1A holistic DPSIR-based approach to the remediation of heavily 2contaminated coastal areas

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14**Abstract**

15This paper proposes a holistic approach to connect anthropogenic impacts to 16environmental remediation solutions. The eDPSIR (engineered-Drivers-Pressures-States-17Impacts-Responses) framework aims at supporting the decision-maker in designing 18technological solutions for a contaminated coastal area, where the natural matrices need to 19be cleaned up. The eDPSIR is characterized by cause-effect relationships that are 20operationally implemented through three multidisciplinary toolboxes: (i) Toolbox 1, to 21connect driving forces with pressures, classifying the state of the system and allowing the 22identification of target contaminants and the extent of contamination; (ii) Toolbox 2, to

23quantify bioaccumulation also by identifying corresponding areas; (iii) Toolbox 3, to 24identify the most suitable remediation solutions for previously identified contaminated 25areas, named contamination scenarios. The eDPSIR was calibrated on the case study of the 26Mar Piccolo in Taranto (Southern Italy), one of the most complex and polluted areas in 27Europe. While the consolidated DPSIR allows for a strategic response by limiting the use 28of contaminated areas or reducing upstream pressures, the eDPSIR made it possible to 29structure with a semi-quantitative logic the problem of assisting the decision-makers in 30choosing the optimal technological remediation responses for each sediment scenario of 31contamination (heavy metal; organic compounds; mixed). Assisted natural attenuation was 32identified as the best remediation technology in terms of treatment effectiveness and 33smallest amount of impacts involved in the project actions. However, considering the 34scenario of mixed contamination, *in-situ* reactive capping reached a good rank with a value 35of the composite indicator equal 99.5%; thermal desorption 36stabilization/solidification recorded a value of 94.1% and 84.6%, respectively. The 37application of these toolboxes provides alternative means to interpret, manage, and solve 38different cases of global marine contaminated sites.

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40Capsule about main findings:

41To develop and apply a new DPSIR-based decision-making approach capable of 42structuring and solving a complex problem such as heavily contaminated coastal areas.

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44Keywords: environmental health risk; marine sediments; multi-criteria analysis; pollution

45clean-up; sustainable remediation technologies

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47List of abbreviations

48ANA = Assisted Natural Attenuation

49BOD = Biochemical Oxygen Demand

50CA = Cluster Analysis

51COD = Chemical Oxygen Demand

52CSM = Conceptual Site Model

53DPSIR = Driving force—Pressure—Status—Impact—Response

54EAC = Environmental Assessment Criteria

55ERA = Ecological Risk Assessment

56HQc = Hazard Quotient for chemistry

57MCDA = Multi-Criteria Decision Analysis

58MOA = Matrix of Alternatives

59PAHs = Polycyclic Aromatic Hydrocarbons

60PCA = Principal Component Analysis

61PCBs = Polychlorinated Biphenyls

62PCT = Paired Comparison Technique

63PI = Preference Index

64RTR = Ratio-to-Reference

65SAW Simple Additive Weighing

66SEA = Strategic Environmental Assessment

67TOC = Total Organic Carbon

68TWG = technical working group

69WOE = Weight of Evidence

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711. Introduction

72The rapid economic development and industrialization have led to intensive urbanization 73with severe environmental problems. The main anthropic impacts, including wastewater 74discharge, industrial operations, and improper waste management, have compromised the 75whole ecosystems worldwide (World Health Organization, 2016). As a result, health 76problems have arisen for humans, due to the deterioration of environmental matrices and 77contamination of the trophic chains. The main pollutants contaminating natural ecosystems 78include microbial pathogens, organic compounds, metals/metalloids, emerging pollutants 79and modern pesticides (Bortone et al., 2020). Some pollutants would leave long-term 80damage to the ecosystem as in the case of Polychlorinated biphenyls (PCBs) (Li et al., 812009). Many approaches of environmental pollution management and mitigation measures 82related to ecosystem and human health are discussed in literature. Among them, the 83Driving force—Pressure—Status—Impact—Response (DPSIR) has been widely documented to 84analyze environmental issues. It is an approach that considers a chain of causal links 85starting with "driving forces" (e.g., economic and social needs), which create "pressures"

86(e.g., emissions, discharges) that in turn can change the state of environmental matrices, 87with consequent impacts on the environment and humans.

88The existing literature on the models adopted to address pollution issues in coastal systems 89demonstrated a range of empirically oriented DPSIR applications. Atkins et al. (2011) 90integrated DPSIR with ecosystem services to create a framework to support decision-91making in the marine environment. Cook et al. (2014) explored the interactions among 92coastal ecosystem pressures, states, and services, quantifying the impact through matrix-93based analyses. Pastres and Solidoro (2012) showed dynamic models can help to improve 94understanding of ecosystem dynamics, thus providing a quantitative basis for the DPSIR 95framework. Zhang and Xue (2013) highlighted the need for further progress in the DPSIR 96logic, which could be achieved by establishing a unified information system and by 97improving indicator development for marine environmental problem assessment. By 98developing a system of indicators, Liu et al. (2018) highlighted how coastal ecosystems are 99dynamic and fixed-point monitoring should be conducted in the long-term to obtain 100dynamic data. Delgado et al. (2020) pointed out that in conflict situations the DPSIR model 101needs validation by local experts and social actors. Recently, Lewison et al. (2016) 102highlighted that the use of quantitative approaches in DPSIR research is relatively limited 103and partly due to the challenges of merging different data types. They concluded by 104showing how the search for cause-effect relationships as quantitative as possible is still a 105work in progress; new research should be directed towards the development of semi-106quantitative relations between the various components of the DPSIR with a view to future

107generalization. Additionally, Bell (2012) explained why DPSIR, by its nature, is a narrowly 108formulated device, incompatible with most of multiple perspectives, which human 109interaction requires. Furthermore, it has been argued to be very limited when applied to 110sustainability research and to be incomplete as problem structuring and communication 111method (Wieket al., 2011).

112The aim of this work is to propose an engineered version of the DPSIR framework able to 113assess which factors contributed to the pollution of a complex marine ecosystem and to 114identify the best technological responses in quantitative terms, such as the selection of 115appropriate remediation technologies. The new implemented framework, called eDPSIR, is 116based on three multidisciplinary steps, indicated with the term "toolboxes", able to connect 117(i) driving forces with pressures and states, (ii) states with impacts, and (iii) technological 118responses with all the previous DPSIR components. One important purpose of this 119implemented DPSIR framework is to overcome the communication gap between scientific 120systems and public. The proposed approach was calibrated considering a complex and 121heavily contaminated site in Southern Italy, the Mar Piccolo of Taranto. To our knowledge, 122this is the first example of an attempt to explicitly integrate pressures, environmental 123issues, and potential remediation actions into a single framework.

