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Assessing environmental pollution levels in marinas

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Highlights:

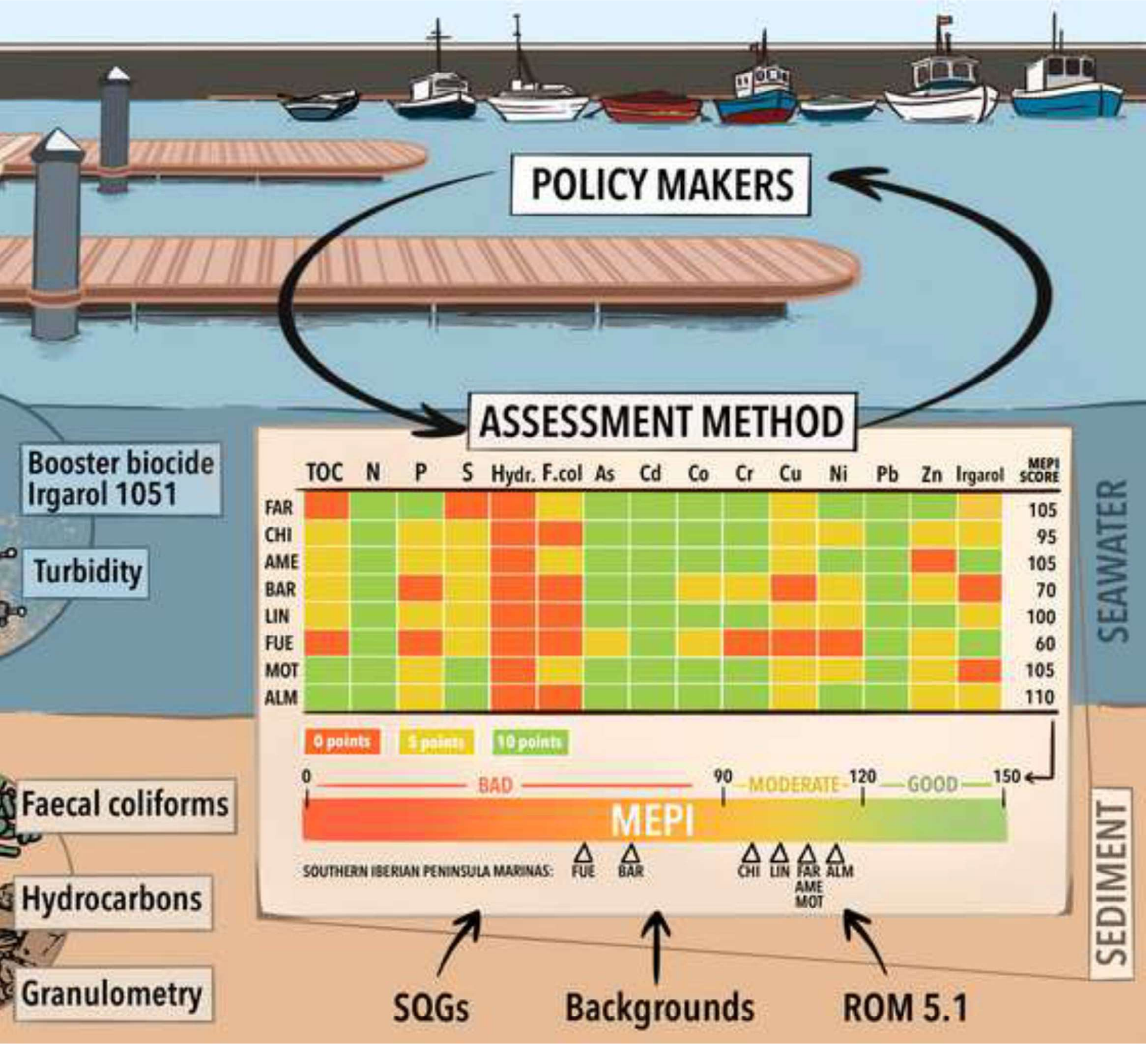
-Environmental pollution of marinas was reviewed and empirically assessed.

-Turbidity, Irgarol, hydrocarbons and faecal coliforms were identified as key stressors.

-A multi-stressor quality assessment based on international guidelines was developed.

-The method was tested in southern Iberian Peninsula marinas

-Marinas ranged from bad to moderated polluted according to the proposed MEPI index


POLICY MAKERS
ASSESSMENT METHOD
Booster biocide Irgarol 1051
Turbidity
Faecal coliforms
Hydrocarbons
Granulometry
TOC N P S Hydr. F.col As Cd Co Cr Cu Ni Pb Zn Irgarol MEPI SCORE
FAR 105
CHI 95
AME 105
BAR 70
LIN 100
FUE 60
MOT 105
ALM 110
0 points
5 points
10 points
0 BAD 90 MODERATE 120 GOOD 150
MEPI
SOUTHERN IBERIAN PENINSULA MARINAS: FUE BAR CHI LIN FAR AME MOT ALM
SEAWATER
SEDIMENT
SQGs
Backgrounds
ROM 5.1

***Highlights (for review : 3 to 5 bullet points (maximum 85 characters including spaces per bullet point)**

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ABSTRACT

Despite the growing interest in recreational boating and the increasing number of marinas along the world coastlines, environmental knowledge of these ecosystems is still very scarce. Detailed data of pollutans in marinas are necessary to provide a global approach of environmental risks in the context of international management strategies. In the present study, a set of 64 variables (30 in seawater and 34 in sediments) were measured to compare marinas from Southern Iberian Peninsula (SIP). Uni and multivariate analyses showed significant differences among marinas, evidencing the importance of management at local scale. The most relevant variables determining these differences were turbidity and the booster biocide Irgarol 1051 in seawater, and granulometry, hydrocarbons and faecal coliforms in sediment. The use of normalization techniques with Al or Fe, and the suitability of different methodologies to measure Total Organic Matter (TOM) in marinas were also discussed. Additionally, we perform a comprehensive literature review of worldwide marina stressors and develop a simple and straithtforward method for assessing environmental quality. The method was tested using SIP marinas and was based on the comparison of 15 selected sediment stressors with background values, concentrations of worldwide sediment quality guidelines (SQGs), and reference conditions/security thresholds established by the programme of coastal waters in port areas (ROM 5.1). A global score was assigned using a new proposed index, Marinas Environmental Pollution Index (MEPI), ranging from 0-150 points according to the environmental quality (<90:bad, 90-120: moderate, >120: good). MEPI of marinas from SIP ranged from 60 to 110 points indicating bad or moderate levels of pollution. Environmental quality is one of the decisive factors for awarding eco-labels or eco-certifications, such as Blue Flags in marinas. Therefore, pollution baseline information and enviromental tools are mandatory for correct assignation of these awards and necessary for assessing the efficiency of the management actions.

Keywords: marinas, recreational boating, environmental pollution, quality assessment, sediment, MEPI index, Irgarol

1. Introduction

The number, distribution and economical importance of marinas are significantly increasing due to coastal tourism demand (Moreno et al., 2009). Although marinas provide substantial economic incomes, boating activities (and associated infrastructure) can degrade the environment and its associated ecosystem services, introducing a range of environmental problems, such as emission of air pollutants, noise, sediment dredging, wastewater discharges, leaks of petroleum derivates (such as hydrocarbons), and accumulation of heavy metals and biocides from antifouling paints (McGee et al., 1995; Konstantinou and Albanis, 2004; Mali et al., 2017; Toh et al., 2017; Chatzinikolaou et al., 2018). Excess of nutrients and a variety of chemical pollutants originated from sewage discharge, industrial activities, agricultural leaching and urban runoff, can also accumulate within marinas (Kenworthy et al., 2018 and references therein). Water renewal in marinas is hindered by fixed structures (sea walls, narrow outlets, floating pontoons, etc) which limit circulation. Thus, marinas represent semi-enclosed environments characterized by reduced hydrodynamic energy, high sedimentation rates and lowered oxygen levels (Rivero et al., 2013; Ng et al., 2019), where seawater and sediment pollution is increased by water retention (Kenworthy et al., 2018). Moreover, marinas have suitable conditions for the establishment and spread of fouling organisms, and are consideredas important hotspots for Non Indigenous Species (NIS) (Martínez-Laiz et al., 2019; Ramalhosa et al., 2019). Therefore, environmental quality assessment of marinas is becoming a priority issue since they behave as sinks for physical, chemical and biological pollutants. However, more efforts are still needed to raise awareness on the impacts of marinas in coastal environments and to provide the tools to better manage them (Burgin and Hardiman 2011).

Ports and marinas management is challenging, as it involves conflicting uses by residents, visitors, boat owners, shipping, industry and other users, thus requiring the integration of multi-disciplinary authorities and stakeholders from different sectors (e.g. engineers, ecologists, economists, governmental bodies) (Chatzinikolaou et al., 2018). Larger ports have access to a range of environmental management tools; however, these management strategies are rare in smaller ports or marinas (Parra et al., 2018). In the context of Water Framework Directive (WFD) and the Marine Strategy Framework Directive (MSFD), marinas can be considered as heavily modified water bodies (HMWB).These are bodies of surface water that, as a result of alterations by human activity, are substantially changed in characters and cannot meet the good ecological status (see Gupta et al., 2005; Petrosillo et al., 2010; Ondiviela et al., 2013). Navigation and recreational boating are among the main activities considered likely to result in a water body being designated as HMWB, but many others (such as water regulation, flood protection or land drainage, among others) are also included (Borja and Elliot, 2007). HMWBs are, then, heterogeneous groups which encompass diverse ecosystems requiring different management. Therefore, the wide variety of HMWBs makes it difficult to assess general regulations for all of them.

During the last years, numerous approaches have dealt with the assessment of the ecological status of natural water bodies, but only a few deal with HMWBs. Indeed, the wide range of pressures on ports and marinas prevents the assessment of some methods that are effective for natural water bodies (Ondiviela et al., 2012, 2013 and references therein). One of the few initiatives towards the development of strategies for the management of HMWBs in ports was "ROM 5.1. Quality of coastal waters in port areas" (Revilla et al., 2007). It consists on a methodological standard included in the Spanish Recommendations for Maritime Works (ROM) to provide port authorities with procedures to explore the connection between port activities and the quality of water bodies (Ondiviela et al., 2012). Using a DPSIR scheme (Driver, Pressure, State, Impact, Respond), ROM 5.1 provides tools for the classification of water bodies in port areas, the environmental risk assessment, the environmental monitoring and the management of contaminant episodes (Ondiviela et al., 2012). This program makes reference conditions and security thresholds available, which is crucial for quality assessment of marinas. The standard of ROM 5.1 has also been applied at European scale and its implementation stressed the importance of considering both water and sediments to evaluate the ecological potential, confirming sediments as main evidence of the contaminant episodes (see Ondiviela et al., 2013). Indeed, regarding pollutants, most studies have focused on sediments (e.g. Mali et al., 2017; Sim et al., 2015) and several methods have been developed to estimate background values and degree of pollution in sediments (Birch, 2016) and to provide interpretative tools for assessing the biological significance of chemical pollution in the context of the Sediment Quality Guidelines (SQGs) (Birch, 2018).

