparse earth and atmosphere network, samples of mixed diet are measured in a quarterly basis in the following sampling points of the north of Spain: Bilbao, Santander, Oviedo and La Coruña (see 4.2). These samples are made up of weekly complete diet samples that include marine food.

4.6 INFORMATION TO THE GENERAL PUBLIC

In compliance with the functions entrusted to the CSN relating to public information, CSN has developed a computer application to give public access to environmental radiological surveillance data in Spain, which can be accessed through the CSN website. In the case of data corresponding to the Sampling Stations Network (REM), the surveillance data can be consulted since 2017 through the CSN website[9] with data since 2006. This website is updated every year after the CSN receives the data from the different entities (laboratories, installations, regional authorities…), usually in the first quarter of the following year.

CSN also elaborates annually a report about the environmental radioactivity monitoring programmes, where all information is summarized and analysed, including charts with time evolution. These reports are available for public access through the CSN website (chapter “04. Informes técnicos”)[10].

The CSN reports annually to Spanish Congress and Senate a summary of the results obtained in these networks and publishes this report in its website[11].

4.7 INTERNATIONAL COOPERATION ON MARINE MONITORING

OSPAR (Convention for the Protection of the Marine Environment of the Northeast Atlantic) is one of the most important international bodies related to the marine environment monitoring in the North-East Atlantic Ocean. CSN takes part in the work of OSPAR in the field of marine radioactivity. Under

---

8 Be-7, K-40, Cr-51, Mn-54, Co-58, Fe-59, Co-60, Zn-65, Nb-95, Zr-95, Ru-103, Ru-106, Cs-134, Cs-137, La-140, Ba-140, Ce-144, Tl-208, Pb-210, Pb-212, Bi-214, Pb-214, Ra-228 and Am-241

9 https://www.csn.es/valores-radiologicos-ambientales-pvra-rem/

10 https://www.csn.es/centro-de-documentacion

11 https://www.csn.es/relaciones-csn-con-las-cortes/informe-anual

Annex IV to the OSPAR Convention, OSPAR is required to produce periodic assessments of the quality status of the maritime area covered by the Convention. Quality Status Reports (QSR) are produced approximately every 10 years, with Intermediate Assessments being produced in between. The next QSR is planned to be issued in 2023 and will assess the period 2010 – 2020. Previous Quality Status Reports were carried out in 2010 (QSR 2010) and 2000 (QSR 2000).

To achieve the OSPAR strategic objective, the OSPAR Radioactive Substances Committee (RSC) carries out periodical evaluations of progress made in implementing the strategy. Four periodic assessments of progress towards the objective of the OSPAR Radioactive Substances Strategy have been published. The Fourth Periodic Evaluation (2016) focused on progress made with regard to radioactive discharges from nuclear and non-nuclear sectors. This is the latest Periodic Evaluation published. A specific workgroup of the RSC is working on the Fifth Periodic Evaluation, which should be ready in 2022. Spain is part of this working group.

5 LABORATORIES

5.1 Center for Research and Experimentation of Civil Work

Center for Research and Experimentation of Civil Work (CEDEX) is a public research institution located in Madrid, functionally dependent from the Ministry of Transport, Mobility and Urban Agenda. It is responsible for the determination of the radiological quality of the Spanish continental and coastal waters.

The Maritime Safety and Security Society (SASEMAR) collects water samples (10 litres) manually and sends them to the CEDEX laboratory for analysis together with a filled-in sampling sheet. No processes are subcontracted to third parties. Tables III and IV present the available counting devices and typical counting times for each analysis.

Table III. CEDEX devices and counting times for each analysis

Radionuclide	Measurement device	Counting time (s)
Gross alpha	ZnS(Ag) scintillation detector	86 400
Gross beta	Proportional counter	18 000
Residual beta or gross beta activity excluding potassium	Atomic emission spectrometry	18 000
H-3	Liquid scintillation counter (Quantulus 1220)	18 000
Gamma emitters (especially Cs-137, MDA < 0.045 Bq/L)	Gamma spectrometry	180 000

Table IV. CEDEX laboratory device characteristics

Device	Manufacturer	Type or Model	Number	Calibration, Maintenance & Performance Procedures
HPGe detectors	Canberra	GR2522, GR2520, GX4020 GC13022	4	ITE-1011 ITM-1302
Proportional counters	Berthold	LB 770-2	1 device (10 detectors)	ITC-1301

Liquid Scintillation counters	PerkinElmer	Quantulus 1220	2 detectors	ITV-1302 ITV-1303
ZnS(Ag) scintillator detector	Canberra	Modular PMT: 2007P HV: NHQ212M NIM 2000 Canberra	8	ITC-1306

Results are recorded in the laboratory information management system named "Meragua". All electronic files generated in the measurements of the samples are archived in a computer server. According to the CSN requirements, if an activity concentration is below detection limit or uncertainty, it is reported as below detection limit (with its value).

