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CoAStal and marine waters integrated monitoring systems for ecosystems proteCtion AnD managemEnt

CASCADE

Project ID: 10255941

Priority Axis: Environment and cultural heritage

Specific objective: Improve the environmental quality conditions of the sea and coastal area by use of sustainable and innovative technologies and approaches

D2.2.9

Submission of at least 2 open-access peer-reviewed and/or conference papers.

Part 2

PP in charge: All PPs

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Preliminary Study on Electrochemical Ion Imprinted Polymeric Film in Sensor Development for Cd(II) Ions Determination in Water †

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Abstract: Preliminary results on an electrosynthesized ion imprinted polymeric film (IIP-film) for Cd(II) ions determination in sensor development are here reported. The sensor was prepared by electropolymerization of 4-aminophenylacetic acid (4-APA) monomer in presence of Cd(II) ions, which acts as the template. The screen-printed carbon electrodes (SPCE) were used as transducer during sensor development, whereas the cyclic voltammetry (CV) and differential pulse voltammetry (DPV) were selected as the electrochemical methods for the synthesis and Cd(II) ions sensing, respectively. The incubation of the developed sensor in NaOH 250 mM involved into remove the template and the formation of specific recognition cavities into the polymer. A multivariate optimization based on central composite design (CCD) was employed to study the effect of three independent parameters on electrochemical performances of the sensor. The electrochemical characterization of sensors was performed in ferrocyanide-ferricyanide redox couple and in KCl 0.1 M, the latter revealing redox properties from the polymeric film. The performances of sensors and the control (non-imprinted film, NIP) was observed in sodium acetate buffer (100 mM, pH = 5) over the Cd(II) concentration range 0.1–10 μM.

Keywords: ion imprinted polymer; 4-APA; electrochemical sensor; Cd(II) ions; electropolymerisation

1. Introduction

Heavy metals pollution refers to a global issue, due to the high toxicity and dangerous effects on environment and human health. Among heavy metals, cadmium is one of the most toxic. Main sources of this ion in environment is industrial wastewater, fertilizers and so on. Currently, the most analytical method for cadmium detection is represented by atomic adsorption spectroscopy (AAS), inductively coupled plasma-mass spectroscopy (ICP-MS) and inductively coupled plasma atomic emission spectrometry (ICP-AES). Those techniques are sensitive, accurate but also expensive, and on-site determination of targets is not so suitable. Due to the complexity of those instrumentation, there is the need to point different methods to be available for on-site determination. Electrochemical methods can be used for that. Moreover, diverse electrochemical methods are explored today for the determination of heavy metals in water environment [1].

Imprinted polymers define robust and artificial materials able to mimic recognition processes of such analytes, such as proteins, small molecules, or ions [2]. The process results in the selective formation of ion-sized imprinted cavities, which are complementary to a specific template in terms of its functional groups. These materials can be easily applied to identify, monitor and remove the target ions in water environment [3]. In this view, the ion imprinted polymers (IIPs) can be described. Their synthesis can be carried out both chemically and electrochemically. The latter leads to the preparation of imprinted films, which are compatible in conjunction with transducers in sensor development [4,5]. Very few works report the electrochemical synthesis of ion imprinted polymers and their application as sensors for metal ion detection [6,7]. With this regard, we propose the synthesis, optimization, characterization and subsequent application of an electrosynthesized IIPs for the electrochemical detection of cadmium (II) in water. The proposed sensor was prepared by electropolymerization of 4-aminophenylacetic acid (4-APA) monomer in presence of Cd(II) ions, which acts as the template. The screen-printed carbon electrodes (SPCE) were used as transducer during sensor development, whereas the cyclic voltammetry (CV) and differential pulse voltammetry (DPV) were selected as the electrochemical methods for the synthesis and Cd(II) ions sensing, respectively. The incubation of the developed sensor in NaOH 250 mM involved the removal of the template and the formation of specific recognition cavities into the polymer. A multivariate optimization was employed for studying the effect of three independent parameters on electrochemical performances of the sensor. The electrochemical characterization of sensors was performed in ferrocyanide-ferricyanide redox couple and in KCl 0.1 M, the latter revealing redox properties from the polymeric film. The performances of sensors and the control (non-imprinted polymer, NIP) were observed in sodium acetate buffer (100 mM, pH = 5) over the Cd(II) concentration range of 0.1–10 μM.

2. Materials and Methods

2.1. Materials

Acetic acid, 4-Aminophenylacetic acid (4-APA, 98%), sodium acetate trihydrate, cadmium nitrate tetrahydrate (98%), and ethylenediaminetetraacetic acid (99%) were purchased from Sigma-Aldrich (Italy). Sulphuric acid and sodium hydroxide solutions were commercially available as analytical reagent grade. All reagents were used without further purification. MilliQ water was used for washing the polymeric film after the preparation. Sodium acetate buffer (100 mM, pH = 5).