Concerning SQGs, estimating potential environmental risks of worldwide coastal navigation on water quality is an important step to design a sustainable global market. The Environmental Risk Assessment (ERA) is the process that evaluates the likelihood of adverse ecological effects ocurring as a result of exposure to one or more stressors (US EPA, 1998). It provides a framework for objectively and systematically evaluating the risks posed by environmental contamination to ecological resources (Chapman and Mann, 1999) and defines the SQGs. ERA methods are mainly developed for commercial harbours and have several limitations (e.g. they lack ecological characterization of the environment, focus on unique stressors or hazards or consider just the impacts generated by accidents) (Gómez et al., 2019). Several methods and models, such as the Complexity Tidal Range Index (CTRI) or the Marina Environmental Risk Assessment (MERA), have been recently applied to prioritize environmental and planning strategies in marinas (e.g. Gómez et al. 2019, Valdor et al. 2019). However, the progress in these tools contrasts significantly with the scarcity of available physico-chemical measures and abiotic datasets from marinas. In this sense, detailed and comparative baseline data of pollutans in the water column and sediment of marinas worldwide are necessary to complete the information of the conceptual models and to provide a whole framework of environmental risks in marinas. Investigation from an integrative perspective is necessary (McVay et al., 2019). The establishment of adequate indicators able to discriminate among marinas is the first step to provide adequate reference conditions and security threshold values. Baselines are also essential as starting points to properly establish long-term monitoring programmes, to complete and update empirical models, to undertake ERAs efficiently, to evaluate backgrounds and enrichments, and to develop adequate SQGs.

Southern Iberian Peninsula (SIP) is an adequate study area since (i) represents an horizontal gradient Atlantic-Mediterranean, (ii) encompasses a very high number of marinas along the coast and (iii) includes detailed reference conditions of natural environments.

To fill the gap between management and data availabiliy, the main *objectives* of the present study were: (*i*) to provide a complete dataset of stressors measured in water and sediment of marinas from SIP; (*ii*) to identify relevant abiotic variables useful to discriminate among marinas, (*iii*) to explore potential spatial patterns of environmental variability among marinas along SIP in a Mediterranean-Atlantic axis, and, eventually, (*iv*) to develop a simple, straightforward and easy-to-apply method to assesss the environemtnal quality of marinas based on sediment background references, ROM 5.1

reference values, and SQGs information. Additionally, we conducted a comparison with literature datasets from marinas worldwide to provide some recommendations about the use of selected stressors from water *vs* sediment, as well as methodological considerations for some of them.

2.Material and methods

2.1. Study area and sampling

Eight marinas were selected along SIP, four in the Atlantic coast (Faro, Chipiona, Puerto América and Barbate) and four in the Mediterranean (La Línea-Puerto Chico, Fuengirola, Motril and Almería) (Fig. 1). Sampling was conducted from 26 June to 2 July 2017. Within each marina, three floating pontoons were randomly selected. At each pontoon, measurement of seawater temperature, salinity and pH were made *in situ*. Salinity (psu) and temperature (ºC) were measured with a WTW LF323-A/SET conductivity meter and pH with a WTW pH 300i/SET pH meter. Turbidity (ntu: nephelometric turbidity units) was also measured, using a WTW Turb® 335 IR turbidity meter, and water transparency (cm) was determined using a Secchi disk (Bertelli et al., 2020; Taillie et al., 2020). Additionally, to quantify the multiple stressors in water and sediments of the marinas (*objective i*) three samples of surface seawater (0.3 m depth) (one per pontoon) were collected, and three sediment samples (one per pontoon) were taken from the bottom using a small van Veen grab (sampling area: 15 x15 cm). The depth of the bottom at the marinas (where the sediment was collected) was 1.1 m (Faro), 3.5 m (Barbate, La Línea and Motril), 4 m (Chipiona and Almería), 5 m (Fuengirola) and 6 m (Puerto America).

All samples were immediately frozen (-20ºC) until the laboratory analyses. Mean values of all environmental variables were obtained using the three measures (one per pontoon) taken in each marina.

2.2. Analytical procedures

2.2.1. Seawater analyses

Analysis of major, minor and trace elements (Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, P, Pb, S, Sr, Zn) in water samples was performed using a VARIAN ICP 720-ES (simultaneous ICP-OES with axially viewed plasma), equipped with ultrasonic nebulizer CETAC U5000AT+ after filtration through Nylon filters (pore size = 0.45μm) and acidification with 2% HNO_3 (30%). Blank and standard solutions for devices calibration were used. For preparation of standards we used <18 MΩ/cm ultrapure water supplied from a Milli-Q Millipore system (Bedford, MA, USA) and TracepureTM HNO_3 from Merck (Darmstadt, Germany). Calibration and Quality Control (QC) solutions were prepared from an ICP multi-element standard solution IV Certipur obtained from Merck and Spectrascan certified reference solution from LGC Standards GmbH (Wesel, Germany). To prevent contamination of the samples with traces of any metal, all material used for sample storing and treatments and all lab ware equipment was soaked in 2% v/v HNO_3 solution followed by two washes with Milli-Q water. The calibration blank was prepared with 2% v/v HNO_3. Analytical blanks and standard reference materials were run in the same way as samples. The accuracy of the analytical methods was assessed through reference water sample (TR-434 Trace of metals in drinking water from INTER 2000 Program and Trace Elements in Estuarine Water CRM 505 No. 048). The recoveries ranged from 89.2–109.4%. The differences in concentrations between analysed and certified values were generally <10%.

Total organic carbon (TOC) was analysed on a Shimadzu TOC-VCSH with ASI-V auto sampler after filtration through Whatman 1 paper (pore diameter= 11 μm). It was obtained by the difference between total carbon (TC) and total inorganic carbon (TIC) which was removed by acidification and measured separately.

The booster biocides Irgarol 1051, Clorothalonil, Dichlofluanid and Sea-Nine 211 (organic compounds used in antifouling paints in combination with Cu) were measured in the water samples by and stir bar sorptive extraction–thermal desorption–GC–MS developed and detailed by Giráldez et al. (2013).

2.2.2. Sediment analyses

pH (pH/redox Crison with the electrode M-506) and electrical conductivity as an estimation of sediment salinity (conductivity meter PCE Instruments) were recorded in the laboratory after adding distilled water to the sediment with 1:2,5 w/v, and 1:5 w/v, respectively.

Particle-size distribution in sediment samples was determined by the hydrometer method, after dispersion by sodium hexametaphosphate (Gee and Bauder, 1986) and classified into three classes: sands (2-0.063 mm), silt (0.063-0.002 mm) and clay (<0.002 mm) (Shepard, 1954; Mali et al., 2017).

For analysis of major, minor and trace element concentrations in sediment (Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, P, Pb, S, Sr, V, Zn), sediments stored at -20ºC after sampling were air-dried, crushed and sieved though a 2 mm sieve and then ground to <60 μm. Samples were digested with aqua regia (1:3 conc HNO3:HCl) in a microwave digester. Quantification of elements in the extracts was achieved using a VARIAN ICP 720-ES (simultaneous ICP-OES with axially viewed plasma) as explained above for water samples. The accuracy of the analytical methods was assessed by carrying out analyses of the BCR (Community Bureau of Reference) sample BCR®-320R, and soil sample reference ISE 2015-4.3 from the Wageningen Evaluating Programs for Analytical Laboratories, International Soil-analytical Exchange (WEPAL; ISE). The recoveries ranged from 95.5–114.7%.

Total organic matter (TOM) and/or total organic carbon (TOC) are traditionally among the most common measures in marine sediments, but analytical methods to measure them differ among studies. Therefore, in the present study three different methods were used with the purpose of comparison and further discussion. TOC was analysed by: (1) using a Shimadzu TOC-VCSH with ASI-V auto sampler as explained for water samples (hereinafter analyzer method "a") and (2) dichromate oxidation and titration with ferrous ammonium sulphate (Walkley and Black, 1934) (hereinafter oxidation method "o"). Additionally, (3) TOM was determined gravimetrically by loss on ignition 450ºC for 24h (Sánchez-Moyano et al., 2010) (hereinafter calcination method "c"). To enable comparisons, the results obtained for the three metods were always expressed as TOM (TOMa, TOMo and TOMc respectively). The conversion factor of 1.724 was used to expressed TOC as TOM in cases (1) and (2) (see Nelson and Sommers, 1996)

N was determined by the Kjeldahl digestion method to convert organic nitrogen to ammonia. The digestate was alkalized, the ammonia distilled into boric acid and titrated with an acid of known concentration (Bremmer and Mulvaney, 1982).

Total hydrocarbons were measured by Soxhlet method using petroleum ether as organic solvent (see Vázquez-Blanco et al., 2000; Kuppusamy et al., 2020; López-Bascón and Luque de Castro, 2020)

Determination of faecal coliforms was conducted by straking on Brilliant Green Bile agar 2% (BGBL) and incubated at 44±1ºC during 48h. Presumptive faecal coliform colonies were confirmed by gas and indol production. For gas production, after cultivation in tryptophan broth, tubes developing turbidity and gas were considered as presumptive positives. Indole production was tested using drops of Kovac's reagent and the result was considered positive if a red-violet complex on the surface was formed. Colonies abundance was reported as number of CFU (colony-forming units) per g (dry weight) of sediment. Quantification limit of the method was 50 CFU/g.

The booster biocides Irgarol 1051, Chlorothalonil, Dichlofluanid and Sea-Nine 211 were also measured in sediment samples following the methodology of García et al. (2020).

2.3. Statistical analyses

The experimental design included two factors: 'Location' (Lo), a fixed factor with two levels (Mediterranean *vs* Atlantic) and 'Marina' [Ma(Lo)], a random factor with four levels, nested with Lo. The Atlantic marinas were Faro (FAR), Chipiona (CHI), Puerto América (AME) and Barbate (BAR), and the Mediterranean ones were La Línea-Puerto Chico (LIN), Fuengirola (FUE), Motril (MOT) and Almería (ALM). The number of replicates was n=3, corresponding with the three pontoons randomly selected for each marina (one measure per pontoon, three measures per marina). Two separate data sets, one for seawater and other for sediments, were considered and analysed separately.

Two Principal Component Analyses (PCAs) were carried out with the environmental matrices of seawater and sediments respectively to explore which variables could be the major contributors to ordination of marinas (*objective ii*). Previously to conduct PCAs, data were transformed with log (x+1) (Guerra-García and García-Gómez, 2005; Clarke and Gorley, 2006). Taking into account the disparity in the literature with respect to data pre-treatment prior to PCA, different options were tested (trace metal normalization, transformation of selected variables, standardization by substracting the mean and dividing by the standard deviation); the highest values of variance explained and the clearest patterns were obtained with log transformation, which was consequently used throughout the whole study. Although the application of the routinely used log (x+1) transformation has been reported to skew the data and mask important trends in some cases (Reid and Spencer, 2009), this transformation has been successfully applied not only to obtain a normal distribution, but also to perform other functions such as standardizing the dataset and reducing the influence of outliers (e.g. Cao et al., 1999; Prieto et al., 2008; Faustino-Queiroz et al., 2020).

To explore environmental variability among marinas along the SIP in a Mediterranean-Atlantic axis (*objective iii*), differences in environmental data among 'Location' and 'Marina' were tested for water and sediment separately by the use of a permutational multivariate analysis of variance (PERMANOVA) following the design explained above. Analysis was based on Euclidean distance measures and Monte Carlo tests were included since the number of unique permutations was low (n=35). Significant p-values were obtained by computing 9999 permutations of residuals under a reduced model as this method gives the most accurate Type I error for complex designs (Anderson, 2005). Additionally, to test the dispersion among samples, a permutational analysis of multivariate dispersions (PERMDISP) was used. To explore differences for each variable, two-way ANOVAs were additionally conducted using the same experimental design. Homogeneity of variances was checked using Cochran's test (Underwood, 1997).