The laboratory keeps the samples for at least 1 year and the reports in electronic format for at least 5 years after analysis and reporting.

The CEDEX laboratory is accredited by ENAC (UNE-EN ISO/IEC 17025) (Accreditation number ENAC 82/LE1955). It regularly participates in international and national intercomparison exercises (Table V).

Table V. CEDEX radiological laboratory intercomparisons and proficiency tests

Assay	Year	Organizer	Matrix
INTERNATIONAL			
Gross alpha, gross beta and tritium, and gamma emitters	2016	IAEA-TEL-2016-03	Water and spruce needles
Tritium, Cs-134 and Cs-137	2016	IAEA-RML-2016-01	Sea water
Gross alpha, gross beta and tritium and gamma emitters	2017	IAEA-TEL-2017-03	Water, milk power and Ca-carbonate from spring water
Tritium, Co-60, Cs-134 and Cs-137	2017	IAEA-RML-2017-01	Sea water
Gross alpha, gross beta and gamma emitters	2018	IAEA-TEL-2018-03	Water and agricultural soil
Tritium, Co-60, Cs-134 and Cs-137	2018	IAEA-RML-2018-01	Sea water
Tritium enrichment by electrolysis	2018	IAEA, IHL (Isotope Hydrology Laboratory). TRIC2018	Water
Rn-222	2018	JRC (Joint Research Centre)	Water
Gross alpha, gross beta and gamma emitters	2019	IAEA-TEL-2019-03	Water and shrimp

Assay	Year	Organizer	Matrix
Tritium, Co-60, Cs-134 and Cs-137	2019	IAEA-RML-2019-01	Sea water
Gross alpha and gross beta	2019	JRC (Joint Research Centre)	Water
Gross alpha, gross beta and gamma emitters	2020	IAEA-TEL-2020-03	Water and fish
Gross alpha, gross beta, tritium and gamma emitters	2021 (ongoing)	IAEA-TEL-2021-03	Water and Japanese bamboo
NATIONAL			
Gamma emitters	2016	Consejo de Seguridad Nuclear (CSN)	Soil
Gamma emitters	2018	Consejo de Seguridad Nuclear (CSN)	Milk power
Rn-222	2019	LaRUC (UC) - Laboratory of environmental radioactivity (University of Cantabria)	Water
Gross alpha, gross beta, residual beta and tritium	2019	Consejo de Seguridad Nuclear (CSN)	Water and sea water
Rn-222	2021 (ongoing)	LaRUC (UC) - Laboratory of environmental radioactivity (University of Cantabria)	Water
I-131	2021 (ongoing)	LARUEX (UNEX) – Laboratory of environmental radioactivity (University of Extremadura)	Water

5.2 Centre for Energy, Environment and Technological Research

The Centre for Energy, Environment and Technological Research (CIEMAT) laboratory is located in Madrid. It analyses a large number of environmental and quality control samples from all Spanish territories. As regards the Cantabrian Sea marine monitoring, the Unit of Environmental Radiology and Radiation Monitoring (URAyVR) carries out the analysis of the Ajo Cape samples taken triennially by the CSN. The unit is accredited by ENAC (UNE EN ISO/IEC 17025; accreditation number ENAC 144/LE471). Tables VI and VII present the laboratory equipment. URAyVR retains the samples and reports in electronic format at least for 5 years after analysis and reporting. No processes are subcontracted to third parties.

Table VI. CIEMAT URAyVR measurement devices

DEVICE	MANUFACTURER	TYPE OR MODEL	NUMBER
HPGe detectors	Canberra	GR2920, GR3321, GR4022, GX3519, BE5030, GC2518, GC3251, BE5030, GX4020, X10022 and GCW3521	11 detectors
Proportional counters	Berthold	LB-770 B	3 devices (30 detectors)
Alpha/Beta Counting System	Canberra	LB4200-8	1 device (8 detectors)
Scintillation counters	Packard	Tricarb 2750 TR/SL, Tricarb 2770, Tricarb 3100 and Tricarb 3110 TR	4 detectors

Samples are seawater, seaweed, fish and molluscs. They are identified as soon as they are received at the laboratory. CSN supplies a sample delivery note containing the sample details (sampling point, client reference and date of sampling) and the characteristics of the sample (water or biota, specifying whether it is food or an indicator organism). The information included in the delivery form is imported directly into the URAyVR database to avoid typing mistakes. At this point, an identification number is assigned to the sample, which allows identifying the sample and its aliquots throughout the process. This reference together with the client's reference allow the samples to be traced unequivocally.