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1252. Materials and methods

126**2.1. Case study**

127The Mar Piccolo of Taranto is a semi-enclosed marine basin in Apulia, Southern Italy (Fig.



Fig. 1. Localization of the Mar Piccolo case study.

133It has a surface area of 21 km² divided in two inlets, called First Seno and Second Seno. 134This area has been subjected to chemical pollution originating from industrial activities for 135several decades, such as the steel plant ex-ILVA, the concrete factory Cementir, the Eni 136refinery station, and the Military Arsenal (Labianca et al., 2018). In addition, this basin has 137been strongly impacted by human activities such as wastewater discharge and mussel 138farming. It is also characterized by scarce water circulation, encouraging organic matter 139sedimentation and subsequent accumulation of pollutants in sediments (Cardellicchio et al.,

1402006). A particular characteristic of this basin is the presence of some submarine 141freshwater springs called "citri". The pressures exerted on the Mar Piccolo have 142contributed to the deterioration of environmental matrices, as shown in **Table 1**.

144 Table 1. Pollution in the environmental compartments of the case study.

N.	Environmental compartment	Local pressures	Pollutants	References
1	Sediment	 Industrial activities (steel, oil, metals, varnishes, chemicals, paints), Agricultural activities (run-off of chemicals), Vessel traffic (Military Arsenal, fishing, mussel farming), Illegal discharges of nutrients and contaminants, Natural phenomena (e.g., absorption). Vessel traffic (Military Arsenal, fishing, mussel farming), 	 PAHs: 56-36370 μg/kg PCBs: 20-9391 μg/kg Metals: As (3.77- 48 mg/kg), Cd, (0-1.5 mg/kg), Cu (9.1-172.7 mg/kg), Hg (0-16.7 mg/kg), Ni (23.33-60.66 mg/kg), Pb (24.42-272.54 mg/kg), V (25.67-95.61 mg/kg), Zn (38.51-602.89 mg/kg). (kg of dry solids). PAHs: >0.2 pg/L PCBs: 4652 pg/L 	(Todaro et al., 2019)
2	Marine water	 Illegal discharges of nutrients and contaminants, Natural phenomena (e.g., desorption, advection, diffusion of pollutants from/through sediments). 	 Metals: Cd (>0.3 μg/L), Cr (>0.3 μg/L) and Pb (>7.2 μg/L) 	(ARPA PUGLIA, 2014)
3	Air	 Industrial gas emissions, Natural phenomena (e.g., fall-out of pollutants). 	 NOx: 5-15 μg/m³, SO₂: 4-12 μg/m³ PM10: 10-20 μg/m³ 	(Trizio et al., 2016)

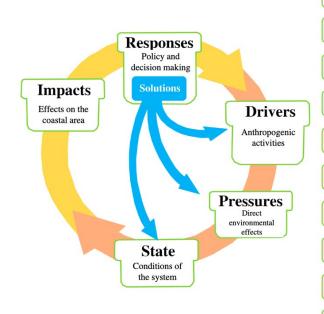
146The ISPRA (ISPRA, 2010) characterization was adopted at the basis of the responses 147undertaken by local authorities, which in summary were expressed in limiting the use of 148part of the Mar Piccolo areas. For instance, after a monitoring plan started in 2011 by the 149Italian Local Health Authority for the evaluation of contamination by PCDD/F and 150DL/PCB in Mussel *Mytilus galloprovincialis* of Mar Piccolo and Mar Grande, the Italian 151ordinance nr. 1989 of 22 July 2011 prohibited harvesting and handling mussels in the First 152Inlet. To date, the Mar Piccolo has been included among the Italian most contaminated site 153of national interest with the Italian Ministerial Decree in 2000 (Ministerial Decree 15410/01/2000, 2000) and is being studied by the Italian Government to choose the most 155suitable solution.

1602.2. Structure of the holistic approach (eDPSIR)

161The classic DPSIR framework considered a chain of causal links starting from "driving 162forces" which create "pressures". The latter determine changes in the "states" of the 163environment (physical, chemical and biological situation), which can "impact" on targets 164such as ecosystems and human health, eventually leading to political or technical 165"responses" (e.g., indicators, policies and regulations) (**Fig. 2a**) (Borja et al., 2006). 166Differently, the eDPSIR provides new relationships between the different elements of the 167DPSIR. Its final goal is to select the most suitable remediation technology according to the 168specific objectives and preferences of a particular stakeholder, and based on technical,

169environmental, economic, and social criteria.

170As the first step, all data and existing information needed to be identified and collected 171(**Fig. 2b**). The data must be reliable and as such comes from official sources (e.g., 172Environmental Protection Agency, universities,). Afterwards, three toolboxes are 173implemented, described in the following paragraphs. The three toolboxes act in synergy 174with the conventional DPSIR logic and follow the typical steps of a contaminated site 175remediation procedure (**Fig. 2c**). Several software packages are used, details of which can 176be found in section 2.3.



Collection and conceptual organization of available data

Recognition of industrial, anthropic, and natural driving forces and pressures

Identification of areas with environmental issues and distribution of the pollution

Evaluation of cause-effect relationships between pressures and states

Development of the Conceptual Site Model (CSM) for risk assessment

Calibration of the ERA tool (for risk assessment) using the dataset of pollutant concentration in sediments, water and mussels

Prediction of the bioconcentration in the target-organisms

Identification of homogeneous contamination scenarios

Selection of the remediation solutions for each homogeneous contamination scenarios by means of MCDA

Identification of the best remediation solutions for each homogeneous contamination scenarios

INPUT DATA concerning:

characterization of contaminated matrices; driving forces; pressures

OUTPUT: Identification of target contaminants and their concentrations in environmental matrices; comparison of concentrations with the Law limit values; delimitation of contaminated areas where the limit values for target contaminants have been exceeded; grouping of contaminated areas by target contaminant

INPUT DATA concerning: Identification of the contaminated areas and their target contaminants; CSM definition on the basis of available data; definition of the data required for the ecological risk analysis

OUTPUT: Knowledge of the ecological risk for each area previously delineated in toolbox 1, identification of the areas where contaminants may reach the trophic chain.

INPUT DATA concerning: Database of remediation technologies; Homogeneous contamination scenarios (organic + inorganic contamination; organic-only contamination; inorganic-only contamination); Risk magnitude.

OUTPUT (Remediation response):

c)

Selection of the best remediation technologies for each homogeneous contamination scenarios to be intended for field investigations

Toolbox 2

Toolbox 1

Toolbox 3

178 ^{a)}

21 22

Fig. 2. (a) Rationale, (b) framework of the holistic approach and (c) input/output data from each toolbox.