Regarding with measures of TOM in sediments, the three methods were compared using Pearson correlations, as the three variables (TOMa, TOMc and TOMo) followed a normal distribution according to Shapiro-Wilk tests. Values obtained with the three methods were also statistically compared using one-way ANOVA and the post-hoc Student Newman Keuls (SNK) was used for pair comparisons.

PCA and PERMANOVA analyses were carried out using PRIMER v.6+PERMANOVA package (Clarke and Gorley, 2006) and ANOVA was conducted on GMAV5 software (Underwood et al., 2002).

2.4. Trace metal normalization

Major, minor and trace elements occur naturally in the environment and this complicates assessment of contaminated marine sediments because measurable quantities of metals do not relate *per se* anthropogenic enrichment (Guerra-García and García-Gómez, 2005). Geochemical normalization can compensate for both the granulometric and mineralogical variability of metal concentrations in sediments (Liu et al., 2003). Al and Fe have been traditionally used as conservative elements to normalise the trace metal contaminants and they can be chosen as normalizers if they have significant relationships with the finest fraction of sediment (see Reid and Spencer, 2009; Ho et al., 2012; Birch, 2020). Therefore, we used Spearman correlations (in this case Shapiro-Wilk tests were significant) to explore the relation of these two conservative elements, Al and Fe and percentage of silt+clay in the sediment. Absolute concentrations of trace metals were graphically compared with the normalized ones and patterns were discussed.

Additionally, the ratio Cu/Mn was also calculated in order to detect anoxic sediments (Jung et al., 1996).

2.5. Sediment quality assessment

The quality assement in marinas (*objective iv*) was focused on sediment stressors due to several reasons: (i) sediments are usually preferred to assess the environmental condition of marine environments because they serve as storage of contaminants being evidence of the pollution episodes (Ondiviela et al., 2012, 2013); on the contrary, water column is dynamic and highly variable requiring considerable temporal sampling (Rodríguez et al., 2006); (ii) the first part of the present study confirmed that sediment variables better contributed to differentiate marinas than seawater ones (see Table 1 of result section, and (iii) most of the background values and guidelines are available only for sediment, and information for seawater is scarcely found in literature for most of stressors.

The main 15 stressors measured in the sediments (TOC, N, P, S, Hydrocarbons, Faecal coliforms, Irgarol and the trace elements As, Cd, Co, Cr, Cu, Ni, Pb and Zn) were selected, and compared with background data, values of ROM 5.1 and SQGs.

2.5.1. Background data

Regarding background concentrations, we considered a total of 35 undisturbed sites with low human impact (maximum 10 m deep) selected to encompass the whole coast of Southern Spain (Sánchez-Moyano et al., 2005). Sediments of these sites were collected from July to September 2000 as a part of a monitoring programme in the context of WFD developed by the *Agencia de Medio Ambiente y Agua* (AMAYA) supported by the *Consejería de Medio Ambiente* of the Andalusian Government, focused on the environmental control of the coast based on the macrofaunal communities and their relationships with the abiotic variables (see e.g. Guerra-García et al., 2013). Data from sediments of little anthropized beaches of the Alboran Sea

(Navarro-Barranco et al., 2019) were also used for comparison. Additionally, background references of uncontaminated sediments of the Iberian Peninsula, including Portugal (Caeiro et al., 2005: Sado estuary) and Spain (Carballeira et al., 2000: Galicia; Borja et al., 1996: Basque Country) (see Birch, 2017) were also considered, together with background established for France (Tolosa et al., 1996), UK (Nagpal, 2004), USA (Lee et al., 2006) and Australia (Preda and Cox, 2002). Global average upper crust (marine shale) reference values provided by Turekian and Wedepohl (1961) were also considered.

2.5.2. ROM 5.1 values

In the context of water bodies classification by WFD, data of marinas from SIP were compared with reference conditions and security threshold values estimated for HMWBs according to ROM 5.1 to assess the stress caused by marinas in terms of ecological quality (Ondiviela et al., 2012).

2.5.3. Sediment Quality Guidelines (SQGs)

Concerning SQGs guidelines from the following countries were considered: Spain (Manchaca et al., 2012; Buceta et al., 2015), France (Kenworthy et al., 2018), Norway (Bakke et al., 2010), USA (Long et al., 1995a, 1995b), Canada (Persaud et al., 1993) and China (Lao et al., 2019).

2.5.4. Methodological procedure

The following methodological protocol was developed to assess a quality status based on environmental pollution stressors of SPI marinas:

(1) Compilation of all literature indicators used in the assessment (backgrounds, ROM 5.1 reference values, and SQGs guidelines).

(2) Assignment of three quality categories for each stressor: good (green), moderate (orange), bad (red) and the concentration ranges for each category, according to the global comparison with the compiled data in (1). Following the precautionary principle, the categories were established in a conservative way, being very restrictive (good assignment can be only reached with low concentrations of the stressor).

(3) Assignment of a score for each stressor according to the colour category. Green was scored with 10 points, orange with 5 points and red with 0 points.

(4) Calculation of MEPI index as the sum of the 15 scores in each marina. The final value of MEPI according to the method proposed could range between 0 (15 red stressors) and 150 (15 green stressors) and let us to compare marinas globally according to a global assement of the sediment quality.

(5) Establishment of environmental status of each marina according to MEPI index. Again, following the precautionary principle we were very restrictive and conservative to establish quality status levels in marinas based on MEPI. We proposed the following intervals: (<90: bad, 90-120: moderate, >120: good). For example a marina with 10/15 green stressors and 5/15 orange stressors would have a MEPI=125 and would be considered as good.

In general, the criteria of background reference values, ROM 5.1 regulations and SQGs recommendations were generally coincident among them and along different countries. However, when these values showed different patterns, the most conservative ones were priorized. The simultaneous consideration of a wide range of reference values for comparison could involve certain degree of subjectivity in the established quality levels based on the colour assignment for each stressor. However, we consider this option more comprehensive than the alternative one by selecting a single indicator for comparison (e.g. backgrounds for sediments of Southern Spain, or only reference conditions of ROM 5.1, or just SQGs from a particular study).

3. Results

3.1. Environmental data of seawater and sediment

All the abiotic data measured in seawater and sediment are included in Table 1. Chlorothalonil, Dichlorofluanid and Sea-Nine 211 were also measured but were not detected in the water column of any marinas; therefore, these variables were not included.

Both seawater and sediment datasets did not reveal globally significant differences between Atlantic and Mediterranean marinas (see PERMANOVA results for factor 'Location' in Table 2). For this factor, ANOVAs only showed significant differences in a few variables (Table 1). However, PERMANOVA showed that factor 'Marina' was strongly significant, revealing environmental differences in water column and sediments among marinas (Table 2). This result was supported by ANOVAs, which confirmed significant differences among marinas for 11 out of 27 variables in seawater and 31 out of 36 in sediments (Table 1).

PCA analyses hightlighted again differences among marinas (Fig. 2). PCA based on seawater data confirmed the results of PERMANOVA and ANOVAs showing a clear separation according to environmental data. Sampling sites were mainly segregated by turbidy and Irgarol (Fig. 2). Axis 1 explained the 66% of the total variance and significantly correlated with turbidity ($r=0.98$, $p<0.001$). Although PERMANOVA did not show significant differences between the Mediterranean and Atlantic, PCA revealed some segregation of Mediterranean (located on the left) vs Atlantic (located on the right) sites along axis 1 with Atlantic sites being characterised by higher turbidity values. Turbidity was, indeed, one of the few variables that showed significant differences between the Atlantic and Mediterranean according to ANOVA (Table 1). Axis 2 explained 28% of the total variance and correlated with Irgarol ($r=0.99$, $p<0.001$) and axis 3 (not represented graphically) explained only the 3% of the remaining variance and correlated with temperature ($r=0.78$, $p<0.001$) and TOC ($r=0.55$, $p<0.01$). Irgarol showed the highest concentrations in the water column in La Línea, and the lowest in Faro and Motril. It is remarkable that much of the variability in seawater is due to only 4 variables (turbidity, Irgarol, temperature and COT). In fact, these variables showed significant differences among marinas (ANOVAs in Table 1). Concerning sediments, although most of the variables showed significant differences among marinas (Table 1), PCA showed that only some of them explained most of the data variability (Fig. 2). Axis 1 explained the 42% of the total variance and signficantly correlated only with the sediment granulometry (clay, $r=-0.44$, $p<0.05$, and sand, $r=0.35$, $p<0.05$). Axis 2 explained the 18% of the total variance and correlated with faecal coliforms ($r=0.65$, $p<0.001$) and S ($r=-0.32$, $p<0.05$). Axis 3 (not represented graphically) absorbed the 10% of the variability and was correlated with hydrocarbons ($r=0.47$, $p<0.01$) while Axis 4 was still explaining a moderate fraction of the total variance (9 %) and correlated with Irgarol ($r=0.43$, $p<0.05$) and Ni ($r=-0.53$, $p<0.01$).

3.2. Total organic matter (TOM)

The values of sediment TOM obtained with the three methods (method "a" through TOC analyzer, method "o" with dichromate oxidation and method "c" by gravimetry quantification of loss on ignition) showed significant differences [one-way

ANOVA, F=40.2, $p<0.001$, n=24; SNK: (a=o) ≠ c)]. For method "c" mean values of TOM in marinas ranged from 3.27 to 27.5%, while method "a" and "o" showed values significantly lower, ranging 0.69–3.22% and 1.32–5.76% respectively. Additionally, while TOMa and TOMo were significantly correlated (r=0.861, $p<0.001$), TOMc was not correlated with either TOMa or TOMc respectively (TOMc vs TOMa: r=0.400, p=0.053 n.s.; TOMc vs TOMo: r=0.348, p=0.095 n.s.; Fig. 3A). The three methods contributed to discriminate among marinas.

3.3. Trace metal normalization

When the usefulness of the two conservative elements Al and Fe were explored through their correlation values with the finest fraction of the sediment (*i.e.* clay), only Al showed a very significant correlation (r=0.972, $p<0.001$) (Fig. 3B). Correlation was also significant when the whole fine fraction including silt + clay (<0.063 mm) was considered (r=0.875, $p<0.001$). Conversely, Fe was not correlated with either clay (r=0.278, p=0.189 n.s.) nor silt + clay (r=0.387, p=0.062 n.s.). Therefore, Al was used as conservative element to normalize trace elements. Although normalized ratios of trace metals showed, in general terms, similar patterns than the absolute values, a clear enrichment gradient towards the Mediterranean was observed for As, Cd, Co, Cu and Pb (Fig. S1, supplementary material).

The ratio Cu/Mn showed the highest value in Faro, followed by La Línea, Fuengirola and Motril (Fig. 3C).