2.2. Apparatus

CV and DPV measurements were performed using a PalmSens potentiostat equipped with a cable connector (DropSens, Milano, Italy) for screen-printed electrodes. PSTrace was the software to control the instrument and data acquisition. The polymeric film was deposited on screen-printed carbon electrode (SPCE). The SPCEs were composed of three-electrode configuration on a planar ceramic support (3.3 × 1 cm) and they consisted of a carbon disk-shaped working electrode (4 mm diameter), a platinum electrode as counter electrode and a pseudo Ag/AgCl paste electrode as reference electrode. SPCE were commercially available (Metrohm, Milano, Italy).

2.3. Preparation of Electrosynthesized Ion Imprinted Polymer and Non-Imprinted Polymer

The preparation of ion imprinted polymer (IIP) based on poly-4-aminophenylacetic (poly-4-APA) films was performed by cyclic voltammetry (CV) in a potential range between −0.2 and 1.2 V vs. pseudo Ag/AgCl, at a scan rate of 50 mV s^{-1} for 40 cycles in a solution of H_2SO_4 0.5 M containing 1 mM of Cd^{2+} ions. The porogen was chosen based on previous works about the electrosynthesis of poly-4-APA on SPE [8]. After the electropolymerization, the electrode was rinsed with MilliQ water and incubated in different solvent (EDTA 100 mM and 250 mM, H_2SO_4 500 mM, NaOH 100 mM and 250 mM) to remove the target. The preparation of the control (non-imprinted polymer, NIP) was obtained with the same protocol, but without adding the template into the polymerization mixture.

The treatment in NaOH 250 mM was also performed on NIP. All prepared sensors were taken in air when not in use.

2.4. Cd^{2+} Ion Sensing

The electrochemical responses of IIP and NIP films towards Cd^{2+} ions were recorded using DPV measurements in the potential range of −0.2 to + 0.4 V, modulation amplitude of 50 mV, step potential of 4.95 mV, and equilibration time of 2 s. Cd^{2+} ions interacted with the imprinted film by drop-casting on the electrode surface an appropriate amount (100 µL) of a solution of sodium acetate buffer (100 mM, pH = 5) containing different concentration of Cd^{2+} ions (0.1–10 µM), by leaving the drop on the electrode for 10 min. After each measurement, the electrode surface was gently washed with sodium acetate buffer for 2 min.

2.5. Experimental Design in Optimization Studies

Multivariate optimization was conducted with the light to optimize the development of IIPs and NIP. The selected optimization model was the central composite design (CCD), which allowed the selection of main three factors affecting the development of the sensors, such as (i) the monomer concentration, (ii) the rate between template-monomer (mainly affecting the number of cavities on the polymeric network) and (iii) the number of CV cycles during the electrosynthesis. MODDE® Software (Umetrics, version 12, https://www.sartorius.com/en/products/process-analytical-technology/data-analytics-software/doe-software/modde) was used for design, mathematical modelling and optimization. The levels of studied independent variables are listed in Table 1.

Table 1. Levels of independent variables considered in this work.

Variable	Low	High
Monomer concentration (X_1)	0.5	5
Rate Cd^{2+}/monomer (X_2)	1	3
Number of CV cycles (X_3)	10	40

The response was the difference of current (Δi, µA) recorded in ferrocyanide-ferricyanide redox probe before and after the electropolymerization of the different imprinted sensors. Based on CCD principle, the design consisted of 2^k fractional factorial points plus 2^k axial points and 1 center point, where k defines the number of central points (in this case, k = 3). Eighteen experiment runs were conducted, and the second-order polynomial equation consisted of linear, quadratic, and first-order interaction terms is shown below (Equation (1)):

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^{\,2} + \sum_{i=1}^{k} \sum_{j(\neq i)}^{k} \beta_{ij} X_i X_j + \varepsilon, \quad (1)$$

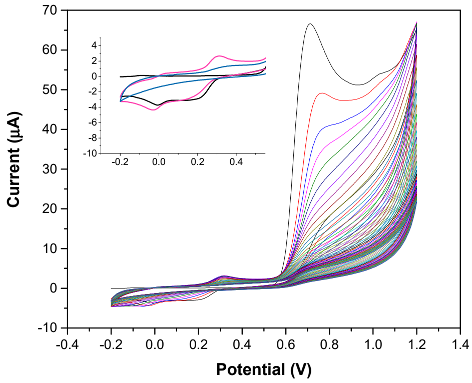
where Y is the response variables, X_i represent the dependent variables, β_0, β_i, β_{ii}, β_{ij} were the regression coefficient for intercept, linear, quadratic and interaction terms, respectively.