The following describes the sample preparation procedures of each analysis:

Gamma spectrometry: Direct measurement of seawater is carried out by introducing the sample (4 L) into a Marinelli geometry. For samples other than seawater, gamma spectrometry is carried out on the ashes obtained after calcination at 450°C. Subsequently, the ashes are calcinated at 650°C for the rest of the determinations.

Strontium: Radiochemical preparation for Sr-90 analysis is performed only in the hard part of clams. The method consists of determination of the activity concentration of Sr-90 in secular equilibrium with Y-90 by means of a continuous gas flow proportional counter. Before radiochemical separation, the ashes obtained by calcination are pre-treated and leached with mineral acids. This pre-treatment allows Sr-90 to be concentrated to reach the required detection limits. The radiochemical separation method is based on selective precipitations (chromates, hydroxides and carbonates) and the subsequent use of chromatographic extraction resins (Sr-Spec resin).

Plutonium: The radiochemical preparation for Pu-238 and Pu-239,240 analysis is performed on biota and seawater samples. In the case of biota samples, initial leaching with HNO_3 is carried out on the ashes obtained after calcination at 650°C. In the case of seawater samples, Plutonium precipitation is performed with NaOH. The tracer used in the radiochemical process is Pu-242. The residue or precipitate obtained is dissolved in 8N nitric acid. In this solution, radiochemical separation is carried out to remove the interferents (Am, Th and U), which consists of several oxidation-reduction reactions combined with two consecutive ion exchange chromatography processes. The Pu eluate is evaporated to dryness and then Pu is electrodeposited on a stainless steel disc. Finally, the disc is measured by alpha spectrometry on an implanted silicon detector (PIPS).

Polonium/Lead: The radiochemical method of separation of Po-210 and Pb-210 in the samples consists of extracting them from the source matrix, auto-depositing Po-210 on silver and measuring it by alpha spectrometry in an Implanted Silicon (PIPS) detector. Seawater samples are acidified with HCl and evaporated to dryness. Solid samples are then acid digested with HF/HNO_3 mineral acid mixtures; HCl/boric acid and concentrated HNO_3 or HCl. The tracer used is Po-208, which is added at the beginning of the analysis. Po-210 is then selectively self-deposited in reducing medium on a silver disc and subsequently measured to obtain the activity at the separation date. The remaining Po-210-free solution is stored for at least 3 months to allow the Po-210 generated by the decay of the Pb-210 present in the solution to grow. After this time, Po-208 tracer is added and a second Po-210 autodeposition is performed on a silver disc. The activity concentration of Po-210 in the second autodeposition is proportional to the amount of Pb-210 that has decayed in that time; they are related to each other through Bateman's equations of radioactive equilibrium.

Radium: Ra-226 radiochemical preparation is performed on seawater and biota samples. In the case of biota samples, an aliquot of the sample is digested in a microwave oven. The obtained aliquot is diluted to 1 L with deionised water. In the case of seawater, 1 L of sample is taken. The radiochemical method starts with the precipitation of the $[Ba\text{-}Ra\text{-}Pb]SO_4$ precipitate. Pb is removed by a solution of AEDT in a strongly ammoniacal medium, in which the main alpha emitters are also separated. The remaining stable interferents are removed by the use of sodium dichromate and the subsequent precipitation of Ba as chloride. Finally, Ba is isolated by precipitation as $[Ba\text{-}Ra]SO_4$. The precipitate is filtered and measured in a ZnS(Ag) solid scintillation detector. Determination of Ra-226 activity is carried out by means of an equation system derived from the Bateman equations.

The following counting methodologies are used:

Gamma spectrometry: CIEMAT gamma spectrometry laboratory has 10 Canberra HPGe detectors connected to an electronic chain consisting of a high voltage source, an amplifier and ADC's. The spectra are acquired and analysed using the Canberra Genie 2000 software. Samples are measured individually for a minimum time of 60 000 s. The radionuclides analysed are: Be-7, K-40, Cr-51, Mn-54, Co-58, Fe-59, Co-60, Zn-65, Nb-95, Zr-95, Ru-103, Ru-106, Cs-134, Cs-137, Ba-140, La-140, Ce-144, Tl-208, Pb-210, Pb-212, Bi-214, Pb-214, Ra-226, Ra-228 and Am-241.