3.4. Quality assessment

Background values, references from programme ROM 5.1 and worldwide Sediment Quality Guidelines (SQGs) were compiled in Table 3. Most of the data in the literature correspond to trace metals; while the available information or reference values for TOC, N, P, S, total hydrocarbons and faecal coliforms is still scarce. Background values were, in general terms, similar among countries and in agreement with the reference conditions established by the ROM 5.1 for HMWBs. Concerning SQGs, although countries used different classification methods (effect ranges, classes, grades), reference values were mostly coincident. For example, basal concentrations for low effects in the biota for As, Cd, Cr, Cu, Ni, Pb and Zn were of the same order for ERL

(Effect range – Low) and category A in Spain, class I in Norway, ERL in USA, LEL (Lowest Effect Level) in Canada and Grade I in China.

Quality categories of concentration ranges (bad, moderate and good) were established for each stressor (Table 4) based on the global information of backgrounds, ROM 5.1 and SQGs values included in Table 3.

For each stressor, good (green) was scored with 10 points, moderate (orange) with 5 points and bad (red) with 0 points.The value of the MEPI index for each marina was the sum of the scores of the 15 stressors (Fig. 4)

According to the proposed methodology based on 15 basic stressors, final score of marinas (MEPI index) from SIP ranged from 60 to 110 points. Marinas with better environmental quality were Almería (110 points), Faro, Puerto América and Motril (105 points). The marinas with the lowest score were Fuengirola and Barbate with 60 and 70 points respectively. Marinas were categorized as bad or moderately polluted, although Almería was close to reach the good status.

4. Discussion

4.1. Environmental data in marinas

This work represents the first comprehensive study assessing environmental pollution in marinas from a multi-stressors approach, based on the measures of 30 variables in seawater and 34 in sediments including natural abiotic variables, organic and inorganic chemical pollutants and bacterial stressors.

4.1.1. Seawater

In general, water parameters were not very useful to discriminate among marinas during the present study (see ANOVA results in table 1). However, turbidity (ntu) and Irgarol 1051 (ng/l) turned out to be especially important (together, they explained the 94% of the total variance in the PCA conducted with the 30 seawater parameters).

Guerra-García et al. (2006) found that turbidity was the factor that best correlated with the intertidal assemblages of flora and fauna in Algeciras Bay, Southern Spain. Similarly, Fernández-Romero et al. (2019) found this variable to be one of the most relevant driving polychaete distribution in marinas (Fernández-Romero et al., 2019). Turbidity can also have important effects on estuarine food webs (Reustly and

Smee, 2020), on dynamics of marine planktonic and epibiont ciliates (Jones et al., 2019; López-Abbate et al., 2019), on biotic interactions between native and invasive species (Santos et al., 2018) and on the visual acuity of marine mammals (Weiffen et al., 2006), among others.

Irgarol 1051 is an organic booster biocide widely used in several antifouling products. Booster biocides are organic compounds used as additives in copper-based antifouling paints. The role of antifoulants is to prevent the colonization of submerged structures by marine organisms, especially boat hulls (García et al., 2020) since biofouling has negative environmental and economical impacts (e.g. increases in fuel consumption, corrosion, potential for the introduction of exotic species) (Saleh et al., 2016 and references therein). Organotins, particularly tributyltin (TBT), were the most widely used compounds in antifouling paints until the 1990s. The International Maritime Organization prohibited totally the use of TBT on January 2008 (Antizar-Ladislao, 2008). Then, the use of organic booster biocides was generalized as alternative in antifouling products (Sapozhnikova et al., 2007). Some of the most frequently used biocides are Irgarol 1051, Clorothalonil, Dichlofluanid and Sea-Nine 211 (Lee and Lee, 2017; Amara et al., 2018). The increased use of biocides during the last years has resulted in high concentrations in the water column and accumulation in the marine sediment, causing ecological as well as ecotoxicological damage to marine organisms (Giráldez et al., 2013; Amara et al., 2018;). Although these biocides are intended to be environmentally less harmful compared to the organotin biocides, the problem of toxicity on several marine species remains (Amara et al., 2018 and references therein).

Although the four dominant biocides were measured in the present study, only Irgarol 1051 was detected in seawater. Irgarol is very toxic to freshwater and marine algae (Albanis et al., 2002; Cai et al., 2006) and has acute toxicity to many aquatic organisms (Amara et al., 2018: table 7). In fact, Irgarol has a long half-life and slow degradation with a potential to impact non-target organims and affect seafood production and other ecosystem services (Egardt et al., 2017). The environmental risk limit for Irgarol in water (ERLwater) is 24 ng/l (van Wezel and van Vlaardingen, 2004) and the values measured in all the studied marinas exceed this concentration. An extensive literature review of worldwide concentrations of the main biocides in marine ecosystems (Table S1, supplementary material) revealed that concentrations of Irgarol exceeding enviromental risk level were measured not only in most of marinas and ports, but also in some bays and estuaries. In many marinas from Europe, North and South America and Asia, Irgarol measures were often above 100 ng/l (Table S1, Fig. 5).

In certain countries, such as New Zealand, UK, Denmark and Sweeden (see e.g. Egardt et al., 2017) the use of Irgarol in anti-fouling products has already been restricted or phased out. In fact, in 2016, following the European Chemicals Agency's Biocidal Products Committee (BPC) within the context of the implementation of the Biocidal Product Regulation (EU) N°528/20127, the EU Commission decided not to continue the authorization of Irgarol as an antifouling product (see Egardt et al., 2017 and Commission Implementing Decision EU 2016/107). However, this biocide is still being used in many countries, and it will be surely present in marine ecosystems for many years, similarly to TBTs, which are still detected in seawater and sediments despite being banned for over 10 years (Antizar-Ladislao, 2008). Furthermore, taking into account that these biocides started to be used very recently (<40 years), potential long-term effects and persistence are not properly understood yet. Therefore, we encourage scientists, authorities and policy makers to include Irgarol as selected stressor in environmental programmes to monitor pollution in marinas. Most of the available information dealing with Irgarol is focused on seawater and sediment concentrations, or ecotoxicological experiments in laboratory conditions (e.g. Key et al., 2008) but data of how Irgarol can affect the structure and diversity of benthic invertebrate assemblages are scarce. Indeed, its effect on the marine communities inhabiting marinas still remains unexplored.

4.1.2. Sediment

Sediment serve as storage of contaminants and greatly influence the quality of interstitial and overlying water through physical (re-suspension), biological (bioturbation) and chemical (desorption and benthic diffusion) processes (Birch, 2017). The most toxic and persistent contaminants and organic compounds may be accumulated or retained in the sediment (Ondiviela et al., 2012) which in turn may provide a valuable record of the historical contamination. Although water, sediments and biological indicators are important to evaluate the ecological potential, Ondiviela et al. (2013) confirmed sediments as the main one. Indeed, the present study revealed that sediment variables were much more useful to discriminate among marinas than seawater ones (see ANOVA results in Table 1). Although the factor 'Location' was not significant for most of variables reflecting the lack of differences between the Atlantic and Mediterranean, the factor 'Marina' was significant for most of variables. This indicates the environmental singularity of each marina according to sediment stressors. Chatznikolaou et al. (2018) already pointed out the singularity of port and marinas. In fact, the importance of local scale in environmental management has been also reported for marine communities inhabiting concrete-made artificial structures, such as cubes or tetrapods (e.g. Sedano et al., 2020). Sediment, therefore, seems to be the advisable compartment to focus the efforts in management strategies due to more discriminant abiotic data.. Therefore, geographically close marinas can have very different environmental quality, which involves singular fitting management

In any case, available information of chemical stressors in marinas is remarkably scarce, even for sediments (Table 5). Although data are abundant for large ports and harbours, studies specifically addressing pollution in marinas are lacking (Mali et al., 2017; Chatzinikolaou et al., 2018; Kenworthy et al., 2018;). Sediment stressors measured in marinas from SIP show, in general terms, similar values to those measured in other worldwide marinas (Table 5). In fact, concentrations values of stressors did not vary greatly among marinas, so sediments seem to be rather similar in terms of chemical pollution accross the globe. Unusually high values have been measured for some metals in certain marinas. For example, Cu concentration exceeded 1000 ppm in the marina of Port Camargue, France, and marinas of Virgin Islands, USA. Mn also exceed 1000 ppm in Veraval harbour, India. Pb and Zn also showed considerably higher values in Craft harbour Nova Scotia, Canada than in the remaining marinas (Table 5).

Trace metal pollution in sediment is a global problem for aquatic ecosystems, involving serious threat to the aquatic biota because of their toxicity, non-biodegradability, and bio-magnification in the food chain (Sundarajan et al., 2017). Marinas and ports usually display high concentrations of trace elements due to harbour activities. Among the many potential sources, metal-based antifouling systems clearly play a major role in contamination (Briant et al., 2013). In the 20th century, Pb-based paints were used to prevent corrosion and fouling. Pb was subsequently replaced by Cu, which has been used as a biocide in antifouling paints for >100 years and its concentration is, therefore, higher in areas where ships are anchored, moored or maintained such as marinas or ports (Biggs and D'Anna, 2012; Chatzinikolaou et al., 2018). Zn is also associated with pollution from antifouling paints in ports and marinas and can cause sublethal effects on benthic organisms (Briant et al., 2013; Chatzinikolaou et al., 2018). As expected, concentrations of Cu and Zn were moderately high in some marinas of SIP according to background information and SQGs (see Fig. 4 and Table 3). The significant enrichment of these two heavy metals is a general trend in sediments of other marinas throughout the world (Table 5). Noticeable concentrations of Cr and Ni were also measured in Fuengirola marina. These trace metals have been associated with industrial origin and with certain lipidic contaminants such as asphaltenes (Guerra-García and García-Gómez, 2005).

The higher ratio Cu/Mn in Faro marina probably indicates a precipitation of copper sulphide and a dissolution of manganese oxide, typical of anoxic sediments (Jung et al., 1996). This is clearly supported by the remarkably high concentration of S measured in this marina (Table 1). Although this marina is one of the less polluted as found in the present study, this anoxic trend could affect negatively benthic communities. High values of Cu/Mn ratios have been reported for enclosed areas of other harbours (Guerra-García and García-Gómez, 2005).

Granulometry, hydrocarbons and faecal coliforms were especially relevant according to sediment PCA. Granulometry has been traditionally considered one of the main factors modulating benthic communities, including those inhabiting harbour areas (Guerra-García and García-Gómez, 2004; Moreira et al., 2005). Total hydrocarbons and faecal coliforms showed very high concentrations in most of the studied marinas, with values exceeding background or SQGs (Figure 4, Table 3). Hydrocarbon pollution is very common in ports, harbours and marinas due to loading and dumping associated with shipping operations, accidental oil spills and recreational boating.

Routine sediment dredging and related activities induce resuspension that may disseminate the faecal pathogens stored in the sediment (Luna et al., 2019). Although faecal pollution monitoring in beaches and bathing areas is exhaustive (Lee et al., 2006), there is a lack of data for marinas, and comprehensive studies to quantify and measure microbial pollution in these habitats are not available. SQGs are also scarce for this stressor in port environments. Taking into consideration the high concentrations of faecal coliforms in most of the studies marinas, we also boost to generalize its measurements in pollution monitoring programmes.

Besides granulometry, faecal pollution, hydrocarbons, S and Ni, Irgarol was also a relevant parameter in sediment characterization according to PCA. It was present in all marinas and exceeded ERL risk values in some of them (Table 3). Although the other booster biocides (Sea-Nine 211, Chlorothalonil and Dichlofuamid) were not detected in seawater, they were accumulated in the sediments, especially Sea-Nine which was present in all marinas. This reveals the generalized use of these antifoulants (especially Irgarol and Sea-Nine) in marinas of SIP.