3. Results and Discussions

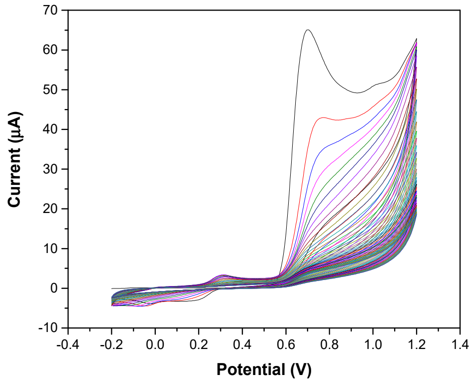
The electropolymerization of 4-APA in presence of Cd^{2+} produced a sensitive polymeric imprinted film for that template, showing superior characteristics against its control. The optimal condition of synthesis was established by employing a multivariate experimental design, and this approach has recently gained interest from scientists regarding optimized sensors and biomimetic sensors. The advantage of using the produced IIPs consisted of revealing a redox property of the polymer, which directly addresses the interaction between imprinted cavities and template. The interaction was visible, close to + 0 V (see related DPV measurements), of which potential is higher than normally observed for the electroactivity of Cd^{2+} in solution.

3.1. Preparation of Electrosynthesized IIP and NIP Films

Figure 1a presented a typical cyclic voltammetry recorded during the electropolymerization of 2.1 mM 4-APA in the presence of 2.1 mM Cd^{2+} ions in 0.5 M H_2SO_4 on a screen-printed carbon electrode. Figure 1b shows the electropolymerization of 4-APA on SPCE, without the template (NIP).



(**a**)



(**b**)

Figure 1. (**a**) The electropolymerization of 2.1 mM 4-APA in the presence of 2.1 mM Cd^{2+} in 0.5 M H_2SO_4. Inset: Focused cyclic voltammetry for (black) 1st, (pink) 2nd (blue), and 17th cycle; (**b**) the electropolymerization of 2.1 mM 4-APA in 0.5 M H_2SO_4. Voltammetric condition: (i) Potential range: −0.2 to + 1.2 V; (ii) scan rate: 50 mV s^{-1}; (iii) CV cycles: 40.

During the CV, the first peak at +0.70 V indicated the formation of cation radicals that promoted the polymerization process, once to the oxidation of 4-APA. Further peaks at +0.19 V and at −0.025 V are related to the reduction of the polymer film on the SPCE surface. Following the second potential cycle, two oxidation waves appeared at the potentials of +0.040 and + 0.305 V, corresponding to the oxidation of the formed polymeric film. After around 17 cycles of polymerization, a decrease in the anodic peaks current was notable, indicating the subsequent formation of the polymer film (see Inset of Figure 1a). Finally, the formation of the film produced a partial blockage of the electrode surface. The electropolymerization of NIP (Figure 1b) followed the same interpretation of the process, with differences in terms of current appeared along the second cycle of CV.

3.2. Optimization of Sensor Performances by Experimental Design

The optimization of performances was possible by a multivariate approach, that considered all factors together, including linear, quadratic and interaction terms in the model. All the selected factors were related to the electrosynthesis process. Among them, with the emphasis to develop imprinted materials, the relationship between all reagent should be described. Preliminary results have shown the factor's importance on responses were the initial concentration of functional monomer (X_1) and the number of CV cycles during the electrosynthesis (X_3). Figure 2 shows the significant coefficients related to factors.

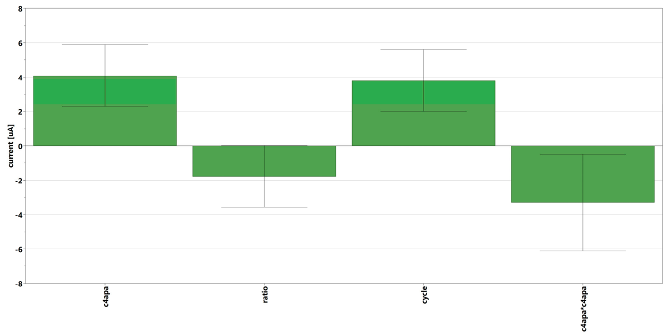


Figure 2. Plot of the significant coefficients obtained from model.