2.2.1 Toolbox 1

181Toolbox 1 aimed at assessing the current state of environmental matrices by using 182recognized biological, physical, and chemical indicators, and consequently correlating 183states with pressures. Three steps were necessary: (i) recognition of industrial, anthropic, 184and natural driving forces and pressures, (ii) identification of areas with environmental 185issues and distribution of the pollution, (iii) evaluation of cause-effect relationships 186between pressures and states.

187First of all, it is necessary to identify and collect all the industrial, anthropic, and natural 188driving forces and pressures through a review of academic and technical documents. The 189second step provides the spatial extent and magnitude of the contamination in a certain 190area, also identifying possible "hot-spots". On this point, the guideline reference is site-191specific. In the Italian case, it was the Italian Decree of the Ministry of Environment n. 192173/2016 (Italian Decree n. 173/2016, 2016), which defined criteria and methodological 193procedures for classifying dredging sediment. The quantitative Weight of Evidence (WOE) 194model is used to integrate and weight sediment chemical measures. The WOE model 195considers the number of parameters exceeding threshold limits, the magnitude of these 196exceedances and the type of contaminant (Priority or Priority Hazardous substances, 197according to the Annex II of the Directive 2008/105/EC). The threshold values considered 198for the calculation of the chemical Ratio-to-Reference (RTR) correspond to the site-199specific intervention values for marine sediments (ICRAM, 2004); in other cases, they may 200correspond with those defined by the local legislator. The Hazard Quotient for chemistry

201(HQ_C) included in the WOE model is calculated according to Eq. 1, where an average 202RTR_w is obtained for all of the parameters with RTR \leq 1 (i.e., below the normative limit), 203while the RTR_w is individually added into the summation Σ for those with RTR > 1 (Piva 204et al., 2011):

$$HQc = \frac{\sum_{f=1}^{N} RTR_{w}(j)_{RTR(f) \le 1}}{N} + \sum_{k=1}^{M} RTR_{w}(k)_{RTR(k) > 1}$$
(Eq.1)

207with N and M as the number of parameters with RTR respectively \leq or >1, while j and k 208are indices allowing to repeat the calculation for N or M times. The values of HQ_C are 209assigned to one of six classes of chemical hazard: absent/white <0.7; negligible/green 0.7–2101.3; slight/azure 1.3–2.6; moderate/yellow 2.6–6.5; major/red 6.5–13; severe/black >13. 211The chemical data from the ISPRA coring campaign in the period 2005-2010 (ISPRA, 2122010) were employed in this analysis. The pollutants considered were metals and organic 213compounds. The sample points are shown in **Fig. S1** in the SI and the coring depths were 2140-10 cm, 10-30 cm, 30-50 cm, 100-120 cm, and in some cases 180-200 cm, 280-300 cm. 215The first two steps made it possible to delineate the contaminated areas and identify the

216target contaminants, in line with the remediation procedure. The third step considers a 217multivariate statistical analysis useful to describe the large dataset, which aims to identify 218correlations among variables, grouped in clusters. Principal Component Analysis (PCA) 219and Cluster Analysis (CA) are used for this purpose, together with a correlation analysis, in 220order to find information about the existing site-clusters with similar pollution 221characteristics and to identify the most important discriminant contaminants (variables) 222within the same cluster. In our case, the statistical analysis was carried out on the total 223matrix consisting of 1023 samples 3 m deep and 20 variables, including moisture content, 224granulometry, metals, metalloids, and nutrients) (ISPRA, 2010). The adoption of the above 225statistical methods required to establish well-defined assumptions. In our case, it was 226hypothesized that the excessive presence of nutrients and organic matter in seawater could 227act as a carrier for many contaminants concentrated in the fine fraction of sediments with a 228high percentage of organic matter. Therefore, high concentrations of metals may be related 229to both organic matter and nutrients in the water column.

2.2.2 Toolbox 2

238Having clear the current state of environmental matrices, the Ecological Risk Assessment 239(ERA) is a useful tool to determine the adverse (ecological) effects occurring as a result of 240exposure of a biological community to potential stressors. At first, it is necessary to 241develop a Conceptual Site Model (CSM) with the goal of understanding how contaminants

242migrate from their sources and the pathways through which migration and exposure of 243potential human or environmental receptors can occur. Two steps are foreseen (i) 244calibration of the bioaccumulation model using a dataset of pollutant concentration in 245sediments, water, and mussels; (ii) prediction of the bioconcentration in the target-246organisms, using a full dataset of a previous coring campaign (ISPRA, 2010). In this study, 247the Mussel *Mytilus galloprovincialis* was considered as bio-target in the Arnot and Gobas 248model, being the most common species in the Mar Piccolo due to the intensive mussel 249farming activity. Eq. 2 shows the bioconcentration process (Arnot and Gobas, 2006):

$$\frac{dC_B}{dt} = (k_1 C_{WD}) - (k_2 + k_E + k_M + k_G)C_B$$
 (Eq. 2)

252where C_B is the chemical concentration in the organism (g/kg), t is a unit of time (1/d), k_1 253is the chemical uptake rate constant from the water at the respiratory surface (l/kg d), C_{WD} 254is the freely dissolved chemical concentration in the water (g/l), and k_2 , k_E , k_M , k_G are rate 255constants (1/d), representing chemical elimination from the organism via the respiratory 256surface, fecal egestion, metabolic biotransformation, and growth dilution, respectively.

257The data employed in the first step derived from the sampling campaign to characterize 258water, sediments and biota concerning the convention of 28/05/2013 between the Special 259Commissioner of the Italian Government and the Apulia Regional Environmental Agency. 260**Table S1** in the SI shows all the chemo-physical information employed in the first step, 261used as input data in the software in order to calibrate the model. The target contaminants 262investigated in this analysis were Polycyclic Aromatic Hydrocarbons (PAHs) although the 263same approach can be extended to any other type of pollutants.

2.2.3 Toolbox 3

266The last toolbox aimed at connecting the responses with the driving forces, pressures, 267impacts, and states with the final goal to identify the most suitable remediation 268intervention, in terms of applicability and social, political, and technological aspects. The 269openness to all these aspects was due to the fact that in socially relevant environmental 270problems, as in the case of the Mar Piccolo, the decision-maker has to find the best 271solution from a technical point of view, motivating the reason for the selection and mainly 272taking into account local economies.