4.2. Methodological remarks

As seawater turbidity has been the most discriminant measure in water column, we propose this to be a crucial measure in water monitoring programs in marinas. The Secchi disk has been traditionally considered a good proxy of water turbidity in oceanographic studies (Weiffen et al., 2006). However, it is not always useful in marinas, especially in very shallow ones (e.g. Faro). In these cases, when the transparency of the water is high, the bottom can be seen from above and the measurement of the Secchi disk, coinciding with the bottom depth, is not suitable. Alternatively, portable turbidimeters are unexpensive, easily handable, and provide accurate measures in nephelometric turbidity units (ntu).

Although total hydrocarbon and faecal coliform concentrations turned out to be important environmental stressors in sediment marinas according to PCA, data are very scarce in literature and, when provided, disparity in methodologies can prevent adequate comparisons. Although gravimetry and FT/IR (fourier transform infrared spectroscopy) methods can yield comparable hydrocarbon measures (Louati et al., 2001), most of the studies do not provide the total hydrocarbon concentrations; since they only focus on Polycyclic Aromatic Hydrocarbons (PAHs) measured by gas chromatography-mass spectrometry (GC-MS). Although some PAHs are particularly relevant (Han et al., 2020), we consider that a general measure of total hydrocarbons can be a good baseline and adequate proxy to explore hydrocarbon pollution in a marina in a general sense (Guerra-García et al., 2003). Concerning faecal pollution, besides the lack of studies in marinas and ports, disparity in methodological procedures and measurement units often make difficult comparisons among sites. Recently, the usefulness of molecular qPCR in monitoring faecal pollution in recreational water quality controls has been asssessed (e.g. Haugland et al., 2016), but little work has been conducted on water and sediment quality in ports (Luna et al., 2012). Furthermore, quantification results obtained from cultivation differ from those obtained by qPCR, difficulting further comparisons (Luna et al., 2019). Hence, the establishment of standardized protocols to unify methodologies should be a priority to undertake intensive data collection about faecal pollution in marinas.

Concerning TOM, the present study reveals major discrepancies in the results obtained with the different methodologies (Fig. 3A). Oxidation and TOC analyzer methods ("o" and "a") yielded similar results, which were indeed highly correlated. However, gravimetry quantification of loss on ignition (method "c") overestimated significantly (up to 8 times higher) the amount of TOM in sediments. This could be due to the presence of calcium carbonate in empty mollusc shells, or to the loss of structural water in sediments rich in clay, which could increase moderately the weight after ignition providing artifact measurements (Kenny and Sotheran, 2013). We, therefore, recommend methods "o" or "a", especially if sediments are rich in the silt and clay fraction, such as those in marinas. Hence, data of literature obtained by method "c" must be taken with caution for comparison purposes since the present study revealed no significant correlation between the results obtained with this method and those yielded by method "a" or "o".

In this work, Al turned out to be better normalizer than Fe since correlated significantly with the amount of the finest granulometric fractions in the sediment. Al is often used as a normalizer for several reasons: it is the most abundant naturally-occurring metal; it is highly immobile so it is not significantly affectedby diagenetic processes and strong redox effects in sediments; and its content is generally not influenced by anthropogenic sources (see Ho et al., 2012). Normalization in the present study revealed an increasing in the enrichment of some metals toward Mediterranean marinas; the patterns differed, however, among elements. Briant et al. (2013) proposed Al-normalized enrichment factors (EFs) of trace elements with respect to the local background according with the equation EF= $(X/Al)_{sed}/(X/Al)_{bg}$, where $(X/Al)_{sed}$ is the ratio between the element X and Al in the sediment and $(X/Al)_{bg}$ is the local background ratio. The main practical problems lies in the fact that Al data are not always available in many studies and/or background sources. Furthermore, Al is a common material used in the construction of floating pontoons in marinas, so we cannot discard some degree of Al pollution , which could bias the enrichment ratios. Taking into account that background values, ROM 5.1 framework and SQGs provide reference values for absolute concentrations of trace metals; that Al data are not always available in the literature; and that potential Al pollution could not be discarded in marinas; we have used absolute metal concentrations for global comparisons and quality assement.

4.3. Developing standardized quality methods to assess pollution level in marinas

Regarding pollution assessment, the majority of efforts have been focused on sediments (e.g. Sim et al., 2015; Mali et al., 2017). Monitoring of sediment quality is an essential tool to assess the possible influence of anthropogenic pressure on ecosystem (D'Alessandro et al., 2018). The method used in the present study was based on the comparisons of a set of 15 sediment stressors with a selected panel of background values, SQGs and ROM 5.1 recommendations (reference conditions and security thresholds developed for HMWBs) (Tables 3 and 4, Fig. 4). The use of background concentrations can inform about contaminant concentrations prior to anthropogenic inputs (OSPAR, 2005; Casado-Martinez et al., 2006). A comprehensive review of the methods for determination of sediment metal background concentrations and enrichment in a wide range of marine environments was undertaken by Birch (2017). In the context of the Sediment Quality Guidelines (SQGs), several numerical approaches have been developed at global scale to provide interpretative tools for assessing the biological significance of chemical pollution (see e.g. Persaud et al., 1993 or Long et al., 1995a, 1995b). Information was extensively reviewed by Birch (2018) who including a variety of empirical and mechanistic techniques, together with sediment quality indices. Although SQGs have been used for over 25 years to aid screening of chemical contaminants for potential adverse biological effects (Birch, 2018), there is a lack of provisions with respect to required quality levels for sediments in Andalusian, Spanish or European regulations (Carrasco et al., 2003). In Spain, after more than 20 years implementing the "Recommendations for the management of dredged material in Spanish ports" (CEDEX, 1994), the Interministerial Commission for Marine Strategies adopted in April 2014 the "Guidelines for the characterization of dredged material and their relocation within waters of the maritime-terrestrial public domain" in the context of the Spanish Law 41/2010 on the Protection of the Marine Environment (Buceta et al., 2015).

Taking into account the great variety of local, regional and global backgrounds and regulations (ROM 5.1 and SQGs) considered for the assessment in the present study (Table 3), information for each stressor was synthesized in three categories of concentration ranges (Table 4). Fortunately, as pointed out by Birch (2017), background concentrations derived by multiple techniques and various organisations across the globe, do not vary greatly (see Table 3) making easier the comparisons between stressors in marinas and reference background concentrations. Applying this method, the total score (MEPI) calculated as the sum of the score for each stressor (green: 10 points, orange: 5 points, and red: 0 points) ranged from 60 (Fuengirola) to 110 points (Almería), indicating that marinas of SIP are characterised by moderate level of environmental pollution. These results support those provided by Gómez et al. (2019) based on an atlas of enviromental risk asssessed through Presure-State-Response model. These authors estimated the environmental risk of Spanish marinas based on the following indicators: density of boats, port operations, dredging probability, land use, flushing capacity, ecological singular elements, alteration by hydromorphological pressures and number of adopted measures and instruments to reduce human pressure and to improve the environmental performance. According to this PSR model, most of the marinas of Southern Spain are characterised by moderate environmental risk (see Gomez et al., 2019: 360). Therefore, our assessment (moderate) based on environmental pollution according to MEPI is in agreement with the environmental risks estimated by these authors

Implementation of the MEPI procedure in future studies could be constrained by the need to measure the 15 selected stressors. However, depending on the availability of environmental data in future studies, some stressors could be replaced by others whenever reference values of backgrounds and/or SQGs are accessible, and table 4 would be continually updating for new stressors. Therefore MEPI could be versatile and adjustable according to the availability of sediment stressors.

The assessment conducted in the present study indirectly involves the effects of pollution on biota through the SQGs, since these guidelines are indicators on the low or severe effects level on the biological communities, mostly based on laboratory essays. However, further studies are necessary in marinas along the SIP to describe benthic communities inhabiting sediments and to assess the direct effects of pollutant in species composition and abundance. In fact, the studies of diversity and structure of faunal assemblages are crucial, especially in anthropized environments, where changes in taxa composition and dominance are specific to human impacts (Tempesti et al., 2020 and references therein). Furthermore, the effect of the combined interaction of various stressors on biota is very difficult to predict using only indirect reference conditions and thresholds, thus requiring exhaustive studies of benthic fauna to explore direct associations between abiotic and biotic data through multivariate approaches.

5. Conclusions

The present study reveals the environmental singularity of each marina, especially regarding sediment stressors. Therefore, geographically close marinas can have very different environmental quality, which involves singular fitting management. Integrating environmental information with economic, social, and ecological priorities can help to design rationale policies for each marina (Karydis and Kitsiou, 2013; Usero et al., 2008, Borja et al., 2017). The simple quality assessment method developed during the present study may contribute to design standards for assigning "sustainable quality seals" to those marinas with lower concentrations of selected stressors according to MEPI index. In this sense, environmental quality is one of the main criteria (together with security, services, information and environmental education) to award a marina with a Blue flag label (Petroman et al., 2010). Eco-labels or eco-certifications provide incentives on marina slip rental, sailboat charter and hotel services (Sipic, 2017). Pollution baseline information is mandatory for the correct assignation of these awards.

Declaration of competing interest.

We declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data related to this article can be found online at https://doi.org/xxxxxxxxx.

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TABLES AND FIGURE CAPTIONS

Figure 1. Map of sampling area showing the location of the eight marinas studied along the coast of the Southern Iberian Peninsula. FAR: Faro, CHI: Chipiona, AME: Puerto América, BAR: Barbate, LIN: La Línea-Puerto Chico, FUE: Fuengirola, MOT: Motril, ALM: Almería.

Figure 2. Principal Component Analyses (PCAs) based on the abiotic variables measured in seawater (A) and sediments (B). Only variables which significantly correlated with axis were included in the graphical plot. TOC: Total Organic Carbon. FAR: Faro, CHI: Chipiona, AME: Puerto América, BAR: Barbate, LIN: La Línea-Puerto Chico, FUE: Fuengirola, MOT: Motril, ALM: Almería. Numbers (1, 2, 3) correspond to each replicate value.

Figure 3. A: Pearson correlations among the values of Total Organica Matter (TOM) in sediments measured by the three methods (a: analyzer, o: oxidation, c: calcination). B: Spearman correlation between Al concentration and percentage of Clay in sediments. C: Ration Cu: Mn in the marinas sampled along the study area.

Figure 4. Quality assessment developed during the present study for marinas of Southern Iberian Peninsula according to the levels established on Table 4. These levels were based on comparison with data of literature, including background levels, ROM 5.1 recommendations and Sediment Quality Guidelines (SQGs) (see Table 3 for details of comparison values). TOC: Total Organic Carbon. FAR: Faro, CHI: Chipiona, AME: Puerto América, BAR: Barbate, LIN: La Línea-Puerto Chico, FUE: Fuengirola, MOT: Motril, ALM: Almería.

Figure 5. Available Irgarol 1051 concentrations in worldwide seawater and sediments. See table S1 for details of references and detailed concentrations of Irgarol and other biocides.

Table 1. Environmental data measured in seawater and sediment at each marina. Values correspond to the mean of 3 replicates (3 pontoons randomly selected in each marina). Results of the two-way ANOVA [Location, fixed factor (Atlantic vs Mediterranean) and Marina, random factor nested to Location] for each variable are included. FAR: Faro, CHI: Chipiona, AME: Puerto América, BAR: Barbate, LIN: La Línea-Puerto Chico, FUE: Fuengirola, MOT: Motril, ALM: Almería, TOC: Total Organic Carbon, TOM: Total Organic Matter, n.d.: not detected, * $p<0.05$, ** $p<0.01$, *** $p<0.001$, n.s. not significant.

