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Verification under the terms of Article 35 of the Euratom Treaty

Technical Report

ITALY

Gela Bay marine environment (Sicily)

18 November 2022

Reference: IT 22-07

VERIFICATION UNDER THE TERMS OF ARTICLE 35

OF THE EURATOM TREATY

FACILITIES	Radioactivity characterisation of the Gela Bay area marine environment, Sicily, Italy in 2006-2011
LOCATIONS	Rome, Italy
DATES	18 November 2022
REFERENCE	IT 22-07
TEAM MEMBERS	Mr V. Tanner Ms E-L. Diaconu
REPORT DATE	9 March 2023
SIGNATURES	

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Abbreviations

EC	European Commission
ENEA	Italian National Agency for New Technologies, Energy and Sustainable Economic Development
HPGe	High-purity Germanium
INAIL	National Institute for Insurance against Accidents at Work
ISAF	Industria Siciliana Acido Fosforico
ISIN	Italian National Inspectorate for Nuclear safety and Radiation protection
ISS	National Institute of Health
MDA	Minimum Detectable Activity
NORM	Naturally Occurring Radioactive Materials

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 Italy of its decision to conduct an Article 35 verification of radioactivity characterisation of the Gela Bay area marine environment in a letter addressed to the Italy Permanent Representation to the European Union. The Italian National Inspectorate for Nuclear safety and Radiation protection (ISIN) 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 ISIN 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

[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] Letter from ISIN to DG ENER, received on 16 September 2022

structure of the monitoring programme and on the implementation of the marine radiological sampling programme.

The verification team met in Rome the following representatives of the Italian National Inspectorate for Nuclear safety and Radiation protection (ISIN):

- Ms. Sonia Fontani, ISIN - Art. 35 and 36 Euratom Treaty contact person – Radiological area, Radioprotection, Radioactive source, Environmental radioactivity and Radiometric laboratory service
- Mr. Leandro Magro, ISIN – Responsible of radiometric laboratory Section, Radiological area, Radioprotection, Radioactive source, Environmental radioactivity and Radiometric laboratory service
- Mr. Guogang Jia, ISIN – Radiometric laboratory Section, Radiological area, Radioprotection, Radioactive source, Environmental radioactivity and Radiometric laboratory service
- Ms. Daniela Conti, ISIN - Radiometric laboratory Section, Radiological Area, Radiological area, Radioprotection, Radioactive source, Environmental radioactivity and Radiometric laboratory service
- Ms. Sara Mariani, ISIN – Radiometric laboratory Section, Radiological Area, Radiological area, Radioprotection, Radioactive source, Environmental radioactivity and Radiometric laboratory service
- Ms. Monica Buchetti, ISIN - Radiometric laboratory Section, Radiological area, Radioprotection, Radioactive source, Environmental radioactivity and Radiometric laboratory service
- Ms. Valeria Innocenzi, ISIN – Radiological area, Radioprotection, Radioactive source, Environmental radioactivity and Radiometric laboratory service
- Mr. Giuseppe Menna, ISIN – Radiological area, Radioprotection, Radioactive source, Environmental radioactivity and Radiometric laboratory service

3 RADIOLOGICAL CHARACTERISATION OF THE GELA BAY MARINE ENVIRONMENT

3.1 BACKGROUND

3.1.1 Site introduction

The former industrial site at Gela, Sicily is situated on the Mediterranean coast at the estuary of Gela River, in the south-western side of Sicily. The site (Fig. 1) produced phosphoric acid used as an intermediate in the fertilizer manufacturing until its final closure in 1992. The production process of phosphoric acid involved the treatment of phosphate minerals (phosphorites), which contain relatively high levels of naturally occurring radioactive materials (NORM), mainly originating from U-238 and Th-232 decay series. The phosphate ore used in the Gela plant came from North Africa.