Strontium: Up to 8 samples are measured simultaneously for 120 000 s in a proportional counter LB 4200 APEX-ALPHA/BETA CANBERRA.

Plutonium/Polonium/Lead: The laboratory is equipped with two Canberra Alpha Analyst units. Each unit contains 12 implanted silicon semiconductor detectors operating under vacuum. These detectors are used for alpha spectrometric measurement of Pu (Pu-238, Pu-239,240) and Po (Po-210 and Pb-210) isotopes. The measurement time for all analysis is 600 000 s.

Radium: The laboratory has 12 Canberra ZnS(Ag) solid scintillation detectors. The measurement time of the radium samples is 60 000 s.

Strontium: Samples are measured in a Canberra proportional counter, which allows simultaneous measurement of 8 samples (LB 4200 APEX-ALPHA/BETA CANBERRA).

Radium: Samples are measured in one of the 12 Canberra ZnS(Ag) solid scintillation detectors.

The methodologies for determining and measuring the analyses are accredited by ENAC based on the ISO 17025 standard. Quality controls of the methods and detectors used are carried out monthly. The quality controls and backgrounds/blanks are stored in the records of each laboratory and reviewed internally once a year and by the accreditation entity every 2 years.

The table below summarises the main characteristics of the detectors used to measure the samples.

Table VII. CIEMAT URAyVR device characteristics

Analysis	Number of devices	Device type	Calibration and maintenance procedure	Standard used	Calculation of results
Gamma spectrometry	10	HPGe detector	• Energy and efficiency calibration • Efficiency checks • Background measurements • Liquid nitrogen charge	Mixed gamma ray sources (provided by LMRI, CIEMAT)	Canberra Genie 2000 software
Radium	12	Solid scintillation detector (ZnS(Ag))	• Energy and efficiency calibration • Efficiency checks • Background measurements	Ra-226 and Am-241 sources (provided by LMRI, CIEMAT)	Microsoft EXCEL spreadsheet
Strontium	1 device (10 detectors)	Proportional counter	• Energy and efficiency calibration • Efficiency checks • Background measurements	Sr-90/Y-90 sources (provided by LRMI, CIEMAT)	Microsoft EXCEL spreadsheet
Plutonium	10	PIPS detectors	• Energy and efficiency calibration • Efficiency checks • Background measurements	Pu-242 sources (provided by National Physical Laboratory (NPL))	Microsoft EXCEL spreadsheet
Polonium/Lead	10	PIPS detectors	• Energy and efficiency calibration • Efficiency checks • Background measurements	Po-208 sources (provided by National Physical Laboratory (NPL))	Microsoft EXCEL spreadsheet

The following data handling procedures are applied:

> **Gamma spectrometry**: All parameters of the measurement, calibration and background used for the determination of the gamma emitters are recorded in the Genie 2000 CNF file. This file is considered the primary file and the one used to store the data. The final activities are extracted from the file and recorded into an Access database. Detection limits are determined according to the Currie criterion by the Genie 2000 software.
>
> **Strontium, polonium, lead, plutonium and radium**: Measurement data are stored in an Excel spreadsheet. Detection limits are calculated according to ISO 11929.

Calculations of the activity concentrations of the analysed radionuclides are performed using MS Excel spreadsheets. The final results are imported into the database. In the case of gamma spectrometry, data are imported directly into the database from the CNF file generated by Genie 2000. According to the CSN requirements, if an activity concentration is below detection limit or uncertainty, it is reported as below detection limit (with its value).

The URAyVR database stores the initial sample data and the final activity concentrations. In addition, the initial data are stored in an initial record made with MS Excel. This data is imported directly into the database. Reports are regularly submitted to the CSN when samples are analysed.

All reports submitted to the CSN are digitally signed and stored on a group hard disk that is only accessible by the persons responsible of the analysis. Back-up copies of this hard disk are made daily. The samples themselves are stored for 5 years in a warehouse protected from light (Storage time for seawater samples is 1 year.).

All laboratory processes are carried out in accordance with the UNE 17025 standard. The laboratory is accredited by ENAC (N°144/LE471). The laboratory participates annually in several intercomparison exercises and proficiency tests organised by the IAEA, CSN and MAPEP, in which most of the matrices analysed in the laboratory are verified (Table VIII). CIEMAT has a quality assurance department that performs internal audits, reviews the procedures and maintains the quality system.