Table 2. Summary of the two-way PERMANOVA results for the enviromental data of seawater and sediments. *** significant differences with $p<0.001$, n.s. not significant. PERMDISP results for the factors 'Location' and 'Marina' are also included. SS=Sum of Squares, MS=Mean Square; MC=Montecarlo.

Table 3. Background values, references conditions and security thresholds from programme ROM 5.1 and Sediment Quality Guidelines (SQGs) obtained from the literature and used to the quality assessment of marinas of Southern Iberian Peninsula. TOC, total organic carbon; Hydroc, Hydrocarbons; F. col., Faecal coliforms; ERL: effect range-low; ERM, effect range-mdial; TEL; threshold effect level; PEL, probably effect level; LEL, lowest effect level; SEL, severe effect level. TOC values included for the marinas from Southern Iberian Peninsula (present study) corresponded with the oxidation method.

Table 4. Quality categories (good, moderate, bad) assigned for each selected stressor on the basis of a global assessment of the background values, references conditions and security thresholds from programme ROM 5.1, and Sediment Quality Guidelines (SQGs) obtained from the literature (see Table 3)

Table 5. Sediment environmental parameters measured in worldwide marinas. IP, Iberian Peninsula; NS, Nova Scotia; TOM, total organic matter; Hydroc., hydrocarbons; n.d., not detected (Number of marinas of each study in parentheses).

Table 1

Table 1. Environmental data measured in seawater and sediment at each marina. Values correspond to the mean of 3 replicates (3 pontoons randomly selected in each marina). Results of the two-way ANOVA [Location, fixed factor (Atlantic vs Mediterranean) and Marina, random factor nested to Location] for each variable are included. FAR: Faro, CHI: Chipiona, AME: Puerto América, BAR: Barbate, LIN: La Línea-Puerto Chico, FUE: Fuengirola, MOT: Motril, ALM: Almería, TOC: Total Organic Carbon, TOM: Total Organic Matter, n.d.: not detected, * p<0.05, ** p<0.01, *** p<0.001, n.s. not significant..

	Atlantic				Mediterranean				2 way-ANOVA	
	FAR	**CHI**	**AME**	**BAR**	**LIN**	**FUE**	**MOT**	**ALM**	**Location**	**Marina**
Seawater:										
Temperature (ºC)	24.20	24.93	23.73	20.00	20.53	17.43	19.87	25.80	n.s.	***
Salinity (psu)	37.13	36.40	36.93	36.50	36.97	36.97	36.53	36.50	n.s.	***
pH	7.81	8.19	8.07	8.08	7.98	7.97	8.00	8.08	n.s.	***
Turbidity (ntu)	3.86	10.63	2.41	2.31	1.05	0.87	1.40	0.80	**	***
Secchi disk (m)	0.87	3.83	6.00	3.50	3.33	5.00	2.83	3.93	n.s.	***
TOC (mg/l)	5.13	4.43	3.97	3.57	3.57	3.33	3.77	3.77	n.s.	***
Ca (mg/l)	437.81	437.69	444.28	441.33	448.36	447.50	439.73	442.56	n.s.	n.s.
K (mg/l)	630.65	627.84	650.83	638.16	643.75	638.86	641.52	636.44	n.s.	n.s.
Mg (mg/l)	1234.88	1223.83	1247.31	1232.88	1249.61	1256.21	1228.45	1231.76	n.s.	n.s.
Na (mg/l)	10503.22	10540.98	10644.98	10553.70	10709.21	10780.54	10677.10	10677.33	*	n.s.
S (mg/l)	996.58	991.04	999.18	990.75	1012.64	1007.21	991.03	1000.37	n.s.	n.s.
B (µg/l)	6075.79	6242.69	6385.07	6343.78	6512.08	6153.80	6179.54	6268.67	n.s.	n.s.
Ba (µg/l)	9.35	15.93	9.42	9.59	10.16	8.54	10.97	8.16	n.s.	**
Al (µg/l)	141.09	188.98	100.18	183.74	104.61	100.98	126.95	65.13	n.s.	n.s.
As (µg/l)	55.17	48.08	34.36	8.02	34.78	14.14	39.51	47.05	n.s.	n.s.
Cd (µg/l)	0.75	1.39	1.80	2.21	0.59	1.87	0.92	1.57	n.s.	n.s.
Co (µg/l)	3.25	4.27	4.89	3.22	3.46	3.47	5.92	3.12	n.s.	n.s.
Cr (µg/l)	5.27	5.12	4.00	6.61	3.99	4.59	4.25	5.24	n.s.	n.s.
Cu (µg/l)	20.06	25.44	14.90	20.07	22.55	17.31	19.32	27.48	n.s.	n.s.
Fe (µg/l)	125.04	318.08	110.25	164.05	71.90	24.96	26.94	48.82	*	n.s.
Mn (µg/l)	12.29	25.63	9.78	19.55	14.49	2.77	4.78	0.56	*	**
Ni (µg/l)	8.08	12.78	13.46	4.81	9.20	10.83	6.69	8.04	n.s.	*
P (µg/l)	25.48	21.62	41.53	22.13	16.65	21.57	20.08	9.62	n.s.	n.s.
Pb (µg/l)	50.77	58.07	56.42	57.58	79.84	42.06	54.04	39.00	n.s.	n.s.
Sr (µg/l)	7505.58	7469.12	7579.49	7514.84	7678.23	7611.00	7638.24	7550.54	*	n.s.
Zn (µg/l)	21.59	33.52	24.21	53.70	33.40	20.22	29.04	25.50	n.s.	**
Irgarol 1051 (ng/l)	71.33	125.00	107.67	128.33	217.33	84.33	55.33	113.67	n.s.	***
Sediment:										
Conductivity (µS/cm)	7456.67	8443.33	7190.00	8116.67	6710.00	8650.00	6150.00	5853.33	n.s.	*
pH	8.48	8.31	8.73	8.52	8.41	8.19	8.05	8.18	*	*
Sand (%)	54.77	3.83	43.70	9.60	42.07	19.20	45.23	77.17	n.s.	***
Silt (%)	28.27	33.63	24.53	21.10	31.13	49.73	51.80	20.93	n.s.	*
Clay (%)	16.97	62.67	31.77	69.30	26.80	31.07	2.97	1.90	n.s.	***
TOMa (%)	2.59	1.49	0.98	2.24	1.15	3.22	0.92	0.69	n.s.	**
TOMc (%)	13.63	19.50	10.86	27.50	15.32	11.22	3.45	3.27	n.s.	***
TOMo (%)	5.37	2.45	2.32	3.52	3.27	5.76	1.62	1.32	n.s.	***
N (%)	0.15	0.13	0.12	0.15	0.16	0.21	0.07	0.09	n.s.	**
Hydrocarbons (ppm)	2633.33	1766.67	1933.33	1500.00	6466.67	1966.67	4566.67	2166.67	*	*
Faecal coliforms (cfu/g)	100.00	1250.00	150.00	1483.33	1016.67	1050.00	566.67	1300.00	n.s.	**
Al (ppm)	15081.86	38216.64	21621.31	43933.50	16858.79	30547.74	10050.57	7699.16	n.s.	***
As (ppm)	12.12	17.41	10.09	16.82	8.85	23.06	16.46	16.17	n.s.	***
B (ppm)	48.86	76.02	57.75	91.07	46.40	38.84	10.17	10.60	*	***
Ba (ppm)	30.20	105.08	67.12	60.23	50.49	98.40	81.46	48.38	n.s.	***
Ca (ppm)	19919.71	112789.45	111798.00	52683.84	192153.11	59053.89	39818.29	44931.92	n.s.	***
Cd (ppm)	0.18	0.14	0.20	0.13	0.49	0.35	0.33	0.33	*	n.s.
Co (ppm)	3.51	8.66	5.86	10.33	4.65	17.95	9.59	7.26	n.s.	***
Cr (ppm)	22.50	49.64	33.32	81.89	60.91	115.19	14.82	7.35	n.s.	***
Cu (ppm)	62.18	93.18	73.56	135.95	69.16	154.28	91.23	58.02	n.s.	**
Fe (ppm)	17181.63	27612.72	18325.20	35109.58	14922.49	36518.32	28455.86	30989.53	n.s.	***
K (ppm)	4182.89	13529.94	8626.57	12587.59	6888.81	9966.23	2339.24	1609.36	n.s.	***
Mg (ppm)	4261.02	12929.07	11564.34	12461.21	16223.01	22830.73	14525.30	11237.26	n.s.	***
Mn (ppm)	76.57	398.58	291.45	501.01	147.78	385.70	275.92	351.47	n.s.	***
Na (ppm)	6348.18	13112.93	9526.21	14972.85	10080.16	12512.63	4475.53	3937.72	n.s.	***
Ni (ppm)	12.13	25.51	15.23	30.09	38.61	211.27	23.76	17.51	n.s.	***
P (ppm)	399.40	916.73	651.57	1113.92	615.46	1165.04	1060.85	702.48	n.s.	***
Pb (ppm)	15.74	35.19	33.99	23.59	22.25	36.88	32.11	38.96	n.s.	*
S (ppm)	11126.76	1927.32	3346.39	3190.93	6809.20	6988.26	1495.68	1165.10	n.s.	***
Sr (ppm)	86.53	273.41	346.18	190.97	974.42	195.01	112.34	134.23	n.s.	***
V (ppm)	38.49	68.92	45.71	97.37	39.50	63.56	29.07	21.88	n.s.	***
Zn (ppm)	67.27	148.37	589.21	166.45	90.99	191.61	136.02	133.42	n.s.	n.s.
Irgarol 1051 (ng/g)	1.31	1.53	0.62	6.69	1.35	0.20	3.64	1.37	n.s.	*
Chlorothalonil (ng/g)	n.d.	n.d.	0.17	n.d.	0.64	n.d.	0.45	n.d.	n.s.	n.s.
Dichlofuamid (ng/g)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	4.40	n.d.	n.s.	n.s.
Sea-Nine 211 (ng/g)	1.54	1.25	0.26	1.51	2.02	0.15	0.87	1.38	n.s.	n.s.

Table 2

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Table 2. Summary of the two-way PERMANOVA results for the enviromental data of seawater and sediments. *** significant differences with p<0.001, n.s. not significant. PERMDISP results for the factors 'Location' and 'Marina' are also included. SS=Sum of Squares, MS=Mean Square; MC=Montecarlo.

Source of variation	**df**	**SS**	**MS**	**Pseudo-F**	**p (MC)**	**SS**	**MS**	**Pseudo-F**	**p (MC)**
		Seawater				**Sediment**			
Location [Lo]	1	5.23	5.23	3.05	0.0615 n.s.	53.38	53.39	1.50	0.2073 n.s.
Marina [Ma (Lo)]	6	10.28	171	20.43	0.0001***	213.12	35.52	8.03	0.0001***
Residual	16	1.34	0.08			70.80	4.42		
Total	23	16.86				337.31			
PERMDISP	Lo	F=4.9608, p=0.0569 n.s.				F=0.5426, p=0.4836 n.s.			
	Ma	F=2.2031, p=0.4765 n.s.				F=5.6111, p=0.0957 n.s.			

