IL NUOVO CIMENTO **46 C** (2023) 40
DOI 10.1393/ncc/i2023-23040-y

Colloquia: SOGIN 2023

Chemical and NORM management in the Contaminated Sites of the Italian National Priority List

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received 7 March 2023

Summary. — Contaminated Sites of the Italian National Priority List, being the legacy of industrial development, have different types of contamination to be managed with different approaches, different legislative procedures, and different control authorities; in particular between chemical and radiological contamination. This may lead to a bottleneck and to a delay in the operations needed for remediation. A synergistic approach involving all affected stakeholders certainly could reduce the time and cost of interventions measures.

1. – Introduction

In Italy, the Contaminated Sites of the Italian National Priority List (SIN) are 42 large industrial complexes that ensured the massive employment of local labor and the creation of economic resources of strategic relevance for the entire country but, in particular during the second half of '900, produced extended and severe chemical pollution. Several of these industrial sites hosted the production of fertilizers with different types of phosphate products. The processing of the phosphate ore implied the generation of large amounts of waste materials, in the form of sludge, gypsum, slags, and air particulate. These wastes were dispersed or disposed of in the surrounding environment with different activity concentrations of natural radionuclides. In the present paper, some site-related cases are illustrated as examples of actual "on-field" experiences. All of those past NORM activities ceased around the beginning of the '90s. The present effort for the remediation of the above-mentioned SIN implies the management of both chemical and radiological pollution and related risks.

Such complex problems of risk management must be faced taking into proper account, in every phase of the remediation plan, the combined actions to identify the best technical

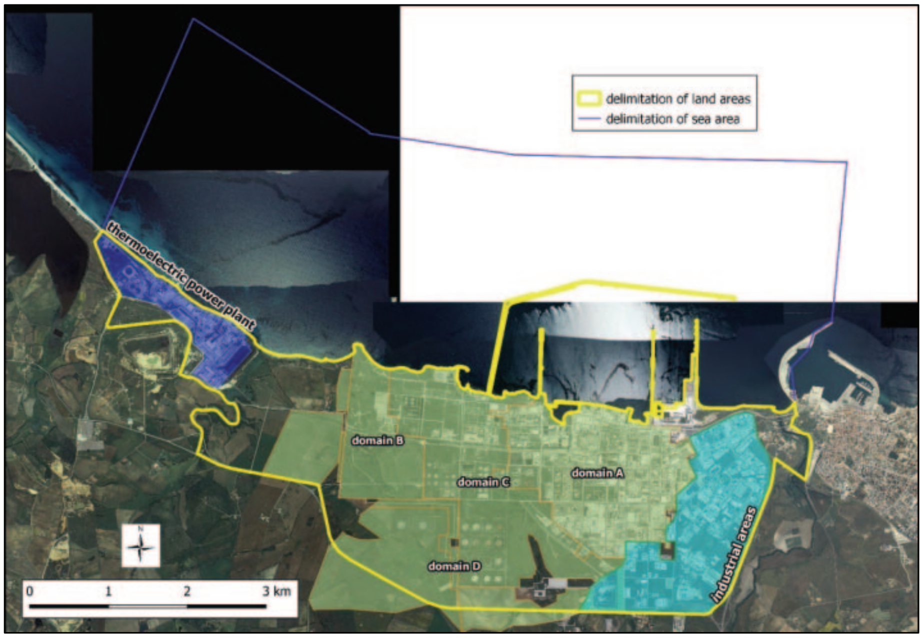

Fig. 1. – Areal Delimitation of SIN Porto Torres.

solutions which, in most cases require the active contribution and mutual interaction of all involved stakeholders.

2. – The SIN of Porto Torres (North Sardinia)

The contaminated site of national interest (SIN) of Porto Torres is located in the northern part of Sardinia; it was defined contaminated area, by the Ministry for the Environment (MASE), with an act dated 7/02/2003. The area of the SIN is 1874 ha (land part) and 2700 ha (sea part). Within the SIN can be distinguished the following macro areas: the petrochemical pole, the thermoelectric power plant, industrial areas and coastal depots, sea area (fig. 1).