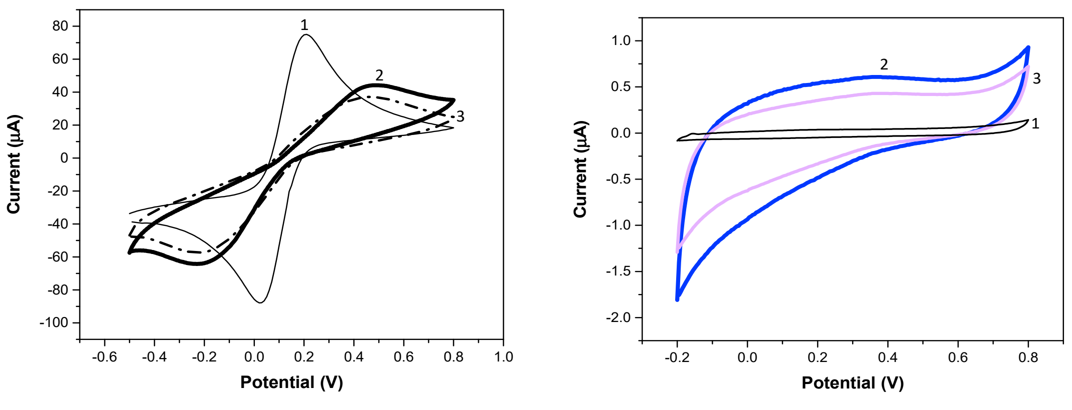
The regression equation for the achieved responses, including significant factors, is reported (Equation (2)).

$$Y = 14.86 + 4.08X_1 - 1.78X_2 + 3.80X_3 - 3.30X_1X_1, \tag{2}$$

After evaluation of Equation (2), it appears as though the monomer concentration is in strong correlation with the other factors. In particular the ratio of monomer/Cd^{2+} should be regulated to assume a correct orientation of cavities on the polymer network. The factor related to the growth of the electrosynthesized imprinted film was also significant, confirming that the deposition of the film on electrode surface is involved in the difference of currents recorded by the electrochemical probe. In light of maximizing the responses, the experimental conditions used for further measurements were (i) 2.1 mM 4-APA, (ii) 2.1 mM Cd^{2+} (ratio 1:1), and (iii) 40 CV cycles during the electrosynthesis.

3.3. Electrochemical Characterization of IIP and NIP Films

The prepared sensors were first subjected to electrochemical characterization in ferrocyanide-ferricyanide redox probe and in KCl 0.1 M, by applying a CV measurement for bare SPCE, IIP film and NIP film after the electrodeposition (Figure 3a,b).



(**a**) (**b**)

Figure 3. (**a**) Electrochemical characterization by CV (fifth cycle) in ferrocyanide-ferricyanide redox probe for (1) bare screen-printed carbon electrode; (2) Cd^{2+}-ion imprinted polymer film/screen-printed carbon electrode (SPCE) and (3) non-imprinted polymer (NIP) film/SPCE after polymerization. Voltammetric condition: (i) Potential range: −0.5 to + 0.8 V; (ii) scan rate: 50 mV s^{-1}; (iii) CV cycles: 5; (**b**) electrochemical characterization by CV (fifth cycle) in 0.1 M KCl for (1) bare screen-printed carbon electrode; (2) Cd^{2+}-IIP film/SPCE; and (3) NIP film/SPCE after polymerization. Voltammetric condition: (i) Potential range: −0.2 to + 0.8 V; (ii) scan rate: 50 mV s^{-1}; (iii) CV cycles: 5.

Both electrochemical characterizations revealed higher electroactivity of the imprinted polymer when compared to NIP film. In addition, as shown in Figure 2b, no signals were obtained for bare SPCE. The electroactivity of IIP film than NIP suggest the imprinting effect of the polymer, where possibly Cd^{2+} ions are possibly able to enhance the overall electrochemical process during polymerization.

The removal of the template ion—to obtain the imprinted cavities—was carried out by exposure of the sensor to different solutions, such as EDTA 100 mM and 250 mM, H_2SO_4 500 mM, NaOH 100 mM, and 250 mM. In all cases, different times of elution were tested, in a range between 1 and 15 min (1, 3, 5, 10, 15 min, respectively). As the most effective method, NaOH 250 mM incubated for 3 min was used. CV markable differences recorded in KCl 0.1 M for NIP and IIPs treated with NaOH 250 mM were visible (Figure 4), confirming the elution of Cd^{2+} ions from the imprinted cavities.