Figure 1. Location of Gela, Sicily

The phosphorites were leached with sulfuric acid to produce phosphoric acid in the liquid phase and phosphogypsum in the solid phase, as an acidic waste by-product. In this process per each ton of phosphoric acid were generated about 5 tons of phosphogypsum. The radionuclides of the phosphate ore, in accordance to their solubility, are shared into the two phases of the so-called wet process: according to the literature about 86% of uranium and 70% of thorium in phosphate rock enter in the liquid phase of the phosphoric acid, while about 80-100% of its Ra-226, Pb-210 and Po-210 is fractionated to the phosphogypsum.

Based on the annual production rate, it has been estimated that during the industrial activity about 5 million tons of processing residues, mainly phosphogypsum, were created. These residues were discharged, until the early 1980s, through a pipe directly as slurry into the sea (about 150 m from the shore) in front of the Gela area; after that time the ISAF realised the waste disposal in a landfill. Consequently, from the early 1980s until 1992, when the production activity ceased, no radioactive residues were discharged into the sea.

Figure 2 presents the site and the discharge routes to the sea and to the landfill area.

Figure 2. Gela industrial site discharge routes

3.1.2 Remediation

In 2000 the Gela industrial area was included in the list of national priority sites for remediation of contaminated areas. In the early 1990s many production activities were stopped, and the ISAF shares held by Montedison and Anic were transferred to Enichem (Syndial from 2003, now Eni Rewind). At present Eni Rewind is the owner and holder of the remediation obligations for the former Anic areas; it manages the ISAF environmental activities.

In 1999 the remediation of the site began, including the permanent safety intervention at the former phosphogypsum landfill. These interventions ensured the isolation of the NORM residues from the external environment through the realisation of a bentonite-concrete composite structure. In accordance with the prescriptions of the Service Conference of 16 December 2005 with the ISIN, the National Institute of Health (ISS) and the National Institute for Insurance against Accidents at Work (INAIL), Eni Rewind was entrusted the radiological monitoring activities on site, in particular fixed sampling locations of groundwater were defined along the perimeter inside and outside of the permanent safety structure.

The remediation of the Gela site is still on-going. In 2022 the Italian authorities gave permission to dismantle the remaining plant components still containing NORM residues and transfer the material to the landfill, essentially removing all remaining NORM-material from the site.

3.2 RADIOLOGICAL CHARACTERISATION OF THE COASTAL MARINE AREA OF GELA

3.2.1 General

The need for a radiological characterization of the coastal marine area of Gela comes from the industrial activities carried out in the past in the Gela Industry Area, involving the use and storage of materials, or the production of residues, which contained natural radionuclides (NORM; Naturally Occurring Radioactive Materials).

In 2006 a radiological survey of the marine environment was initiated, based on a subset of the sampling points identified in the larger chemical characterisation plan. The Ministry of Environment, Land and Sea commissioned to the ISIN the elaboration of the radiological characterization plan, taking

into account the releases into the sea of NORM residues (phosphogypsum). In 2009 the implementation of the radiological characterization plan was entrusted to a contractor company: the radiological measurements were carried out by Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA). 10% of the analyses and measurements were repeated by the ISIN for verification and validation of data.

3.2.2 Sampling and analysis

Seawater, sediment and marine biota samples (in total 232 samples) were collected and analysed in order to assess the radiological impact of the released NORM in the marine environment. Sampling locations very far from the industrial site were considered as 'clean' control sites for natural background.

Seawater sampling included 8 sample locations (yellow squares in Fig. 3); for each sample 50 litres for alpha spectrometry and beta measurements were collected and 5 litres for liquid scintillation analysis. Sampling was carried out by a rosette sampler and within 24 hours of collection the samples were filtered with 0.45 mm cellulose nitrate filters in order to separate the water from the particulate matter in suspension; the two matrices were separately analysed.

Marine biota was sampled by collecting bivalves (mussels) and necto-benthonic species, for each sample 1.5 kg of fresh weight was used for gamma spectrometry analysis and 200 cc of dry weight for Pb-210 and Po-210 measurements.

Marine sediments were sampled using a core sampler at depths 0 – 200 cm. Samples were sliced on 10 cm samples. Samples were subjected to a radiochemical analysis and gamma spectrometry; some samples were kept and analysed only if other samples had showed elevated radioactivity levels. Altogether 159 sediment core samples were analysed.