The whole area of the petrochemical plant, which covers more than 1100 ha, in terms of plant size, quantity, variety, and quality of the contaminants, is the most polluted area of the SIN. This huge part of the SIN was divided into 4th different sub-areas: domain "A" (310 ha), which includes the industrial facilities and the areas of the tanks; domain "B" (100 ha), on the western part of the site, in which are included so-called landfill sites of "Minciaredda" and "Cava Gessi"; domain "C" (320 ha), between "A" and "B", in which is located the "Palte area" and domain "D" (about 350 ha), in the southern part of the site. The area of the thermoelectric power plant "Fiume Santo" extends for about 140 ha and it is located westward, outside the main part of the SIN.

The environmental characterization of the different parts of the site, according to the Italian legislation (D.lgs. 152/2006), has proceeded with different times and modalities, depending on the private entities, the size of the areas, the presence of active or abandoned industrial activities and the type of contamination and pollutants.

2˙1. *Chemical contamination and remediation activities in the SIN of Porto Torres.* – Linked to the presence of several primary and secondary sources of contamination, spread all over the SIN, a heterogeneous situation emerged. Soil pollution, due especially to primary source of contamination, is mainly located in domain "B" where "Minciaredda" and "Cava Gessi" are placed. A second primary source was identified in domain "C", where the "Palte Fosfatiche" and the "Peci DMT" deposits are located. Generally speaking, the main contaminants identified in soils belong to BTEX (up to 750 times more than the concentration limit), chlorinated aliphatics (up to 1800 times more than the concentration limit), and metals (to a lesser extent) [1].

Groundwater beneath the SIN also revealed the presence of primary sources of contamination such as LNAPL and DNAPL, mainly found in domains "A" and "B". Groundwater contamination is evenly distributed throughout the whole site and very significant concentrations; for example, maximum concentrations compared to the regulatory limits of more than one million times for BTEX and chlorinated compounds have been reached.

In brief, such a delicate framework has required many environmental safety actions and remediation. Concerning groundwater, since 2005 a hydraulic barrier along the seaside, in the northern part of the SIN, has been installed together with several Pump&Treat activities and treatment modules. Moreover, in recent years, a multi-phase extraction system is active in the northern part.

Looking upon the soil, in 2020 the Nuraghe integrated Project started. The core of the project is a multifunctional platform built for the treatment of excavated land and waste. The main action is planned in the area of "Minciaredda" with a desaturation process (MPE system) that will remove liquids and toxic gases before the waste removal activities.

The site of Porto Torres, in addition to being very extensive and having multiple industries (active and abandoned), has an extremely complex geological and hydrogeological structure.

In particular, the presence of two main carbonate aquifers [2] implies an important secondary permeability, due to geological fractures linked to karst or structural phenomena, which implies that the water reservoir is strongly anisotropic. The direct implication is the presence of pathways of contaminants is often not linear and is strictly dependent on the number, size, and orientation of the fractures and karst conduits.

The fact that it is not possible to identify clearly a direct correlation between sources of contamination, pathways, and receptors, often complicates the remediation from both technical and administrative points of view.

2˙2. *Radiological contamination and remediation activities in the SIN of Porto Torres.* – In this environmental framework, in addition to the so-called "conventional contamination", also the theme of radiological remediation occurs.

In the past, in Porto Torres industrial areas, there was industrial activity linked to the production of phosphoric acid. The main process was the wet one, through which the phosphoric acid is produced from phosphate rock. The residues and waste of this technique are phosphogypsum, refining sludge, scales, sediments, and slag. The most important pollutants are radioactive elements, such as the U-238 decay chain and heavy metals.

In Porto Torres these kinds of materials were stored in the following landfills: "Cava Gessi landfill": 21 ha - 2×10^6 m^3 (phosphogypsum + chlorinated solvents) and "Palte phosphate storage area": 37.000 m^2 - 9.500 m^3 of palte phosphate and 21.000 m^3 of palte phosphate mixed with soil (fig. 2).

Fig. 2. – Identification of NORM affected area in Porto Torres.

These areas are now object to specific characterization, in order to determine the radiological risk and, if necessary, to prepare the arranged mitigation and remediation actions.

In the future, combining information on chemical and radiological contamination could help to fill some information gaps related, for example, to the definition of the conceptual model of the site or to the definition of the best solution for the remediation of the area.

From this point of view, the "Palte" represents a significant example of the challenges posed by the problem of managing the overall risk related to the presence of some particular kind of polluting materials. This type of highly hygroscopic, fine-grained, industrial sludge was generated by the refining process of the phosphoric acid solution, as the product was intended for human consumption (e.g. detergents) and had to be purified from the presence of heavy metals. The result is a NORM technically enhanced in the content of specific chemical elements which poses both toxicological and radiological hazards, with particular regard to uranium concentration. Moreover, the sludge exhibits low pH and they eventually shall be treated, by means of a dedicated plant, in order to get chemically inert and insoluble residuals, before being posed in a safe location, in compliance with both the regulation on industrial waste management and the radiation protection.