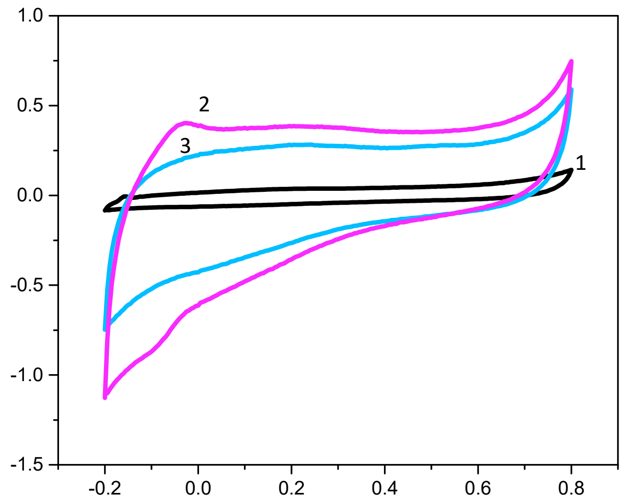
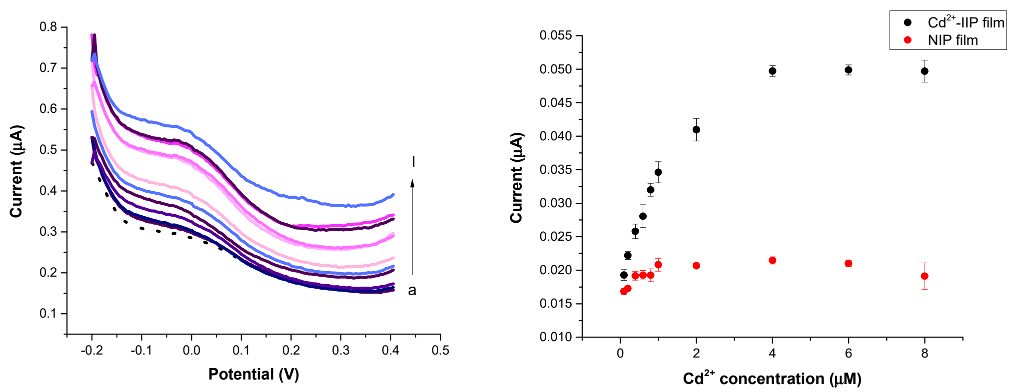


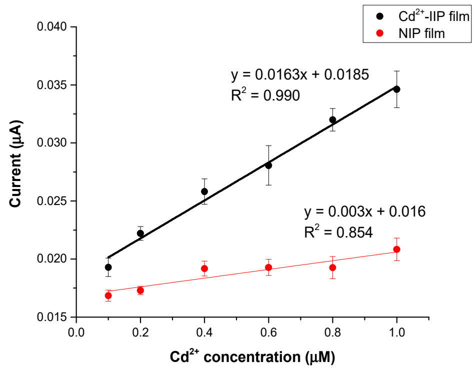
Figure 4. Electrochemical characterization by CV (fifth cycle) in 0.1 M KCl for (1) bare screen-printed carbon electrode; (2) Cd^{2+}-IIP film/SPCE; and (3) NIP film/SPCE after treatment in 250 mM NaOH for 3 min. Voltammetric condition: (i) Potential range: −0.2 to + 0.8 V; (ii) scan rate: 50 mV s^{-1}; (iii) CV cycles: 5.

3.4. Electrochemical Performances of IIP and NIP Film

The electrochemical sensing of Cd^{2+} ions was performed by DPV measurements on NIP and Cd^{2+}-IIP film: 100 mM sodium acetate buffer (pH = 5) was selected as the electrolyte solution for the determination of Cu^{2+} ions. DPV measurements recorded for Cd^{2+}-IIP film are shown in Figure 5, and related calibration curves are also reported (Figure 5b).



(**a**) (**b**)



(**c**)

Figure 5. (**a**) Differential pulse voltammograms recorded for Cd^{2+}-IIP film after the exposure to (a) blank (sodium acetate buffer), (b) 0.1, (c) 0.2, (d) 0.4, (e) 0.6, (f) 0.8, (g) 1.0, (h) 2.0, (i) 4.0, (j) 6.0, (k) 8.0, (l) 10 μM of Cd^{2+} ions in the presence of sodium acetate buffer; (**b**) comparison of the electrochemical responses between Cd^{2+}-IIP and NIP films along all tested Cd^{2+} concentration; (**c**) comparison between responses from Cd^{2+}-IIP and NIP film in the linear range revealed between 0.1 and 1 μM Cd^{2+} ions.

As shown from Figure 5a, the current responses value increased within the tested Cd^{2+} ion concentration. However, saturation reached value upper than 2 μM, due to the occupancy of cavities. Notably, the imprinted sensor shows high affinity and specificity towards Cd^{2+} ions compared to that obtained for NIP films, confirming the imprinting effect on this polymer. The linear regression was established between 0.1 and 1 μM, with a sensitivity of 0.0163 μA μM^{-1}. In addition, it was possible to evaluate the imprinted factor as 6.86, highly indicating the specific recognition of template from imprinted cavities on Cd^{2+}-IIP films.

The proposed imprinted sensor shows high sensitivity and possesses superior specific properties towards Cd^{2+} ions. These preliminary results are currently encouraging us to perform further experiments in regard to selectivity properties of the imprinted polymer against NIP and its application to real water matrices, of which discussion will be presented soon.