Table VIII. CIEMAT URAyVR radiological intercomparisons and proficiency tests

Assay	Year	Organizer	Matrix
INTERNATIONAL			
Gamma Emitters (natural and anthropogenic), Tritium Strontium 90	2020	IAEA (IAEA-RML-2020-01)	Seawater
Gamma emitters (natural and anthropogenic) Gross alpha and beta activity, Strontium 90	2020	IAEA (IAEA-TEL-2020-04)	Water, fish ashes and filters
Gamma emitters (natural and anthropogenic) Gross alpha and beta activity, Strontium 90	2020	US. Department of Energy (MAPEP-43)	Soil, vegetable and filters
Gamma emitters (natural and anthropogenic) Gross alpha and beta activity, Strontium 90	2020	US. Department of Energy (MAPEP-42)	Soil, vegetable and filters
Gross beta activity	2020	European Commission, JRC	Water
Gamma emitters (natural and anthropogenic) Gross alpha and beta activity, Strontium 90	2019	US. Department of Energy (MAPEP-41)	Soil, vegetable and filters
Natural gamma emitters	2019	National Physical Laboratory (NPL)	Slag
Gamma emitters (natural and anthropogenic) Gross alpha and beta activity, Strontium 90	2019	US. Department of Energy (MAPEP-40)	Soil, vegetable and filters
Gamma emitters (natural and anthropogenic) Gross alpha and beta activity, Strontium 90	2019	IAEA (IAEA-TEL-2019-04)	Water, filters and shrimp
NATIONAL			
Gamma emitters (natural and anthropogenic) Gross alpha and beta activity, Strontium 89 and 90	2020	CSN	Soil
Gamma emitters (natural and anthropogenic), Tritium Gross alpha and beta activity, Strontium 89 and 90	2019	CSN	Water

6 VERIFICATIONS

6.1 INTRODUCTION

Verifications were carried out in accordance with the agreed programme (Annex 1). This chapter summarises the verifications carried out the by the verification team. The team has assessed the monitoring arrangements based on their own expertise and comparison with similar arrangements in other Member States.

The outcome of the verification is expressed as follows:

- A '*Recommendation*' is made when there is a clear need for improvement in implementing Art. 35. These are included in the main conclusions of the verification. The Commission requests a report on the implementation of the recommendations – lacking implementation of a recommendation can lead to a reverification.
- A '*Suggestion*' is made when the verification team identifies an action, which would further improve the quality of the monitoring.

In addition, the team may '*commend'* particularly good arrangements, which could serve as a best practice indicator for the other EU Member States.

6.2 MONITORING PROGRAMME

The monitoring programme for radioactivity in the coastal marine environment in Galicia and the Cantabrian Sea is conducted as a part of the national environmental radioactivity monitoring programme in Spain. It consist of sampling seawater and marine biota on regular intervals at representative locations along the north coast of Spain. In addition, the programme includes monitoring of radioactivity in mixed diet samples containing also marine food (seaweed, fish and molluscs). Marine sediments are not included in the programme, in accordance with the international practice in agreement with the OSPAR convention and the Euratom Article 36 recommendation.

Samples are analysed in accredited laboratories (CEDEX and CIEMAT). Monitoring results are made available to the public at the CSN website and in regular monitoring reports issued by the CSN. In addition, Spain provides marine radioactivity data to OSPAR and participates in the OSPAR Radioactive Substances Committee.

Verification team notes, that while sampling of seawater is carried out on annual basis, marine biota sampling is based on triennial sampling in accordance with agreement 2005-8 of the OSPAR framework.

> *Verification team suggests that the CSN consider an annual sampling programme for the most important marine foodstuffs produced in the area.*

6.3 CEDEX LABORATORY

6.3.1 General

Verification team visited the CEDEX laboratory in Madrid[12]. The laboratory employs altogether about 400 staff members, seven of whom work on the water radioactivity measurements. The laboratory measures water radioactivity of samples from 15 sampling stations on the Spanish territorial waters. Four of these stations are in the Galicia Coast and Cantabrian Sea. CEDEX laboratory is accredited according to ISO/IEC 17025.

[12] C. de Alfonso XII, 3, 28014 Madrid

The laboratory has very good facilities for sample preparation, analysis and storage. The room layout has been clearly arranged to support each task. No staff shortage was reported.

Verification team commends the clear and logical laboratory organisation and layout.