273Based on the results of the first toolboxes, three different scenarios of contamination can be 274considered: SC1 with metals, PAHs and PCBs present in the same concentration (mixed 275contamination); SC2 with concentration of metals higher than PAHs and PCBs (prevalent 276inorganic contamination); SC3 with concentration of PAHs and PCBs higher than metals 277(prevalent organic contamination). For each contamination scenario, the framework for

278selecting the technological remedial alternatives consisted of two phases: (i) Multi-Criteria 279Decision Analysis (MCDA), aimed at evaluating the three most suitable technologies; (ii) 280application of a coaxial matrix for each technology recognized in the previous step, with 281the goal of identifying the best technology.

289The MCDA (OECD-JRC, 2008) offered a scientifically sound decision framework and 290entailed the following steps: (i) preliminary screening of potential remedial technologies 291(so-called alternatives); (ii) identification of the selected alternatives and evaluation criteria 292(qualitatively and quantitatively based); (iii) construction and normalization of the 293alternative matrix; (iv) construction of the composite indicator (which was indicated as 294Preference Index - PI); (v) identification of the optimum alternatives.

295The initial panel of remediation technologies was identified by a technical working group 296(TWG) through a critical analysis of technical and scientific reports (ACE, 2014; FRTR, 2972002; Hasegawa and Rahman, 2016; SMOCS, 2012; UNIDO, 2007) (Fig. S2). The criteria 298used to evaluate the technologies were classified in applicability and technological aspects 299(Table S2). The criteria were defined on the basis of current technical-scientific literature 300and were all independent of each other; more information on the grade assignable to each 301criterion is shown in Table S3 of Appendix. Clearly not all criteria weighed the same; 302according to Sabia et al. (2016), the weighing of the criteria requires the definition of the 303priority order. The priorities adopted by the TWG are shown in Table S2. Then, each 304criterion was given a weight, according to the relation with all other criteria, and 305qualitative criteria were transformed into quantitative for better homogenization and

306comparison. The Paired Comparison Technique (PCT) (Goletsis et al., 2003) was used for 307a pairwise comparison. At this point, the Matrix of Alternatives (MOA) was drawn up with 308the potential technologies in the rows and the evaluation criteria in the columns. A 309normalization of the matrix was subsequently carried out according to the max/min method 310(Eqs. 3 and 4):

$$x_{ij}^- = x_{ij} / \text{Max}(x_j) \tag{Eq. 3}$$

$$x_{ij}^- = Min(x_j)/x_{ij}$$
 (Eq. 4)

313where, x_{ij} is the performance of the alternative i-th (each remediation technology) with 314respect to the j-th indicator (each evaluation criterion), w_j the weight of each indicator, x_{ij}^{-} 315the normalized value of x_{ij} calculated with Eqs. 3 and 4 if the criterion is to be maximised 316or to be minimised, respectively. As result, all the values of the normalized matrix were in 317the range [0, 1].

318Through the Simple Additive Weighing (SAW) method (Afshari et al., 2010), the MOA 319was resolved, multiplying each element of the matrix with the weight of the corresponding

320column. The PI of the single alternative was calculated following Eq. 5:

$$PI_{i} = \sum_{j=1}^{m} x_{ij}^{-} \cdot w_{j}$$
 (Eq. 5)

323where ^Wj represents the weight for each evaluation criteria.

324At the end, the best technological alternatives were identified with the highest PI, 325according to the scenario considered, and distinguishing between *in-situ* and *ex-situ* 326alternatives. Lastly, coaxial matrices (Ruppert et al., 2015) put in relation and in a logical 327sequence, respectively: (i) project actions, (ii) impact factors, (iii) environmental 328components, and (iv) potential environmental and human risks. Drawing up different 329evaluation analysis sheets, it was necessary to quantitatively assess all possible impacts of 330each remediation project action using the range [-4, +4], with -4 for the worst system 331alteration and +4 for the best one. Summing all the impact values and converting them into 332a scale of 100, it was possible to quantify and compare all the impacts of every single 333remediation project.

3352.3. Software used in the proposed methodological framework

336The Arnot and Gobas model (Arnot and Gobas, 2004) was considered; through the 337Aquaweb software, site-specific evaluations of bioconcentration/bioaccumulation factors 338in organisms of aquatic food webs from chemical concentrations in water and sediments 339(available at https://arnotresearch.com/aquaweb/), were conducted. The QGis software was 340used for data visualization (available at 341https://www.qgis.org/it/site/forusers/download.html). Lastly, three different types of 342software were used to conduct multivariate analyses: The Unscrumbler software and 343SIMCA for the Principal Component Analyses (PCA) and Statistica10 for the Cluster 344Analyses (CA). The knowledge of these specialist tools was fundamental and preparatory 345to the understanding of the three developed toolboxes.

346

347**3. Results**

3483.1. A complex environment: organization of available data

349Before presenting the results of the 3 toolboxes, it was essential to organize the massive 350amount of available data. Researchers investigated both the economic aspects, linked to the 351activity of shellfish farming, and the hydrogeological aspects, and showed the complexity 352of this coastal system. Numerous studies concerning multiple aspects have been carried 353out; the Mar Piccolo is undoubtedly a complex system, an accurate schematization of it is 354surely the most important starting point towards remediation (**Fig. 3**).

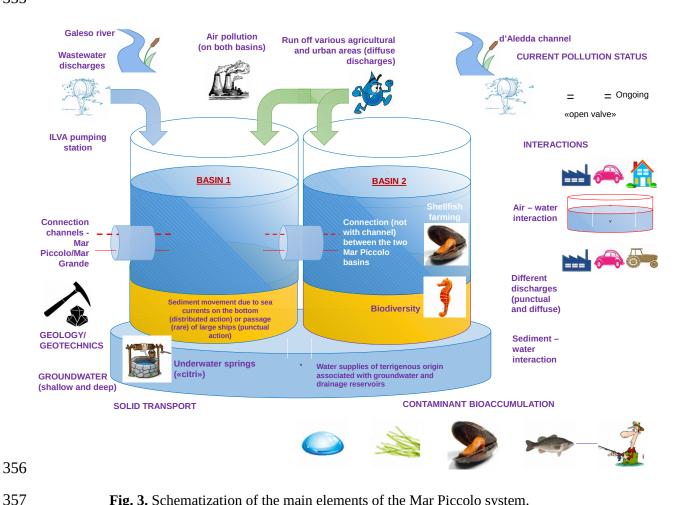


Fig. 3. Schematization of the main elements of the Mar Piccolo system.

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359The two basins are immersed in saturated soil and at some spots in contact with the water 360table. The two basins are connected to each other and to the open sea, the Mar Grande. 361There are numerous pressures from point discharges of wastewater and watercourses (the 362Galeso River and the D'Aiedda channel) to the surface run-off as a result of rain. There is 363also the presence of highly impacting activities, such as shipyards and the military arsenal. 364Several organic/inorganic pollutants are present in sediments (**Table 1**); interstitial and free 365waters are instead affected by wastewater discharges, characterized by non-negligible 366concentrations of organic substances (Biochemical Oxygen Demand, Chemical Oxygen 367Demand) and nutrients, and other contaminants such as PAHs and PCBs. **Table 2** 368summarizes most of the information available to date.