4. Conclusions

Preliminary study on electrosynthesis of ion imprinted polymeric sensor on SPCE transducer for Cd^{2+} ion determination in water is reported here. The electrosynthesis of the imprinted cavities revealed the newly approach to produce highly sensitive films towards environmental targets. In this light, the developed imprinted polymeric film shows greater sensitivity than NIP film, with an imprinting factor of 6.86. Those achieved preliminary results open the possibly to employ this sensor for quantitative determination of Cd^{2+} ions in water. Further experiments to evaluate more properties of the sensor are currently under study.

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Conflicts of Interest: The authors declare no conflict of interest.

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Article

Modelling Beach Litter Accumulation on Mediterranean Coastal Landscapes: An Integrative Framework Using Species Distribution Models

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Abstract: Beach litter accumulation patterns are influenced by biotic and abiotic factors, as well as by the distribution of anthropogenic sources. Although the importance of comprehensive approaches to deal with anthropogenic litter pollution is acknowledged, integrated studies including geomorphologic, biotic, and anthropic factors in relation to beach debris accumulation are still needed. In this perspective, Species Distribution Models (SDMs) might represent an appropriate tool to predict litter accumulation probability in relation to environmental conditions. In this context, we explored the applicability of a SDM–type modelling approach (a Litter Distribution Model; LDM) to map litter accumulation in coastal sand dunes. Starting from 180 litter sampling plots combined with fine–resolution variables, we calibrated LDMs from litter items classified either by their material type or origin. We also mapped litter accumulation hotspots. LDMs achieved fair-to-good predictive performance, with LDMs for litter classified by material type performing significantly better than models for litter classified by origin. Accumulation hotspots were mostly localized along the beach, by beach accesses, and at river mouths. In light of the promising results achieved by LDMs in this study, we conclude that this tool can be successfully applied within a coastal litter management context.

Keywords: Litter Distribution Model (LDM); beach litter accumulation; river mouth; coastal dune vegetation zonation; protected areas; Central Adriatic

1. Introduction

The presence of macro litter on sandy beaches and dunes can cause health problems [1,2], economic losses [3,4], and damage to natural biodiversity [5–8]. In light of that, investigating litter distribution and accumulation is a key issue in coastal zone sustainable management [9]. Currently, the amount of waste washed up on coasts is a matter of great concern in Mediterranean countries, and waste mismanagement has been considered one of the most important environmental problems affecting the coastal courtiers of Europe [6,10,11]. The European Commission adopted the Marine Strategy Framework Directive (MSFD, 2008/56/EC), a normative instrument aimed to achieve a 'good environmental status' of European marine and coastal environments [12]. Under the MSFD, Member States are committed to undertake specific actions related to research, monitoring, and surveillance programs to keep "the properties and quantities of marine litter below a threshold above which it could cause harm to the coastal and marine environment" [12].

Marine litter can be categorized according to its source (e.g., households, agriculture, industrial activities, recreational activities, fisheries, shipping [10]) and to its composition (e.g., plastic, glass, paper, aluminum, polystyrene [13]). Overall, the distribution of litter types classified by source is supposed to be mainly related to the presence of anthropogenic structures and human activities [14,15]. On the contrary, the distribution of litter items grouped by material types likely depends on their physical characteristics, e.g., buoyancy and resistance to seawater, as well as their susceptibility to be blown by the wind [9,16].

Several studies looked into the geographical patterns of beach litter [9] and suggested that they are influenced by abiotic and biotic drivers, such as, e.g., vegetation structure [17,18], dune morphology [19], wind, wave action, tides [20], and the density of debris materials [18,21,22]. On the other hand, other studies suggested that marine litter accumulation is mostly influenced by anthropogenic sources [23], identifying polluted rivers [24–26], densely populated urban areas [17,27], seashore mass tourism [25,28], fisheries, shipping, and thriving aquaculture activities [29–31] to promote litter accumulation on the coasts. Although the role of these factors in driving litter occurrence is well known, and the importance of a comprehensive approach for dealing with anthropogenic litter pollution has been recognized [12], only a few studies have investigated the simultaneous effect of these drivers on litter accumulation patterns on the coasts. Moreover, studies integrating geomorphologic, biotic, and anthropic factors are needed to better understand and predict accumulation patterns of beach litter [15,21,25]. In this regard, most of the studies investigating beach litter transportation and accumulation patterns focused on very complex numerical modelling approaches (e.g., [32–35]), which somehow hamper their applicability within a context of coastal zone management aimed at fulfilling MSFD recommendations.

In the fields of ecology and conservation biology, a class of modelling approaches called "Species Distribution Models" (SDMs) [36] designed to quantify the occurrence probability of a species in a given area has been successfully used by scientists and environmental managers (e.g., [37–39]). More recently, SDMs were successfully applied in non-strictly ecological contexts. In fact, such a technique has been used to predict the occurrence probability of roadkill events for wildlife (e.g., [40,41]) or the susceptibility of a given region to landslides [42]. Considering these examples, SDMs might analogously represent an effective tool for predicting litter accumulation probability in relation to given environmental conditions. Being based on statistical relationships between the occurrence of a given event (dependent variable; here, litter fragments) and geographical predictors (explanatory variables), SDMs could be suitable for both assessing litter–environment relationships and for predicting their spatial distribution. Furthermore, quantifying the relationship between observed spatial patterns of litter occurrence and different geographical variables (e.g., geomorphology, vegetation, anthropogenic factors, etc.) can enhance our knowledge about the process of litter accumulation.