Table I illustrates the sampling programme. Figure 3 presents the sampling locations. Table II presents the analytical methods used.

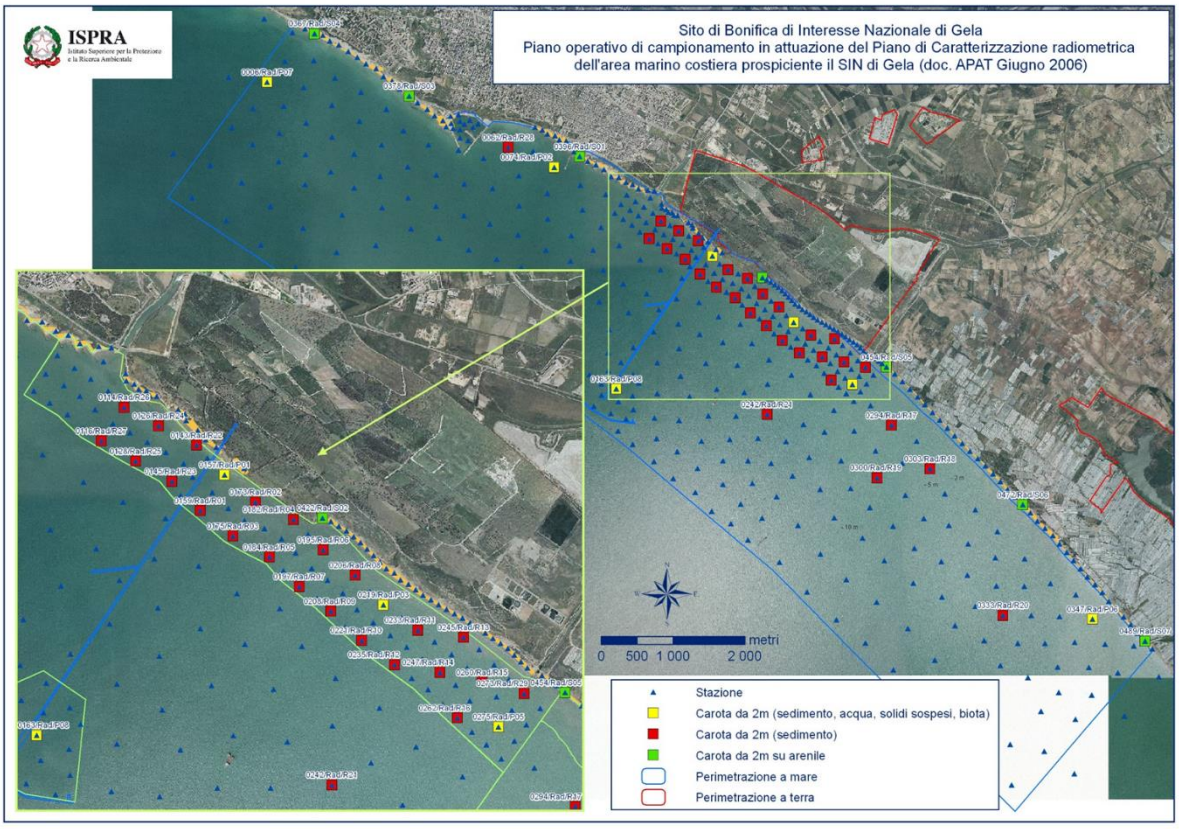

Figure 3. Sampling points of the marine radiological characterisation programme

Table I. Sampling programme

Coastal area	
Corer dimensions: diameter (min.) x length [cm x cm]	10 x 200
N. sampled sediment cores (,)	37
N. sampling points for vertical profile characterisation (sediment cores, seawater, suspended particulate, marine biota)	8 / 37
N. samples (sections) for each sediment core ( ) for gamma spectrometry analysis	4
N. samples (same sections of gamma analysis + surface layer) for each sediment core ( ) for alpha and beta emitters analysis	5
N. samples (sections) for each sediment core ( ) for gamma spectrometry analysis	1
N. samples for each sediment core ( ) for alpha and beta emitters analysis (same sections of gamma analysis + surface layer)	2
Total analysed sediment samples (4x8 + 5x8) + (1x29) + (2x29)	72 + 87 = **159**
Beach	
Gamma dose rate measurement	7 + 1 (blanc) =8
Corer dimensions: diameter (min.) x length [cm x cm]	10 x 200
N. sampled cores ( )	7
N. samples for each core (3 + 2)	5
N. total samples (3x7 + 2x7)	35
N. total cores ( )	**44**
***N.* analysed samples**	**194**