6.3.2 Sample reception

About 60 marine water samples (10 litres) are received annually (Fig. 4). Sample receipt record is kept on paper; the incoming sample data are also recorded in the laboratory database (LIMS).

No remarks.

Figure 4. CEDEX laboratory water sample receipt and storage area.

6.3.3 Sample preparation

Laboratory layout facilitates separate preparation areas for different measurements. The available sample preparation techniques include the following:

- Electrolytic enrichment for H-3 measurement (Fig. 5). The process consists of distillation (salt and K-40 removal) and electrolysis at a low temperature (2-3°C). The condensate is then measured in a liquid scintillation counter with LSC Cocktail Ultima Gold LTT. The enrichment process significantly lowers the H-3 detection limit (factor 10 -15).
- Oven for preparing samples for Potassium measurement using an Atomic Absorption Spectrometer.
- Evaporation equipment for preparing samples for Gross alpha/beta measurement.
- Container for concentrating Cs in marine water samples using ammonium molybdophosphate (50 litres sample).

Verification team commends the sophisticated H-3 electrolytic enrichment process.

Figure 5. Electrolytic low-temperature enrichment system for H-3 analysis

6.3.4 Counting rooms

The two radiological counting rooms of the CEDEX laboratory house the following equipment:

- Low-level counter Berthold LB 770-2 for Gross-beta measurements
- Gross-alpha counting system using 8 photomultipliers
- 2 Quantulus 1220 liquid scintillation counters
- 4 HPGe gamma spectroscopy systems using Genie 2000 measurement software

Verification team was informed that the calibration of the gamma spectroscopy systems includes regular check of system efficiency and energy stability, but control of the system resolution (FWHM) is not included in the routine process.

Verification team suggests that the CEDEX laboratory include control of the HPGe-system resolution in the routine system checks by establishing a trend graph of the peak width of the Co-60 1332 keV peak (FWHM).

6.3.5 Reporting

The monitoring results produced by the CEDEX laboratory are included without delay in the CSN KEEPER database; paper reports are prepared on an annual basis.

No remarks.

7 CONCLUSIONS

All planned verification activities were completed successfully. The information supplied in advance of the visit, as well as the additional documentation received during and after the verification activities, proved very useful.

The information provided and the verification findings gave rise to the following observations:

(1) Overall, the environmental radioactivity monitoring programmes of the marine environment in Spain comply with the requirements of the Article 35 of the Euratom Treaty.

(2) The verification activities found that the facilities needed to carry out monitoring of levels of radioactivity in the marine environment in the Cantabrian Sea and Galicia are adequate. The Commission ascertained that these facilities are available and running efficiently.

(3) Two suggestions have been formulated. Notwithstanding these suggestions, the verified parts of the monitoring system for environmental radioactivity in the marine environment in the Cantabrian and Galicia coastal waters in Spain are in conformity with the provisions laid down under Article 35 of the Euratom Treaty.

(4) The team's recommendations are set out in the 'Main Conclusions' document addressed to the Spanish competent authority through the Spain Permanent Representative to the European Union.

(5) The verification team acknowledges the excellent cooperation it received from all people involved in the activities it undertook during its visit.

Annex 1. Verification programme

EURATOM ARTICLE 35 VERIFICATION

Cantabrian Sea marine environment

18 OCTOBER 2021

Monday 18 October

10.00 **Opening meeting**
(CSN, C. Pedro Justo Dorado Dellmans, 11, 28040 Madrid)

- Welcome address
- European Commission Art. 35 verification programme introduction
- Verification planning

11.00 **Verification of monitoring arrangements of marine radioactivity in the Galicia and Cantabrian coastal marine environment**

- Introduction (CSN)
- Monitoring programme (CSN)

13:30 **Verifications at the CEDEX laboratory**
(CEDEX, C. de Alfonso XII, 3, 28014 Madrid)

- Laboratory introduction (CEDEX)
- Laboratory equipment
- Quality assurance
- Review of documentation

17.00 **Close**

EC verification team

Mr. Vesa Tanner ENER D3

Annex 2. Marine sampling location SMA-01

NOMBRE DEL CODIGO DE TOMA: SMA - 01
NOMBRE DEL LUGAR DE TOMA: CABO AJO - CANTABRICO
PERIODICIDAD: TRIMESTRAL **PROFUNDIDAD:** SUPERFICIAL