Table 2. Milestones achieved on the basis of current literature.

Symbol ^(a)	Aspect	Main considerations	References
()	Movement of sea currents in Mar Piccolo – Mar Grande connection channels	On average the current is directed towards the Mar Piccolo (entering) in the deeper layers, while in the more superficial layers the current is directed towards the Mar Grande (outgoing).	De Pascalis et al. (2015); Armenio et al. (2016); De Serio e Mossa (2016)



Interaction
between
sediment and
bottom sea
currents (erosion
and
accumulation
phenomena).

The values of the maximum tangential load (responsible for erosion) at the bottom of the seabed are mainly located in the Mar Piccolo - Mar Grande connection channels (Canale Navigabile, Canale Porta Napoli) and at the ILVA water pumping station (which brings water to the industrial area). A tendency towards sediment deposition has been observed in the remaining parts of the Basin I and in the Basin II.

De Serio et al. (2018)



Wastewater discharges

Mar Piccolo is subject to both private and public discharges. A total amount of 100,000 equivalent inhabitants is estimated to be discharged into Mar Piccolo. 85% of the inlet flow rate is discharged into the Basin II, with an amount of organic matter equal to 6.7 t/d of BOD_5 . The daily release of nitrogen and phosphorus into the basin has been quantified as 17.2 t/d and 0.3 t/d, respectively.

Cardellicchio et al. (1991); Caroppo et al. (1995); Scroccaro et al. (2004)



Run off of agricultural and urban areas in the Mar Piccolo catchment area (diffuse discharges) The hydrographic grid coincides with the Galeso basin and its main tributaries with 60 km² surface. It also includes the industrial area of Taranto. The basin is subject to flooding and shows an erosive character, resulting in solid transport from the Galeso river basin.

Blonda et al. (2014).



Geology and geotechnics

Geology: Holocene deposits between 0.5 m and 11 m; Recent alluvial deposits between 1 m and 20 m; Terraced marine deposits between 5 m and 30 m; Sub-Apennine clays from 10 m to 65 m; Gravina limestones with thickness less than 2-3 m; Mesozoic limestones from -20 m to -80 m depth, decreasing towards the Gulf of Taranto.

Geotechnics: Clay percentage increases with the depth (from 11 to 12 m of seabed) and reduces that of sand, while the silt is maintained constant; Bioclasts (with dimensions greater than 1 mm) are an element of non-homogeneity of the clayey-lime matrix, impacting on the regularity of the mechanical characteristics. The natural water content and plasticity index show a "fluid" behaviour of the material depending on chemical characteristics of the interstitial fluid (mix of sea water, organic substances, contaminants).

Blonda et al. (2014); Vitone et al. (2016); Valenzano et al. (2018); Sollecito et al. (2019)



Biodiversity

The Mar Piccolo is a "transition" environment (WFD, 2000/60 EC). There is a peculiar vegetal and animal biodiversity, mainly composed of nitrophilic macroalgae and filter organisms. The high biodiversity found (in number and species) together with the high self-regulating and reactive capacity of the system, are indicators of the great potential for recovery of the area from a naturalistic and environmental point of view. There are also protected species.

Prato and Biandolino (2005); Blonda et al. (2014)



Water supplies of terrigenous origin associated with groundwater and drainage reservoirs The deep aquifer located in the Altamura limestones drains the water coming from an upstream hydrological basin of 900 km² and undergoes a rapid decrease in piezometric heights (piezometric cadence of 2‰) causing the formation of terrestrial (Galeso, Lavandaia, Marangio, Battentieri, and Riso) and underwater springs (among the best known are the citri Galeso and Citrello in Basin I, Le Copre, Calandrea, and Mascione in Basin II). Limited knowledge is available about surface aquifers. No information is available on the interconnections between deep and superficial aquifer. This situation is made even more complex by the consistent intrusion of seawater.

Blonda et al. (2014).

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371The complexity described above makes it possible to evaluate how remediation should take 372place and how to cope with the existing sources of contamination, either by removing them 373or by regulating them. Consequently, at the same time as remediation, the decision-maker 374decides to act on the driving forces and pressures involved.

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3763.2. Towards remediation

3773.2.1 Toolbox 1: state of the environment and role of discharges in sediment pollution 378The literature on the Mar Piccolo identified the driving forces as well as the pressures. 379With 195,279 inhabitants in 2019, Taranto is the most populous city in the Mar Piccolo 380catchment area. Different types of agricultural areas fall into the Mar Piccolo catchment 381basin (vineyards, olive groves, and arable crops are the most abundant), covering the 55% 382of the total agricultural area of the basin. ISTAT data revealed an increase of agricultural 383fertilizers employed between 2003 and 2008 and phytosanitary products between 2013 and 3842016 (ISTAT, 2018) and, assuming run-off as one of the main impacts, pollutants and 385nutrients were brought into the Mar Piccolo. The industrial area on the west side is

386responsible for high impacts in terms of pollutant production and distribution (steel factory, 387oil refinery, and cement factory). Severe pressures can be attributed to the presence of the 388Italian Navy shipyard with its dry docks on the south side of the first bay as well as to 389intensive mussel farm production (with 30,000 tons/y) and the fishing-boat activity in both 390bays. Nine sewage discharge pipes and small rivers together with D'Aiedda channel 391drained nutrients into the basin (Cardellicchio et al., 2007). The D'Aiedda channel 392discharges the treated wastewaters of 8 municipalities into the second bay (Scroccaro et al., 3932004). About 18,272 m³/d are released into the Mar Piccolo, the 85% of which into the 394second bay. The demography, agriculture, and industry were considered as the main 395driving forces. The corresponding pressures were the discharges of nutrients and 396contaminants, pollution in groundwater and in air. The state of the environment, on the 397other hand, was characterized mainly as marine sediment, seawater, and biodiversity; the 398latter was not presented in this study. The calculation of the HQc index allowed visualizing 399the map of chemical hazard for the marine sediments in a GIS environment at different 400depths from the sea floor (Fig. 4a-f).