Table II. Analytical methods

RADIONUCLIDE	**MEASUREMENT TECHNIQUE**
Ra-226	Gamma spectrometry; Alpha spectrometry; Liquid Scintillation Counting
Pb-214	Gamma spectrometry
Bi-214	Gamma spectrometry
Pb-212	Gamma spectrometry
Bi-212	Gamma spectrometry
TI-208	Gamma spectrometry
Ac-228	Gamma spectrometry
Pa-234m	Gamma spectrometry
Th-234	Gamma Spectrometry
U-238	Alpha Spectrometry; ICP Mass
U-235	Alpha Spectrometry; Gamma Spectrometry; ICP Mass
U-234	Alpha Spectrometry
Po-210	Alpha spectrometry
Pb-210	Beta counting; Gamma Spectrometry
Cs-137	Gamma spectrometry
K-40	Gamma spectrometry

3.2.3 Results

The aim of the study[4] was to estimate the NORM concentrations in the residual product and by-product of the past industrial activity, and to investigate the NORM contamination in the sea area of Gela. For radioecological comparison Lecce was selected as a control site of natural background, it being geologically near and similar to the Gela area.

The conclusions of the radiological characterization were that the concentration activity of natural radionuclides (U-238, U-235, U-234, Ra-226, Po-210, Pb-210) of the analysed seabed samples were comparable to the local natural background on the seabed (U-238Sec ≈20 Bq/kg, Th-232Sec ≈15 Bq/kg). Only in one core sample of marine sediment, at 60-100 cm depth, the concentration activity of natural radionuclides equal to about 4 or 5 times the background values were measured; it was considered as a single not homogeneous spot. Considering that all the radionuclides of the U-238 and Th-232 decay series were in secular equilibrium (U-238Sec ≈125 Bq/kg, Th-232Sec ≈32 Bq/kg) the radionuclide content of the sample can be related to geological features of the spot and not to anthropogenic ones. It has been reported that as a naturally occurring radionuclide the abundance of U-238 ranges from 1.24 to 249 Bq/kg in the Earth's crust.

The seawater and marine biota analysis showed that the levels of activity concentrations of natural radionuclides were fully compatible with the natural background of uncontaminated areas.

3.2.4 ISIN control measurements

Radiochemical analyses were performed by the ISIN laboratories to measure the activity concentrations of Po-210, Pb-210, uranium and radium isotopes on 4 phosphogypsum samples, 5 phosphoric acid samples collected in 2018 inside the industrial area and 20 marine sediment core samples collected in 2010 in front of the industrial Gela site.

The activity concentration of U-238 in the analysed marine sediments ranged from 10.4 to 26.3 Bq/kg with a mean value of 16.7±5.6 Bq/kg, that of U-234 from 10.9 to 27.2 Bq/kg with a mean value of 17.0±5.6 Bq/kg and that of U-235 from 0.285 to 1.28 Bq/kg with a mean value of 0.698±0.283 Bq/kg. The U-234/U-238 and U-235/U-238 ratios in the samples were 1.02±0.04 and 0.0415±0.093, respectively. According to the ratios, it was concluded that the uranium in the sediment samples is of natural origin. These results were comparable and statistically lower than the activity concentration of the control site (U-238 ranging from 13 to 26.4 Bq/kg with mean value of 21.6±4.4 Bq/kg; U-234 ranging from 12.9 to 27.4 with a mean value of 22.5±4.6 Bq/kg and U-235 ranging from 0.51-1.29 Bq/kg with a mean value of 0.913±0.249 Bq/kg). It is also pointed out that the activity concentrations of U-238 in the Gela sediment samples were in the lower band of the natural range abundance of the Earth's crust; therefore, the radioecological impact of the discharged NORM as phosphogypsum is not observable.