Table 3

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Table 3. Background values, references conditions and security thresholds from programme ROM 5.1, and Sediment Qua
literature and used to the quality assessment of marinas of Southern Iberian Peninsula. TOC, total organic carbon; Hydroc
effect range-low; ERM, effect range-mdial; TEL; threshold effect level; PEL, probably effect level; LEL, lowest effect le
included for the marinas from Southern Iberian Peninsula (present study) corresponded with the oxidation method.

		TOC (%)	N (mg/kg)	P (ppm)	S (ppm)	Hydroc. (ppm)	F. col. (cfu/g)	As (ppm)	Cd (ppm)	Co (ppm)	Cr (ppm)	Cu (ppm)	Ni (ppm)
Marinas Southern Iberian Peninsula		0.8-3.3	70-210	399-1165	1165-11127	1500-6467	100-1483	9-23	0.1-0.5	3.5-17.9	7-115	58-154	11-211
Background													
Andalusian coast, Spain		0.1-0.4	<500	-	-	<1-55	-	-	<0.5	-	<1-40	<10-72	5-114
Alboran Sea, Spain		<0.2	-	-	<1500	-	-	-	-	-	28-60	<3	13-25
Galicia, Spain		-	-	-	-	-	-	-	-	-	30-54	20-35	31-38
Basque country, Spain		-	-	-	-	-	-	-	-	-	11	18	12
Portugal		-	-	-	-	-	-	20	1	-	50	35	-
France		-	-	-	-	-	-	-	-	-	-	-	-
UK		-	-	-	-	-	-	-	-	3.8-7.5	-	-	-
Europe (Adriatic Sea) and USA		-	-	-	-	-	<1000	-	-	-	-	-	-
Queensland, Australia		-	-	-	-	-	-	<17	<0.4	<11	2-24	<13	<9
Global shale		-	-	-	-	-	-	13	0.3	19	90	45	68
HMWB-ROM 5.1													
Reference conditions		<0.6	<600	<500	-	-	-	<40	<0.5	-	<100	<50	<50
Security thresholds		4	3600	1200	-	-	-	200	5	-	1000	400	400
Sediment Quality Guidelines (SQGs)													
Spain	ERL	-	-	-	-	-	-	7.5	0.2	-	25.6	27	21
(Basque country)	ERM	-	-	-	-	-	-	16	1	-	58.6	82.5	36
	TEL	-	-	-	-	-	-	13	0.4	-	35.1	40.4	25.8
	PEL	-	-	-	-	-	-	26.3	1.8	-	66.5	95.5	38.7
Spain	A	-	-	-	-	-	-	35	1.2	-	140	70	30
(CEDEX)	B	-	-	-	-	-	-	70	2.4	-	340	168	63
	C	-	-	-	-	-	-	280	9.6	-	1000	675	234
France	ERM	-	-	-	-	50-200	-	-	-	-	-	-	-
Norway	Class I	-	-	-	-	-	-	<20	<0.25	-	<70	<35	<30
	Class II	-	-	-	-	-	-	20-52	0.2-2.6	-	70-560	35-51	30-46
	Class III	-	-	-	-	-	-	52-76	2.6-15	-	560-5900	51-55	46-120
	Class IV	-	-	-	-	-	-	76-580	15-140	-	5900-59000	55-220	120-840
	Class V	-	-	-	-	-	-	>580	>140	-	>59,000	>220	>840
USA	ERL	-	-	-	-	-	-	8.2	1.2	-	81	34	20.9
	ERM	-	-	-	-	-	-	70	9.6	-	370	270	51.6
Canada	LEL	1	550	600	-	-	-	6	0.6	-	26	16	16
	SEL	10	4800	2000	-	-	-	33	10	-	110	110	75
China	Grade I	-	-	-	-	-	-	20	-	-	80	35	-
	Grade II	-	-	-	-	-	-	65	-	-	150	100	-
General (antifoulants)	ERL	-	-	-	-	-	-	-	-	-	-	-	-

Table 4. Quality categories (good, moderate, bad) assigned for each selected stressor on the basis of a global assessment of the background values, references conditions and security thresholds from programme ROM 5.1, and Sediment Quality Guidelines (SQGs) obtained from the literature (see Table 3)

Sediment stressor	GOOD	MODERATE	BAD
TOC (%)	<1	1-3	>3
N (mg/kg)	<500	500-3500	>3500
P (ppm)	<500	500-1100	>1100
S (ppm)	<1500	1500-8000	>8000
Hydrocarbons (ppm)	<50	50-200	>200
Faecal coliforms (cfu/g)	<100	100-1000	>1000
As (ppm)	<20	20-100	>100
Cd (ppm)	<0.6	0.6-5.0	>5.0
Co (ppm)	<10	10-20	>20
Cr (ppm)	<70	70-100	>100
Cu (ppm)	<50	50-100	>100
Ni (ppm)	<20	20-100	>100
Pb (ppm)	<50	50-100	>100
Zn (ppm)	<100	100-500	>500
Irgarol (ng/g)	<1.0	1.0-2.5	>2.5

Table 5

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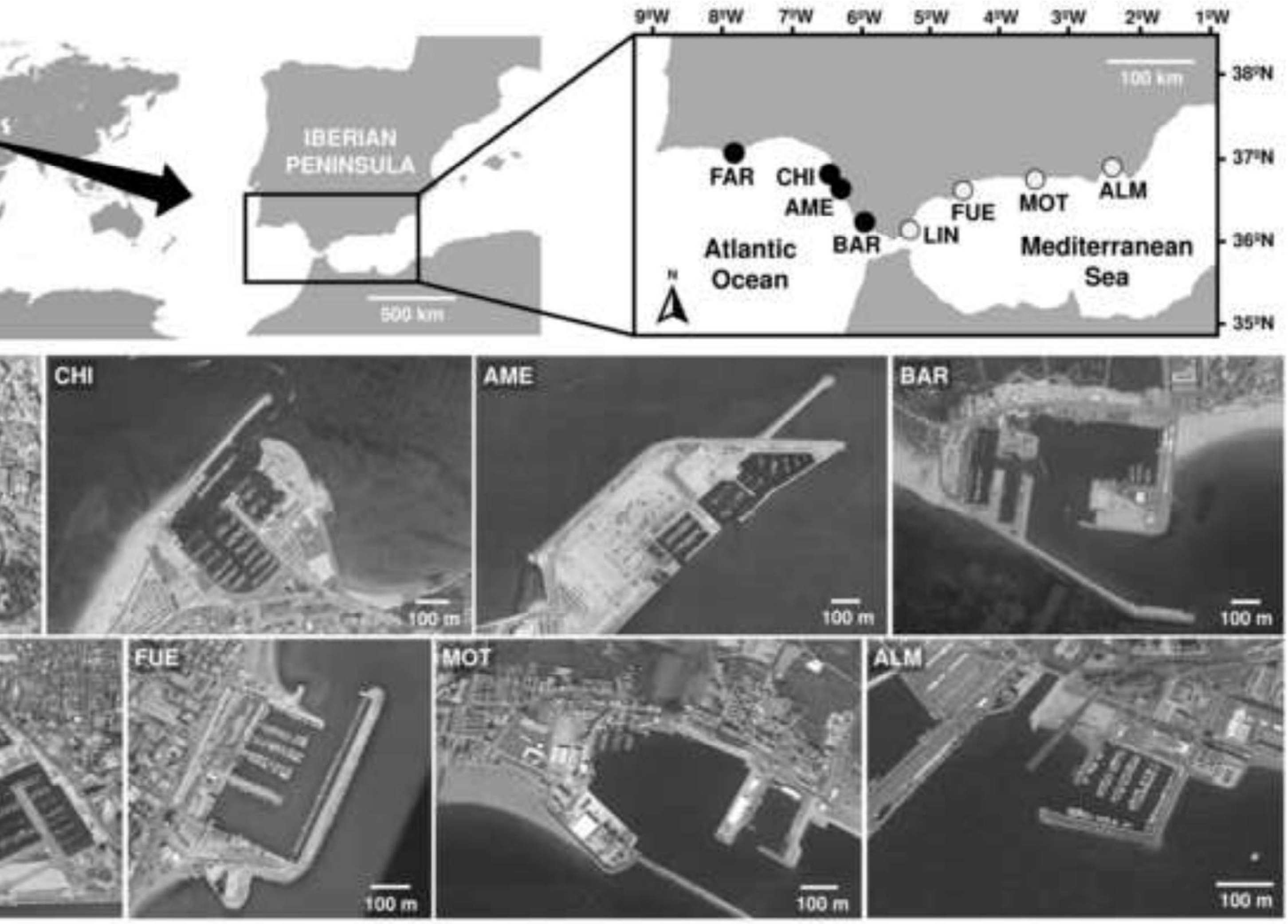
Table 5. Sediment environmental parameters measured in worldwide marinas. IP, Iberian Peninsula; NS, Nova Scotia; T
n.d., not detected (Number of marinas of each study in parentheses).

Marinas (number)	Silt+clay (%)	TOM (%)	N (%)	Hydroc. (mg/g)	Al (mg/g)	Fe (mg/g)	As (ppm)	Cd (ppm)	Cr (ppm)	Cu (ppm)	Hg (ppm)	Mn (ppm)	Ni (ppm)	Pb (ppm)	(p
Europe:															
Southern IP, Spain/Portugal (8)	22-90	3-27	0.1-0.2	1.5-6.5	8-50	15-36	9-23	0.1-0.5	7-115	58-154	-	77-501	11-211	16-39	22
Saladillo-Algeciras, Spain (1)	2-75	3-13	<0.2	0.3-4.4	-	-	-	-	-	-	-	-	-	-	
Kosterhavet, Sweeden (2)	-	-	-	-	-	-	-	0.8-1.2	25-95	35-82	-	-	-	18-30	
Port Camargue, France (1)	-	-	-	-	29-50	-	6-15	-	-	10-1497	0-0.8	-	-	8-94	
Brest, France (3)	-	-	-	0.1-1.3	-	-	-	-	-	42-93	-	-	-	-	
Ligurian Sea, Italy (2)	-	2-16	-	-	-	-	5-24	0.0-0.4	10-27	10-208	-	-	8-29	8-54	
Apulian coast, Italy (4)	3-78	1-28	0.1-1.2	-	1-23	-	5-15	0.1-1.0	0-69	3-173	0.1-1.4	-	4-28	5-195	11
Rapallo, Italy (1)	4-70	3-18	-	-	-	-	-	-	-	-	-	-	-	-	
Cagliari, Italy (1)	48	-	-	-	31	34	9	4.1	82	103	-	266	34	144	7
Heraklion, Greece (1)	41	-	-	-	13	20	5	2.2	128	114	-	338	75	25	3
America:															
Craft harbour NS, Canada (31)	-	-	-	-	-	-	1-62	0.1-3.8	1-305	1-220	0-1.8	-	-	1-583	
Chesapeake Bay, USA (1)	58-89	3-36	-	-	-	-	-	-	-	-	-	-	-	-	
Virgin Islands, USA (2)	3-99	1-17	-	-	6-53	11-34	4-22	0.0-0.7	1-16	13-1535	-	71-113	8-14	2-179	
Africa:															
Ceuta, Spain (North Africa) (1)	31-32	6	0.1-0.2	4.3-4.9	16-33	23-36	20-25	n.d.	46-63	128-139	-	240-332	32-44	100-109	
El Kantaoui, Tunisia (1)	15-34	-	-	-	10-14	5-13	1-13	0.1-1.0	24-44	172-193	-	76-91	9-20	1-25	17
Asia:															
Bandar Al Rowdha, Oman (1)	-	-	-	-	-	-	7	0.1	240	55	n.d.	-	710	4	n
Veraval harbour, India (1)	-	-	-	-	-	27-31	-	0.3-0.9	98-175	20-35	-	606-1011	40-64	129-233	
Oceania:															
Clyde estuary, Australia (1)	-	-	-	-	-	-	-	-	-	70	-	-	-	20	

mage



IBERIAN
PENINSULA
500 km
9ºW
8ºW
7ºW
6ºW
5ºW
4ºW
3ºW
2ºW
1ºW
100 km
38ºN
37ºN
36ºN
35ºN
FAR
CHI
AME
BAR
LIN
FUE
MOT
ALM
Atlantic
Ocean
Mediterranean
Sea
N
CHI
AME
BAR
FUE
MOT
ALM
100 m