3. – The SIN of Gela (South Sicily)

The Gela SIN, due to its previous industrial history, which began in the 1950s, was among the first sites identified at a national level to be subjected to remediation. The

Fig. 3. – Areal Delimitation of SIN Gela.

site, bounded by decree DM 10/01/2000, includes both land (about 765 ha) and sea areas (4600 ha) as shown in fig. 3.

In the SIN Gela, there is a multi-company plant, including a refinery, a thermoelectric plant and chemical industries, and waste landfills of residuals from the industrial cycles. Moreover, there are some areas of crude oil extraction well and n. 2 oil collection centers in the south part; the public areas include the coast and port of Gela and the site of naturalistic interest, "Riserva Naturale Orientata Biviere di Gela" Sites of Community Importance (SIC) belonging to the Natura 2000 network.

3·1. *Contamination and remediation activities in the SIN of Gela.* – During the characterization activities, carried out in the early 2000s, a significant presence of organic and inorganic pollutants was detected in soils and groundwater. Main soil contaminants are metals (arsenic, mercury), carcinogenic chlorinated aliphatic compounds (mainly 1,2-dichloroethane, vinyl chloride), ammonia, BTEX (benzene, ethylbenzene, toluene, p-xylene), PCBs, PAHs (polycyclic aromatic hydrocarbons), $C \leq 12$ and $C > 12$ hydrocarbons. In the groundwater the contaminants are Al, V, As, B, Mn, Cd, Pb, tetraethyl Pb, Fe, Co, Sulfates, Chrysene, Ni, PAHs, BTEX, MTBE, PCBs, p-xylene, o-xylene, total hydrocarbons (n-hexane), vinyl chloride, 1,2-dichloroethane, 1,1-dichloroethylene, trichloroethylene, toluene, chloroform, hexachlorobenzene, dibenzo(a,h) anthracene, benzene, ethylbenzene, styrene, toluene. Figure 4 shows the status of soil remediation procedures in the SIN, updated to June 2022 [3].

As regards soil, the human health and environmental risk assessment (AdR) of part

Fig. 4. – Status of soil remediation procedures in the SIN, updated to June 2022

of the refinery areas was approved in 2021 (M.D. No. 211) and for which the preliminary service conference (CdS) of the remediation project and the containment measures (MISO) are underway; for the remaining areas eliminated because they are highly impacted, prevention and mitigation measures are underway pending the arrival of a new version of AdR. In accordance with the technical approach adopted by ISPRA and ARPA for the SIN of Gela (so-called "Gela model"), the criteria according to which they should be excluded have been identified from the AdR, at least initially, all the areas that had presented one or more of the following critical issues:

1) presence of Light Non-Aqueous Phase Liquid (LNAPL) in the groundwater;

2) consistent exceeding of the residual saturation concentration in the unsaturated zone;

3) high concentrations of contaminants in soil gas.

The primary sources of contamination must in fact be removed or made safe in order to avoid further propagation of the contamination.

For the multi-company plant of Gela, by decree dated 06/12/2004 and subsequent amendments, the Groundwater Remediation Project was approved, consisting of a double containment system at sea consisting of a hydraulic barrier and 2 sections of plastic diaphragms, carried out taking into account the hydrogeological structure of the site (fig. 5). The drained waters are sent to a special Groundwater Treatment Plant (TAF). Periodic monitoring is implemented on the systems in operation to check their efficiency/effectiveness.

Fig. 5. – Summary scheme of the systems of the PDB groundwater remediation project [4]

With regard to soil remediation, in addition to the "dig & dump" technique, in situ treatment technologies have also been used. Among these, the most important example is the remediation by thermal desorption of Vasca A Zona 2 of the Gela Refinery (fig. 6), which is about to enter the final stages of the treatment.

The area of Vasca A Zona 2 constitutes one of the 5 tanks of the old landfills located in the south-eastern portion of the Gela Refinery, in Isola 32. The tank is presented in the plan as a quadrilateral having an area, measured at ground level, of approximately

Fig. 6. – Area "Vasca A zone 2" (ISPRA/ARPA/Eni inspection photo of May 2019)

5.700 m^2. The tank bottom is located at a height of about 7.5 m from the ground surface, while the water table lies in a sandy aquifer at a depth of about 12-14 m from the ground floor. The tub is affected by reclamation interventions on the basis of a specific project authorized by the Decree of the time Ministry of the Environment in the 1st version in 2004 and in the following two variants in 2009 and in 2018.