The activity concentration measured in the phosphogypsum waste samples was not very high (mean value 38±23 Bq/kg) and this is in accordance with the wet production process mentioned in section 3.1.1. During the acid attack of the phosphate ore uranium enters mainly in the liquid phase (the phosphoric acid) and not in the solid residual phase (phosphogypsum); actually, very high average value 2039±176 Bq/l of uranium activity concentration was measured in acid phosphoric samples. For this reason, it is important to consider that uranium is not the radionuclide of major radiological concern in case of phosphogypsum; Ra-226 is assumed to be the radionuclide guide to detect phosphogypsum contamination, since in that case a strong disequilibrium between activity concentration of Ra-226 and U-238 (Ra-226 >> U-238) should be observed. In the analysed sediment samples of Gela, Ra-226 ranged from 12.6 to 32.7 Bq/kg with a mean value of 19.6±6.0 Bq/kg, which

[4] ISPRA technical report "Attività di supporto tecnico operativo di analisi e di validazione di dati relativamente alla caratterizzazione radiometrica dell'area marino costiera prospiciente il Sito di Interesse Nazionale di Gela", 2011.

is comparable with the average values of U-238 (16.7±5.6 Bq/kg), therefore no radium contamination was observed in the sediment samples. As for the activity concentration of Th-234, considering that it is the first daughter in the U-238 decay chain, with a short lifetime ($T_{½}$=24.1 days) and, considering that the release of phosphogypsum into the sea occurred until 1980s and no available soluble uranium compounds were detected, the concentration activity of Th-234 and U-238 can be considered equal, being daughter and parent in secular equilibrium.

According to the ISIN conclusions, the radiological characterisation in the sediment samples in Gela Bay area showed that the activity concentrations of Po-210, Pb-210, radium and uranium isotopes in the samples are comparable to natural background and no contamination was observable. Therefore, it can be assumed that the NORM, together with phosphogypsum released in the investigated inshore, could have been diluted or dispersed a long way to sea from the inshore area by tide and wave action of the seawater during the past 30 years.

4 VERIFICATIONS

4.1 INTRODUCTION

Verifications were carried out based on documents and reports provided by the ISIN. This chapter summarises the verifications carried out the by the verification team. The team has assessed the monitoring 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.

4.2 GELA BAY RADIOLOGICAL CHARACTERISATION 2006 - 2011

The verification team discussed the radiological characterisation carried out at the Gela Bay marine environment with the ISIN experts and it has reviewed the documentation provided by the Italian authorities. The team concludes that the radiological characterisation of the coastal marine area of Gela, Sicily was made in a very thorough manner; it covered the relevant geographical area and included all the relevant environmental components (sea water, bottom sediments and marine biota).

Industrial activities at the Gela site have ceased, a bentonite-concrete composite structure has been built to isolate the phosphogypsum landfill, and there have been no further discharges of NORM material into the marine environment, so the results of the radiological characterisation carried out in the period 2006 - 2011 remain valid.

> *The verification team suggests that the competent authorities continue to follow the dismantling of the ISAF plant and the remediation activities of the site.*
>
> *The verification team commends the level of detail and the impressive analytical effort made to characterise the radiological status of the marine environment at the Gela Bay.*

5 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) The verification activities found that the radiological characterisation carried out in 2006 – 2011 at the Gela Bay marine environment was adequate. The Commission ascertained that the process was carried out in sufficient environmental coverage and in good radioanalytical manner.

(2) Industrial activities at the Gela industrial area have ceased and there have been no further releases of NORM material into the marine environment, so the results of the characterisation remain valid.

(3) At present there is no need for further monitoring actions or further radiological characterization of the coastal marine area of Gela, considering that industrial activities at the Gela industrial area have ceased and there have been no further releases of NORM material into the marine environment.

(4) It is suggested that the competent authorities continue to follow the dismantling of the ISAF plant and the remediation activities of the site.

(5) The team's conclusions are set out in the 'Main Conclusions' document addressed to the Italian competent authority through the Italy Permanent Representative to the European Union.

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