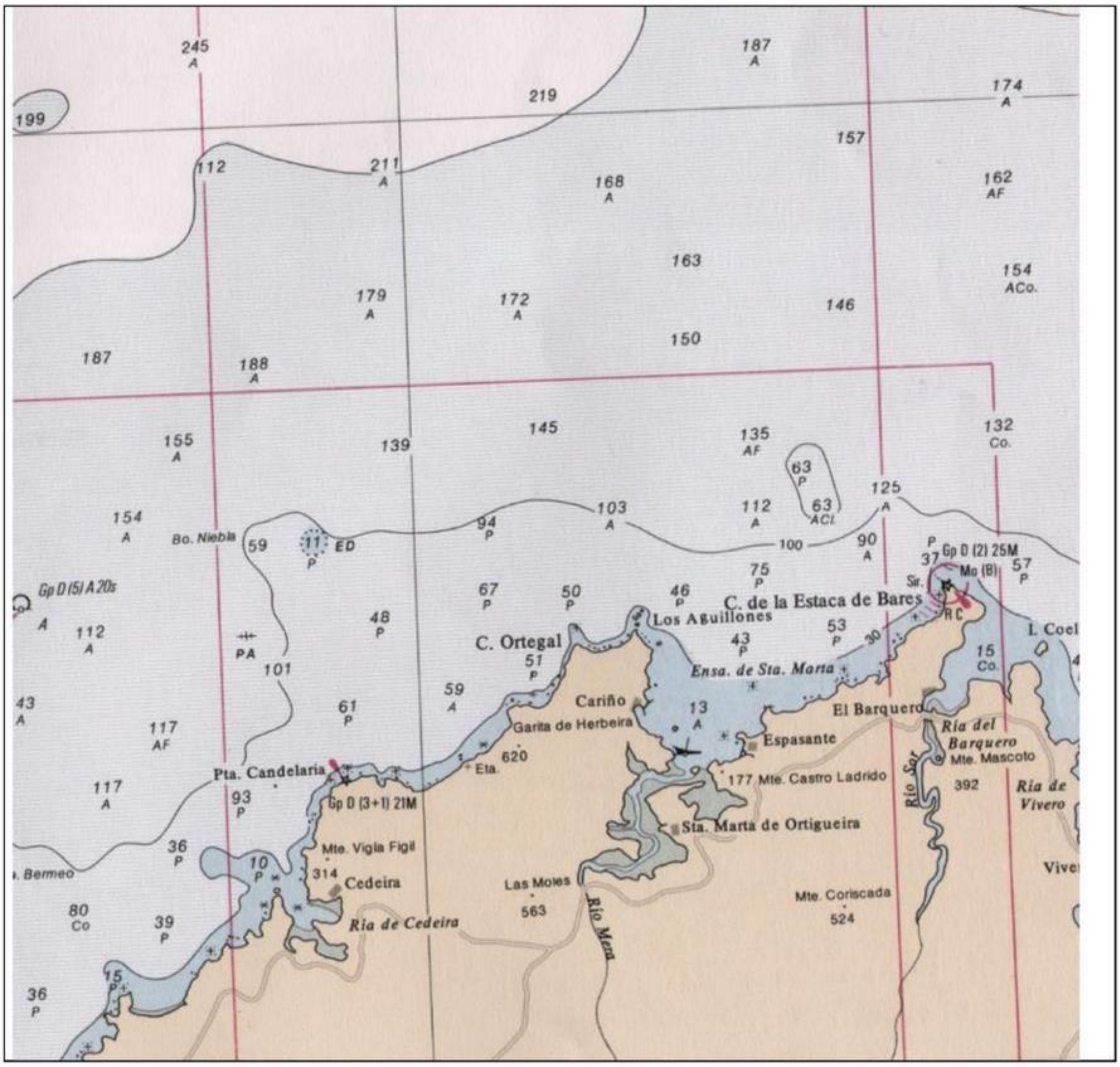
COORDENADAS:	**LONGITUD:** 3º 40´ 10´´ W	**LATITUD:** 43º 32´ 07´´ N

ESCALA 1/200.000

OBSERVACIONES: En este punto las muestras se toman por medio de la embarcación SALVAMAR DENEB.