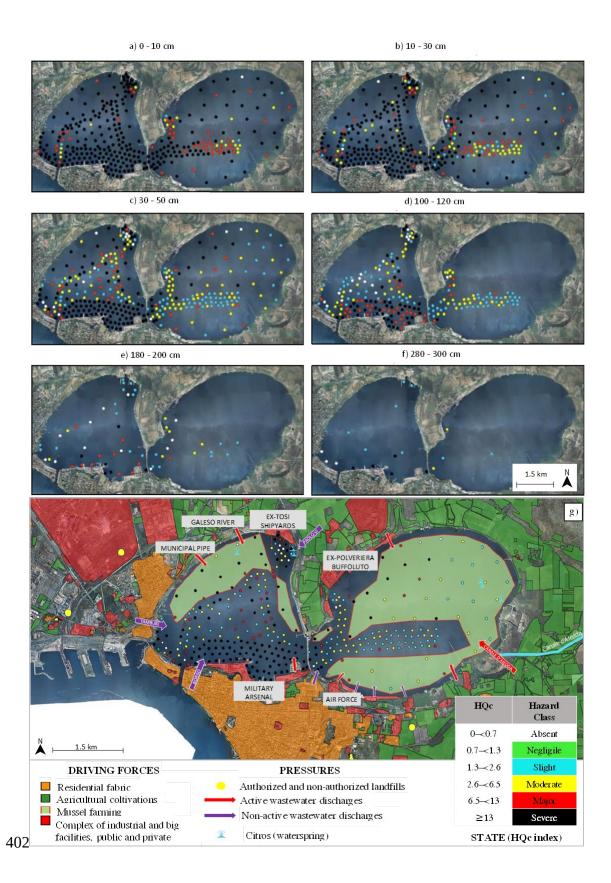


Fig. 4. HQc chromatic maps showing sediment chemical hazard at different depth (a) 0-10 cm, (b) 10-30 cm, (c) 30-50 cm, (d) 100-120 cm, (e) 180-200 cm, (f) 280-300 cm; (g) D-P-S map of toolbox 1.

407It was possible to observe that the first layers of the sea bottom were the most polluted (0-40810 cm, 10-30 cm, 30-50 cm), showing a spread contamination in both inlets. The area of 409the Marine Arsenal appeared to be the most critical zone, since even deep layers (100-120 410cm, 180-200 cm, 280-300 cm) recorded high values of the HQc index. The two inlets 411presented a macroscopic difference, with more serious, widespread, and deep 412contamination in the First Bay.

413The approach adopted in Toolbox 1 allowed understanding the quantitative role of each 414contaminant in the definition of the HQc index. For this purpose, histograms were 415elaborated for each sub-area visible in **Figs. S3-S26** in the SI. The interpretation of the 416histograms made it possible to identify various results, summarized in **Table S4**.

417The map in **Fig. 4g**, which represents an average of the layers 0-100 cm, visually links 418main pressures with environmental status. The histograms for each sub-area (Figs. S3-S26) 419revealed the main pressures that induced that contamination. Lastly, the multivariate 420analysis allowed identifying possible cause-effect relationships between pressures and 421states. Considering the correlations between nutrients and metals (**Figs. S27-S28**), results 422showed how the excessive presence of nutrients and organic matter in the marine water 423could act as carrier for many contaminants in the fine fraction of sediments, the latter 424characterized by high percentage of organic matter. It was possible to identify two macro 425clusters: (i) TOC (Total Organic Carbon), As, Cd, Zn, Hg, Pb, Cu, clay and gravel; (ii) P_{tot},

 $426N_{tot}$, Cr, Fe, Al, V and Ni. While in the first cluster TOC was considered as the responsible 427correlating element, in the second cluster the responsible ones were the nutrients (N_{tot} and $428P_{tot}$). Furthermore, high concentrations of Cr, V, and Ni showed high correlation, 429underlining their apparently natural presence in surficial layers.

430These findings through statistical procedures confirmed the validity of the assumptions 431made in Toolbox 1, highlighting the role of discharges on sediment pollution. Mali et al. 432(2017) corroborated our results and underlined that the nature and origin of organic matter 433within different layers, the rates of dilution by terrigenous sediments, the grain size 434fractions, the aggregation phenomena, and the original composition of the sediment parent 435rocks can influence the contaminant pattern as the main controlling factors.

436

4373.2.2 Toolbox 2: quantifying bioaccumulation

438Toolbox 2 quantified the impact on humans based on the conceptual site model in Fig. 3.

439

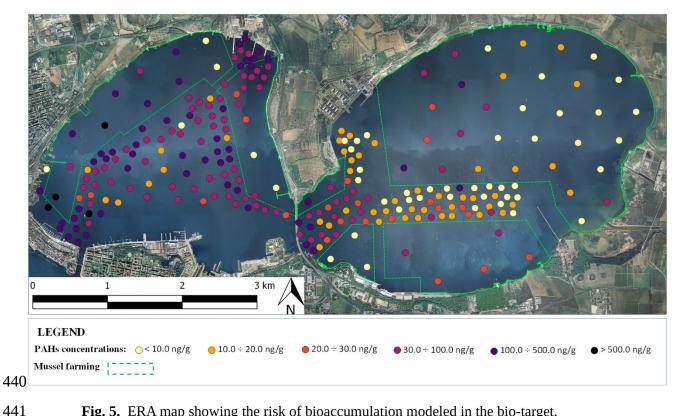


Fig. 5. ERA map showing the risk of bioaccumulation modeled in the bio-target.

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443The CSM showed how the contamination of the seawater was attributed to urban/industrial 444discharges/deposition as well as the runoff of the surrounding soil. There was a supply of 445spring water from the bottom of the Mar Piccolo, while aeriform pollutants could pass 446from air to water. Contamination could reach human via trophic chain. The quantification 447of such impacts on human included the selection of the target contaminant; the analysis 448was conducted considering only PAHs, although the same could be replicated in the case of 449metals and PCBs, which were abundant in the study areas. All the site-specific data input, 450together with the PAH contamination values in sediments and water, were entered in the 451software Aquaweb. A good fitting of the calibration process ($R^2 = 0.87$) between the PAH 452concentration in the bio-target (mussel) sampled and modeled was achieved as visible from

453**Fig. S29**.

454This approach was subsequently extended to the entire case study and, starting from the 455concentration values in sediments and water, it was possible to predict the bio-456concentration in mussels present in both inlets. The results showed how the areas used for 457the mussel farming recorded high modeled values of contamination in the bio-target (**Fig.** 4585), highlighting a hazardous threat to the trophic chain. According to the Commission 459Regulation (EU) No 835/2011 of 19 August 2011, amending Regulation (EC) No 4601881/2006, as regards maximum levels for PAHs in foodstuffs (sum of benzo(a)pyrene, 461benzo(a)anthracene, benzo(b)fluoranthene and chrysene), the maximum concentration 462acceptable in mussels is 30 ng/g. Peaks higher than 100 ng/g were recorded in the First 463Inlet especially on the west shore and in the central area. As an example, in 2011 mussel 464farming in the First Inlet was banned following detection of PCBs and dioxins in the 465mussels for human consumption (ASL TA, 2011).