Figure 2

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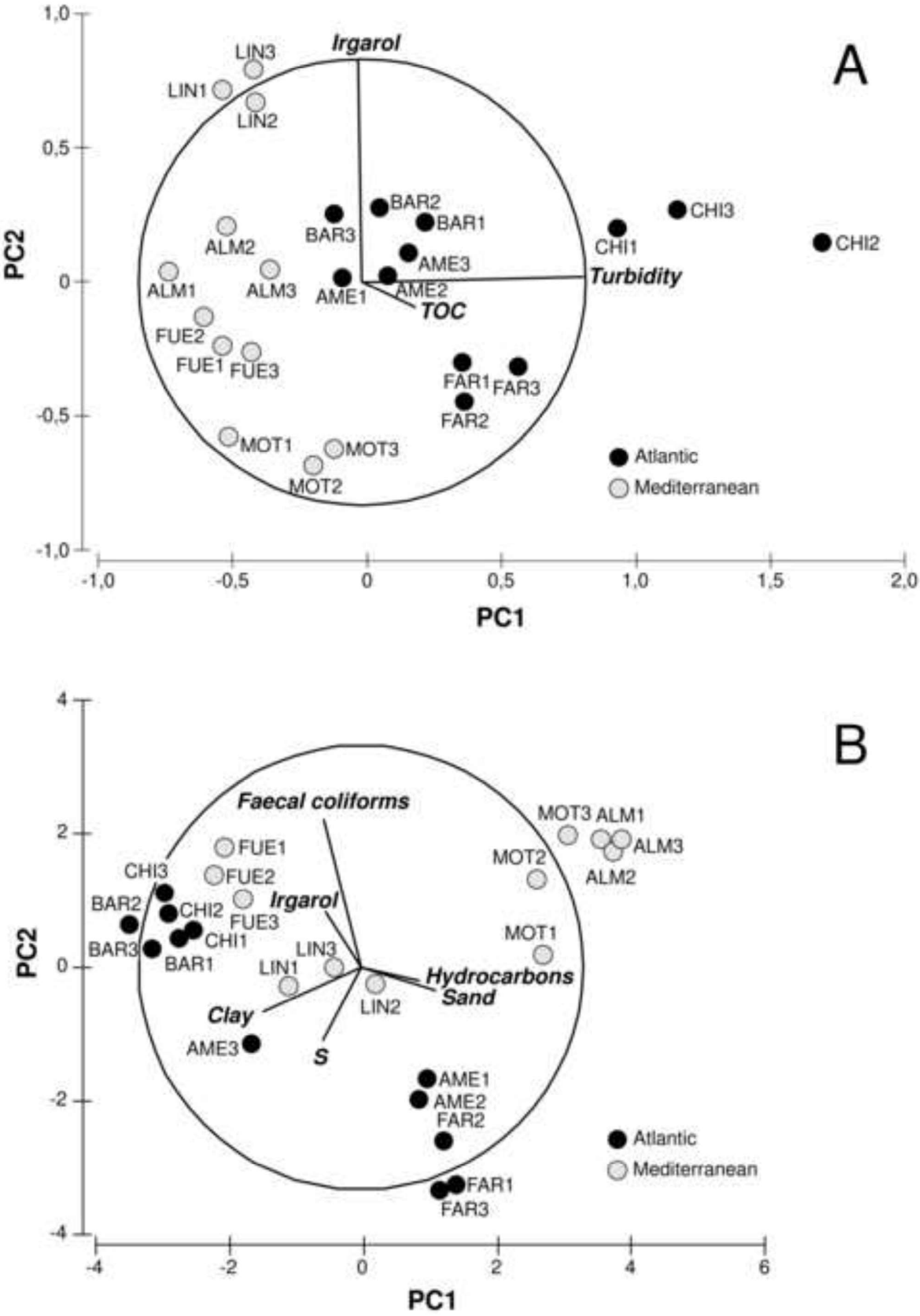

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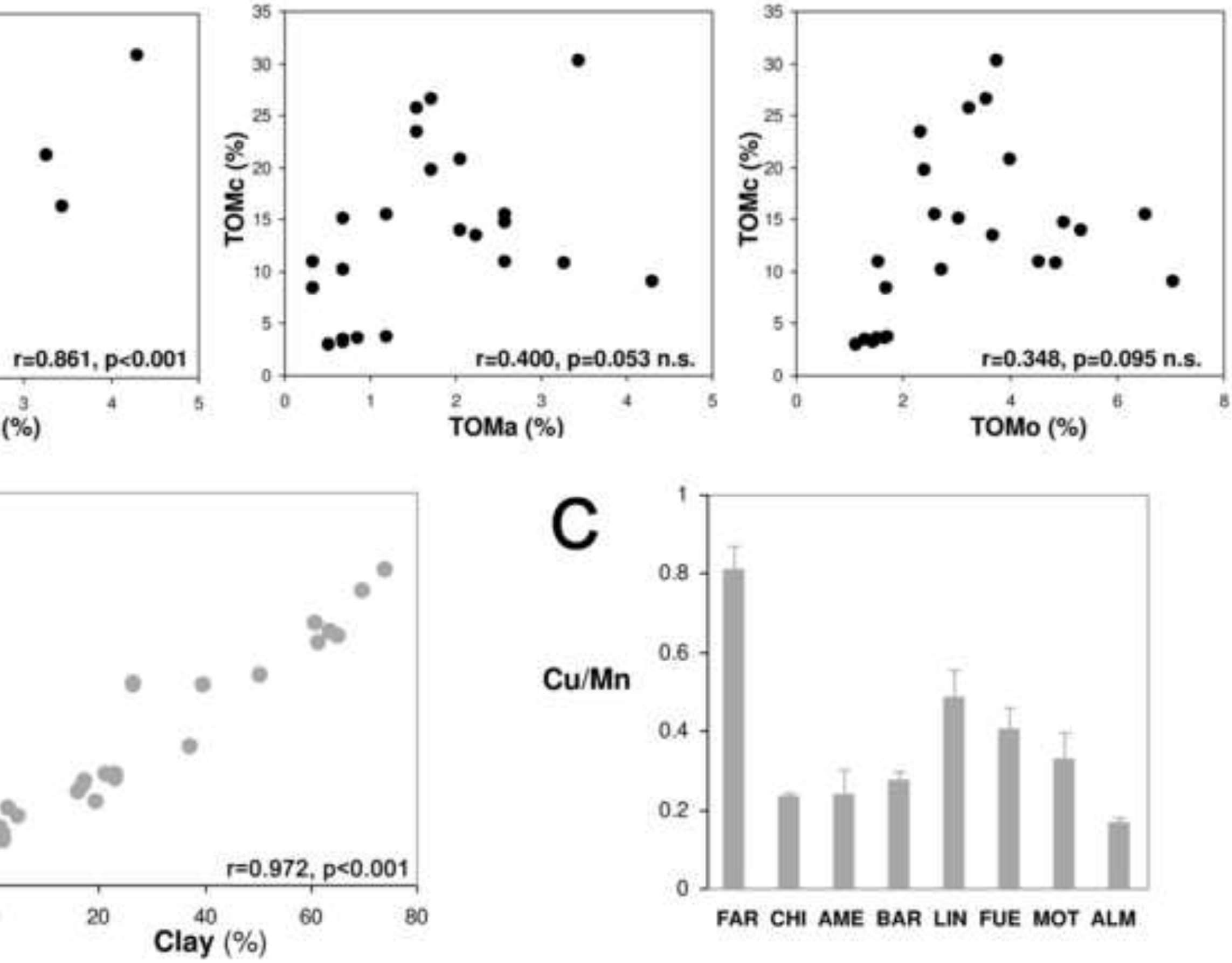
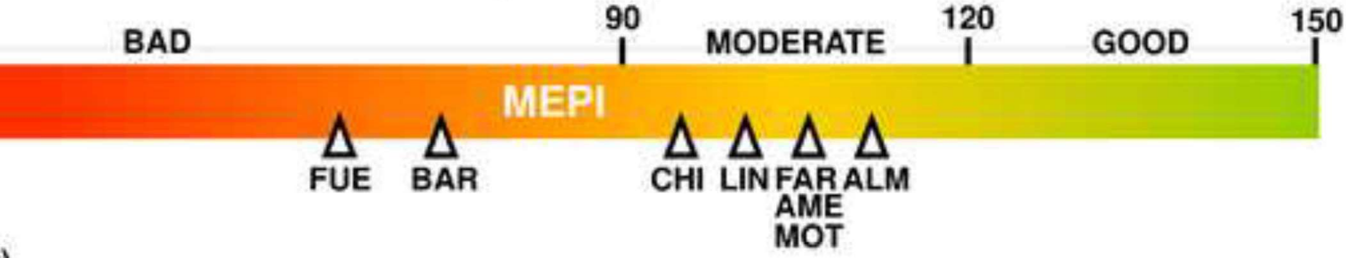


r=0.861, p<0.001
(%)
TOMc (%)
r=0.400, p=0.053 n.s.
TOMa (%)
TOMc (%)
r=0.348, p=0.095 n.s.
TOMo (%)
r=0.972, p<0.001
Clay (%)
C
Cu/Mn
FAR CHI AME BAR LIN FUE MOT ALM

mage

S (ppm)	Hydr. (ppm)	F.col. (cfu/g)	As (ppm)	Cd (ppm)	Co (ppm)	Cr (ppm)	Cu (ppm)	Ni (ppm)	Pb (ppm)	Zn (ppm)	Irgarol (ng/g)	MEPI SCORE
11127	2633	100	12	0.2	3.5	22	62	12	16	67	1.3	105
1927	1767	1250	17	0.1	8.7	50	93	26	35	148	1.5	95
3346	1933	150	10	0.2	5.9	33	74	15	34	589	0.6	105
3191	1500	1483	17	0.1	10.3	82	136	30	24	166	6.7	70
6809	6467	1017	9	0.5	4.7	61	69	39	22	91	1.3	100
6988	1967	1050	23	0.3	17.9	115	154	211	37	192	0.2	60
1496	4567	567	16	0.3	9.6	15	91	24	32	136	3.6	105
1165	2167	1300	16	0.3	7.3	7	58	18	39	133	1.4	110

r in each marina (see Table 4): 10 points 5 points 0 points



)

ERATE)

Figure 5

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