In this perspective, we proposed a study aiming to explore the applicability of a SDM–type modelling approach (a Litter Distribution Model; LDM) to map beach litter accumulation in coastal dunes accounting for the simultaneous effect of geomorphologic features, biotic conditions, and anthropogenic pressure. In doing that, we assume litter accumulation is not homogeneous across the coastal landscape but it occurs wherever the combined influence of dune morphology, vegetation pattern, and anthropic pressure is favorable to the litter occurrence. Within this framework, the specific objectives of our study were to (i) calibrate LDMs for litter items classified by source and material type; (ii) compare LDM predictive accuracy for these two main classification schemes; (iii) explore which factors are associated with beach litter accumulation hotspots; and (iv) map such accumulation hotspots.

2. Materials and Methods

2.1. Study Area

The study was conducted along 88 km of the Adriatic coastline in central Italy (Abruzzo and Molise regions; Figure 1), considering six protected areas where Mediterranean coastal dune vegetation is well represented [43]. The analyzed coasts include long, continuous sandy beaches and recent (Holocene) dunes usually occupying a narrow strip along the sea-shore, where the deposition of sediments and organogenic materials is mostly due to wave motion and wind [44,45]. Under natural conditions, these dunes are characterized by a complex sequence of ecosystems that follow the sea-inland eco–geomorphological gradient [46,47], ranging from herbaceous annual plant communities on the strandline lower zone of the beach, crossing through perennial herbaceous vegetation on embryonic and shifting dunes, to patchy Mediterranean scrubs on the inland stabilized dunes, and to *Pinus* sp. woods in the foredunes [48,49]. As for most of the Mediterranean coasts, several tracts of the study area are seriously impacted by human pressure that promotes both land-based [50,51] and ocean-based litter production, movement, and deposition [17,19]. Since the mechanical cleaning of beaches affects the integrity of natural dune eco–morphology [43], beach litter removal in protected areas is done with low frequency [16]. In light of that, protected areas offer an ideal scenario to investigate which environmental, geomorphological, and anthropogenic factors may drive beach litter accumulation.

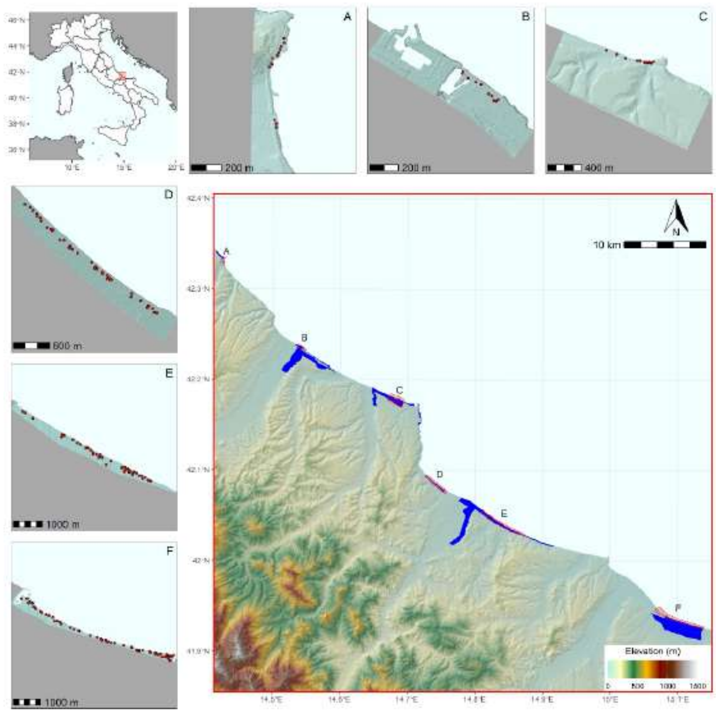

Figure 1. Overall study area and individual protected areas where the models were built: (**A**) Punta dell'Acquabella Regional Natural Reserve; (**B**) Lecceta di Torino di Sangro–Foce del fiume Sangro (SAC IT7140107); (**C**) Punta Aderci–Punta della Penna (SAC site IT7140108) (**D**) Marina di Vasto (SAC IT7140109) (**E**) Foce fiume Trigno–Marina di Petacciato (SAC IT7228221); (**F**) Foce fiume Saccione–Bonifica Ramitelli (SAC IT7222217). The position of each litter sampling plot is indicated by a red dot.