466Toolbox 2 has, therefore, quantified the hazard related to the contamination of 467environmental matrices by PAHs. As reported in Tongo et al. (2017), chronic intake of 468toxic organic compounds has adverse impacts on human and an excess of threshold values 469indicates potential health risk from consumption.

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4713.2.3 Toolbox 3: selection of technological remediation actions

474Before presenting the results of Toolbox 3, it is interesting to note that the current 475responses adopted by local authorities included the limitations on the use of marine areas 476and at the same time regularizing the pressures on the environment. The results shown

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477below are valid if and only if pressures are eliminated and contained. The resolution of the 478matrix of alternatives (MOAs) was carried out for each scenario of contamination. Figs. 479**S30-32** showed the resolution of SC1, SC2 and SC3, respectively. The highest PI score, 480and consequently, the best technology identified was Amendments, for all the three 481scenarios investigated (Table S5). Amendments are represented by any material (e.g., 482activated carbon, organoclay, apatites, etc.) that may interact with the contaminated 483sediment/soil enhancing the pollutant sequestration and degradation. In the scenario SC1, 484where the contamination was given by metals, PAHs, and PCBs simultaneously, the PI 485index pointed out ex-situ solution (Vitrification); in the scenarios SC2 and SC3, the PI 486index highlighted the Assisted Natural Attenuation (ANA) as *in-situ* solution. The first five 487 positions for the *in-situ* technologies, and in all the scenarios evaluated, were: (1) 488Amendments, (2) Assisted Natural Attenuation, (3) Monitored Natural Attenuation, (4) 489Containment barriers, and (5) Passive capping. For the *ex-situ* technologies, vitrification 490appeared to be the most promising alternative in all scenarios. Solvent extraction and 491thermal desorption were the second or third position depending on the scenario, while 492mechanochemistry and confined disposal facility reached the fourth and fifth position, 493respectively, in all the three scenarios considered.

494It is noteworthy that the excavation and disposal option, together with the application of 495nanoremediation technologies, represented the last choices to undertake. Although some 496positive outcomes are present in the literature (Xue et al., 2018), nanoremediation requires 497a robust preliminary field-scale study to investigate the treatment feasibility (Lofrano et al.,

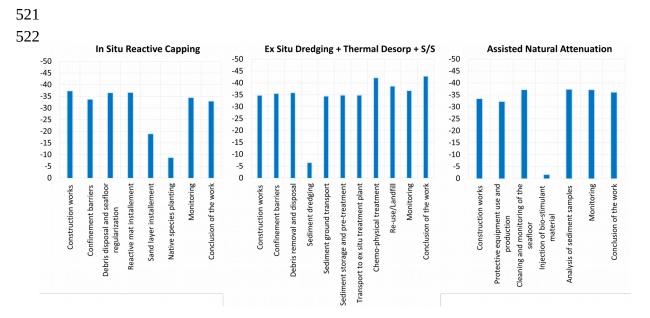
4982017).

499The adopted multi-criterial approach has allowed integrating a diversity of criteria in a 500multidimensional way and in a large variety of contexts.

501The second phase of Toolbox 3 consisted of the development of three coaxial matrices for 502the best treatments identified through the MCDA analysis. The best three technologies 503identified through the multicriteria approach were: (i) reactive capping, as an *in-situ* 504treatment with the use of amendments, (ii) a train-treatment based on dredging plus 505thermal desorption plus stabilization/solidification (S/S), as *ex-situ* approach, and (iii) 506assisted natural attenuation. To achieve this, it was necessary to define the project actions, 507which vary for each technology, the impact factors, the environmental components, and the 508relative potential environmental and human health risks. The coaxial matrices (**Figs. 33S**-5093**5S**), together with the evaluation matrices, allowed identifying and evaluating 510quantitatively the activities responsible of environmental disturbance for each project 511action. A quantitative evaluation sheet was developed for each project action and 512subsequent impacts and environmental components and alterations. An example of an 513evaluation matrix is shown in **Table S6** in the SI.

514All the scores were summed and converted in a scale of 100 for comparison. The activities 515with the highest negative scores would take the most impactful actions, and vice versa. The 516sum of the total impacts for each project action proved to be a useful global information on 517the remediation treatment itself, easily comparable with the other technologies. The results 518identified Assisted Natural Attenuation as the best solution in terms of environmental 519impact and compatibility (**Fig. 6**). ANA recorded a value of impacts equal to 214, whereas

520reactive capping and train treatment reached 238 and 375, respectively.



523**Fig. 6.** Comparison of final scores from the coaxial and evaluation matrices for the three treatments considered.

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526This systematic approach allowed identifying and quantifying the total impacts related to 527remediation treatments, which permitted reducing contamination in environmental 528matrices and still involved impacts and repercussions. Once the most suitable remediation 529technologies have been identified, they can be applied to the contaminated areas identified 530with Toolbox 1 without neglecting the pressure that should be contained.

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5324. Discussion

533This study deals with what would have been done without eDPSIR and what is the added 534value of implementing eDPSIR. The DPSIR was characterized in a broader sense by 535incorporating plans and programs with the intent to place restrictions on the use of the site

536and/or containing the pressures. The eDPSIR, instead, guided the decision-maker in the 537selection of remediation technologies.

538Concerning the correlation between D-P-S, most of previous studies applied the DPSIR 539framework qualitatively, with a non-depth, statistically based assessment of the roles of P 540on S (Lofrano et al., 2015; Skoulikidis, 2009; Zebardast et al., 2015; Zhang and Xue, 5412013). Other authors used a set of key indicators, such as Environmental Assessment 542Criteria (EAC) (Cinnirella et al., 2013), Ecological Effect Index (EEI) (Lu et al., 2019), 543and those within the Strategic Environmental Assessment (SEA) (Cheng et al., 2011), but 544the investigation of cause-effect relationships was still missing. In contrast, starting from 545the site-specific sources of pollution, Toolbox 1 in this study clarified the state of 546contamination in marine sediments, with specific correlations with pressures (see the 547histograms of Figs. S3-S26). The multivariate analysis has proven to be a useful tool to 548assess the correlation between contaminants in water and sediment (Mali et al., 2017). 549However, it was essential to make science-based hypotheses, which then needed to be 550verified. This toolbox revealed the target contaminants and the extension of the 551contamination, in line with a remediation procedure. Other assumptions can be made using 552the same method.