Annex 3. Marine sampling location MAS03

CÓDIGO DEL PUNTO DE TOMA: MAS03
NOMBRE DEL LUGAR DE TOMA: CABO ORTEGAL
PERIODICIDAD: TRIMESTRAL **PROFUNDIDAD:** SUPERFICIAL



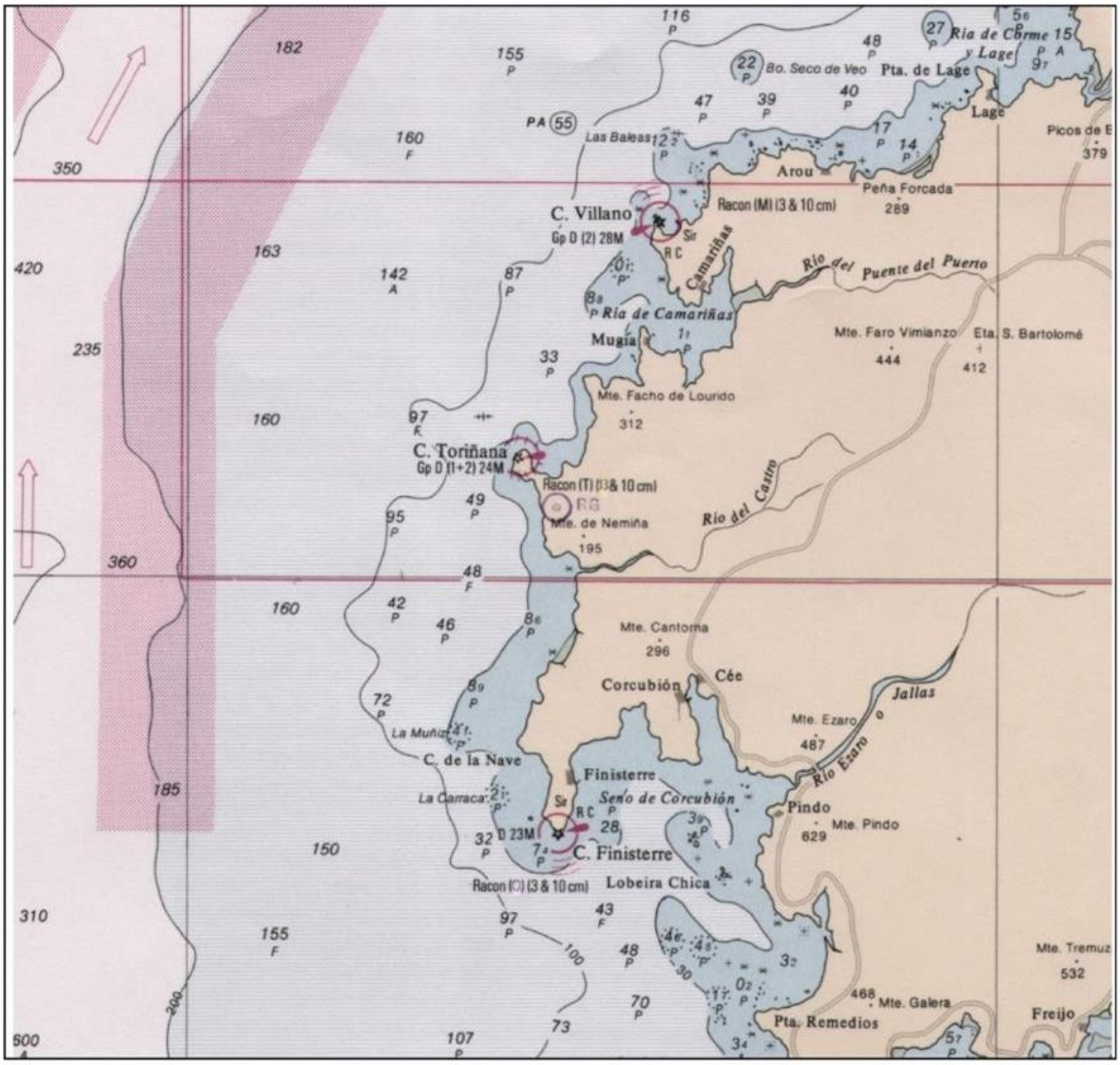
COORDENADAS:	**LONGITUD:** 7º 48' 0" W	**LATITUD:** 43º 51' 60" N

OBSERVACIONES:

La muestra la toma SALVAMAR SHAULA

Annex 4. Marine sampling location MAS04

CÓDIGO DEL PUNTO DE TOMA: MAS04
NOMBRE DEL LUGAR DE TOMA: CABO VILLANO
PERIODICIDAD: TRIMESTRAL **PROFUNDIDAD:** SUPERFICIAL



COORDENADAS: **LONGITUD:** 9º 22' 0" W **LATITUD:** 42º 59' 60" N

OBSERVACIONES:

La muestra la toma SALVAMAR ALTAIR