This project involves a first phase of removing the oily residues present in the tank, with disposal in authorized plants, and the subsequent reclamation of the underlying unsaturated soils and surrounding the tank itself, contaminated by BTEX, THC>12, THC< 12, PAH, chloromethane, vinyl chloride and 1,2-dichloroethane [5].

3˙2. *Radiological contamination and remediation activities in the SIN of Gela.* – In addition to the so-called "conventional contamination", also the theme of radiological remediation occurs. In the past, in the Gela complex, there was a plant for the production of phosphoric acid by wet process, through which the phosphoric acid is produced by attacking the phosphate ore with a sulphuric acid solution. The residues and wastes of this technique are phosphogypsum (calcium sulfate dihydrate), scales, sediments, and slag. The most important pollutants are radioactive elements belonging to the U-238 decay chain and heavy metals. In Gela, highly diluted phosphogypsum was initially discharged into the sea, by means of a pipeline, a few hundred meters from the coastline, in front of the industrial complex.

At the beginning of '70s, the direct release to the sea of gypsum slurry was prohibited, due to major changes in the environmental regulation. A new pipeline was then realized to pump the slurry into a landfill, located above a deep layer of clay (fig. 7). Over the

Fig. 7. – Phosphogypsum landfill before complete remediation (ISPRA/ARPA/Eni inspection photo of 2006).

years, until the early 1990s, when the plant was abruptly shut down and all phosphate production ceased, the landfill was fed by gypsum sludge. The total amount of phosphogypsum, partially dried by the sun, is of the order of several million tons, distributed over an area of over 20 ha, with a depth varying between, indicatively, 10 m and 20 m.

At present, the radiological remediation of the site is in progress and some relevant results have already been achieved:

- The permanent remediation of the phosphogypsum landfill has been finalized, including a water treatment plant for the leachate. Inside the perimeter of the former landfill, a new "specific NORM Landfill" has been authorized and realized for further residues that will be generated during the next decommissioning phases.
- Several plants for waste inertization have been realized and monitoring plans aimed to check and assure the correct operation of the process, have been defined, taking into account the technical indications formulated with the contribution of all of the experts, from all of the involved stakeholders.

The next steps are related to the dismantling of the dismissed Phosphate Plant building (fig. 8). These areas are now subjected to specific characterization, in order to determine the radiological risk (fig. 9) and, if necessary, to prepare the arranged mitigation and remediation actions. The plan will last for some decades.

4. – Conclusion

The effort for the remediation of the above-mentioned SIN implies the management of both chemical and radiological pollution and related risks. The reported examples of

Fig. 8. – Dismissed Phosphate plant (ISPRA/ARPA/Eni inspection photo of 2006)

Fig. 9. – Dismissed Phosphate plant. Restricted area due to gamma radiation emission from scales in tanks, mostly, Radium sulphate concretions (ISPRA/ARPA/Eni inspection photo of 2006).

Gela and Porto Torres, as largely dismissed industrial complexes that are deeply affected by chemical pollution and NORM residues, are useful to remark the need for combined and coordinated work, among several stakeholders, since the very beginning phase of each operative project, such as the characterization of the environmental status, the collection and sharing of the information and the definition of the "conceptual model" of the site.

The regulatory approach to the decision about the feasibility/admissibility of environmental projects (submitted by the liable company) is based on the "splitting" in sub-decisions about the compliance with the requirements set by each regulatory framework, for each of the separate aspects of the global problem.

In theory, the global decision about the accuracy of the suggested solution for the management of each kind of risk and benefits involved should run on parallel railroads. In practice, even an unpredicted stop on a singular railroad is able to cause the prolonged stop of the whole process and the loss of timing and coordination between the separated phases of the decisional process.

The effect of a non-approval of a specific aspect of the problem, may affect the whole project and force a complete revision of the plan. Such a "feedback effect" causes a loss of time and resources.

In conclusion, it is recommendable that such complex problems of risk management, that include themes of different nature such as chemical and radiological pollution, be faced by taking into proper account, in every phase of the remediation plan, each aspect of the problem itself. The combined actions for the finding of the best technical solution require the active contribution and the mutual interaction of all involved stakeholders.

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