553With reference to the S-I link, the literature focused mainly on the definition and then 554application of key indicators to quantify impacts on the environment and on human 555(Barjoveanu et al., 2010). However, what makes the impact assessment accurate is the 556CSM. Toolbox 2 aimed to standardize this phase. As for toolbox 1, it was necessary to 557define the hypothesis to be subsequently verified: the presence of bioaccumulation 558phenomena of pollutants, moving them from sediment and water to mussels, creates

559serious risks for human consumers. Also, toolbox 2 allowed highlighting the areas where 560contaminants may reach the trophic chain.

561With reference to closing the cycle through D-P-S-I-R linkage analysis, Toolbox 3 562provided new *modus operandi* on how to select the optimal remediation technologies. First 563of all, it is important to be aware that a complex environmental problem is being 564addressed; this is not always clear to decision-makers. Such a complex problem must 565include a multi-disciplinary and participatory approach, which can be extended to all 566stakeholders in the area. Stakeholders participation is essential to make the whole decision-567making process traceable first and then to take into account the needs of the area (De Feo 568and De Gisi, 2010). A remediation intervention can be seen as a new development for local 569economic activities and aspects to be considered among evaluation criteria. While the use 570of multi-criteria analysis to address complex environmental problems is well established, 571the elements of MCDA needed to be set up. The current references (ACE, 2014; FRTR, 5722002; Hasegawa and Rahman, 2016; SMOCS, 2012; UNIDO, 2007) provided only 573preliminary information for defining the initial panel of remediation technologies. The 574definition of the assessment criteria and their priorities, together with the weighting vector, 575had to take into account multiple aspects, ranging from the technological ones related to 576remediation technologies to the social ones.

577The eDPSIR goes in the direction outlined by Lewison et al. (2016) that highlights the 578presence of qualitative relationships, limited interdisciplinarity, the use of DPSIR as a 579cognitive tool, the need to develop decision support tools, as well as the need to include 580local stakeholders in the decision-making process. The eDPSIR could allow overcoming

581these bottlenecks. The last aspect to highlight is the eDPSIR generalization. **Table 3** shows 582the synoptic framework enabling the eDPSIR holistic approach to be extended to other 583territorial contexts.

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Table 3. Starting conditions summary for the eDPSIR application.

Starting conditions

Type of data to be determined for the "Driving Forces" and their source:

It is necessary to have information about all the possible activities/phenomena that led to pollution.

Type of data to be determined for the "Pressures":

Starting from the driving forces, the related produced pressures are identified (emissions, waste, discharges, etc.) in quantitative terms.

Type of data to be determined for the "States":

Having clear sources and impacts of pollution, it is fundamental to understand the states of the environmental compartments in order to recognize which are the areas most impacted (concentrations of pollutants in sediments, water, and air). In order to make this step more reliable, homogeneous data should be used (e.g., data on chemical characterisation of sediments on samples taken on the same day and with the same sampling method).

Type of data to be determined for the "Impacts":

The aim in this case is to diagnose the phenomena involved from contaminated matrices on targets such as ecosystems and humans (through risk assessment

analyses). Impact assessment methodologies should be selected on the basis of data availability.

Type of data to be determined for the "Responses":

The responses can be programmatic and technological. In this study, emphasis was placed on both. Technological responses include remediation; programmatic responses relate to the need to control/limit ongoing sources of contamination.

Software/tools to be used:

Aquaweb, QGis, Simca, Unscrumbler, Statistica10

Other methodological tools

WOE criteria, Arnot and Gobas model, coaxial matrices, multi-criteria analysis

References for remediation technologies

See the Manuscript, sections 2.3.3 and 3.2.3, and the Appendix (Supplementary material).

Community involvement

It is allowed, directly in the phase of weighing the criteria for the assessment of remediation technologies.

586

587**5. Conclusions**

588Based on the findings of the Mar Piccolo of Taranto case study, the eDPSIR has proven to 589be a valuable tool to support decision makers in choosing the technological response for 590the remediation of heavily contaminated coastal areas. The introduction of the three

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591toolboxes, each with its own specific features, made it possible to investigate and quantify 592cause-effect relationships. Considering the toolbox 1, "state of the environment and role of 593*discharges in sediment pollution*", the application of the Hazard Quotient-Chemical index 594on a large data set allowed identifying the extent and magnitude of contamination for target 595organic (PAHs and PCBs) and inorganic contaminants. The <u>multivariate statistical analysis</u> 596Principal Component Analysis proved to be a valuable support to the understanding of the 597dynamics according to which the excessive presence of nutrients and organic matter in 598seawater could act as a carrier for many contaminants concentrated in the fine fraction of 599sediments with a high percentage of organic matter, providing useful information to the 600decision-maker in formulating programmatic responses, such as containing/controlling 601pressures. Considering Toolbox 2, "quantifying bioaccumulation", it made it possible to 602quantify the hazard of contaminated marine sediments and highlighted how contaminated 603sediment and water can pose a serious threat to humans and wildlife, which may come into 604contact and absorb toxic and carcinogenic compounds through the food chain. Toolbox 2 605allowed visualizing the areas where contaminants may reach the trophic chain. The most 606site-specific aspect in Toolbox 2 was the definition of the conceptual model linking 607contamination sources with migration paths and natural and human targets. As to Toolbox 6083, "selection of technological remediation actions", the application of a MCDA first and 609coaxial matrices then allowed identifying the Assisted Natural Attenuation as one of the 610best solutions starting from a panel of 50 remediation technologies. The MCDA was 611characterized by transparency and replicability, which are fundamental characteristics in

612the case of complex environmental and social problems. As a final consideration, the three 613toolboxes have proven to be an added value to existing literature. They have made the 614DPSIR as an environmental decision support system rather than a cognitive tool of the 615environmental phenomena in a system. Since the proposed toolboxes are hypothesis-based, 616the eDPSIR presents itself as a tool open to new formulations to be verified.

617

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625Credit author statement

626Claudia Labianca: Data curation, Formal analysis, Investigation, Software, Validation, 627Visualization, Roles/Writing - original draft; Sabino De Gisi: Conceptualization, Formal 628analysis, Methodology, Resources, Supervision, Roles/Writing - Original Draft, Writing - 629review & editing; Francesco Todaro: Data curation, Investigation, Software, 630Visualization, Writing - review & editing; Lei Wang: Visualization, Validation, Writing - 631review & editing; Daniel C.W. Tsang: Resources, Supervision, Validation, Writing - 632review & editing; Michele Notarnicola: Conceptualization, Funding acquisition, Project

633administration, Resources, Supervision, Writing - Review & Editing.
634

635**Declaration of competing interest**

636The authors have no conflicts of interest to declare.

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638Appendix A. Supplementary data

639The following is the Supplementary data to this article:

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