2.2. Beach Litter Occurrence Data

Beach litter sampling was conducted through a stratified random protocol based on 2×2 m survey units [18,27]. Specifically, we randomly placed 180 plots considering, as sampling strata, the EU habitat types, occurring in the study area (RanVegDunes database [48]; Natura 2000 habitat maps, ftp://ftp.dpn.minambiente.it/Natura2000/TrasmissionECE_2013/schede_mappe). This sampling protocol and plot size are particularly effective for describing the highly heterogeneous eco-geomorphology and the fine-scale environmental gradients that characterize coastal dunes [47,52]. Sampling was carried out in the vegetative period (April–May 2018), making sure no mechanical cleaning had been carried out in the study area for several months [30]. All the plots were visited only once by a single team of researchers. Litter items > 2.5 cm (i.e., macro litter [53]) were collected, visually inspected, and recorded following the OSPAR protocol (Convention for the Protection of the Marine Environment of the North-East Atlantic) [54]. The position of each sampling unit was georeferenced using a GPS in order to ensure future monitoring campaigns [30]. Subsequently, we classified litter items according to two macro categories, i.e., material type and origin, following the OSPAR protocol and its successive integrations proposed for the Mediterranean area [55] (Table 1).

Table 1. Macro categories adopted for classifying litter washed up on the analyzed coastal dunes along with the list of items recorded for each category. The number of polluted plots per category and the relative % are also reported.

Material Type	Items	N. Plot	% Plot
PLASTIC	Bottles cups, pull tabs plastic, plastic bottles, plastic drums, fishing nets plastic, plastic plates, plastic forks, plastic bags, plastic sheets, soap containers, snack cards, straws, food trays, packaging of medicines, monofilament lines.	113	38.0
POLYSTYRENE	Polystyrene boxes, polystyrene cups.	88	29.6
GLASS	Glass bottles.	19	6.4
ALUMINUM	Drink cans.	13	4.4
MIXED MATERIALS	Cigarette butts, lighters, fluorescent light tubes, light globes, processed timber, rags, clothing, shoes, hats, tableware, toys, tires and inner tubes, rubber/chewing gum, wires, building materials, nappies, cotton buds, syringes, plasters, sanitary pads, foams, strapping bands, buoys, fishing nets not plastic, fishing related, ropes.	59	19.9
Origin			
CONTAINERS	Bottle cups, pull tabs plastic, plastic bottles $\leq$ 2 L, plastic bottles > 2 L, plastic drums > 2 L, glass bottles.	78	22.1
FISHING AND BOATING	Buoys, fishing nets not plastic, fishing nets plastic, fishing related, monofilament lines, ropes, polystyrene boxes.	100	28.3
FOOD AND BEVERAGE	Drink and food packages, cups, food trays, drink cans, ice cream sticks, chip forks, plastic plates, straws, snack cards, chips bags.	48	13.6
PACKAGING	Foams, papers and cardboard, plastic bags, plastic sheets, strapping bends, soap containers.	50	14.2
OTHER	Fluorescent light tubes, light globes, processed timber, rags, clothing, shoes, hats, tableware, toys, tyres and inner tubes, rubber/chewing gum, wires, building materials. Cigarette packaging, cigarette butts, cigarettes lighters. Sanitary packaging, nappies, cotton buds, syringes, plasters, packaging of medicines, sanitary pads.	77	21.8

We calibrated a different LDM for each material type and origin category. Frequently, occurrence data used in species distribution modelling come from online databases and/or citizen science initiatives. Accordingly, such data may be spatially biased and need to be appropriately filtered (e.g., [41]). Since our modelling exercise relies on litter data gathered through a statistically robust sampling design, we avoided implementing any *ex–ante* data filtering, even though we found it appropriate to test for spatial autocorrelation in LDM residuals [39] in all cases (see below).

2.3. Geographical Covariates

We hypothesized that the occurrence of accumulation areas can be affected by a set of geomorphologic, biotic, and anthropogenic predictors. Starting from airborne LIDAR imagery (acquired in 2008 by the Italian Ministry of Environment, within the PST–A mission), we derived two geomorphological predictors, i.e., curvature and slope, along with a canopy height model (CHM). Indeed, dune morphology (concavity, convexity, height) and vegetation (forests, shrublands or herbaceous plant communities) may likely play a role in influencing litter movement as well as in capturing litter items transported by wind [7,16], thus regulating spatial patterns of accumulation areas. In addition, we considered a number of predictors related to human activity, such as the Euclidean distance from artificial surfaces [56] (derived from the imperviousness products of the Pan-European High Resolution Layers of the Copernicus service; https://land.copernicus.eu/pan-european/high-resolution-layers/imperviousness), from harbors, and direct access to the beach [17,27]. We also included the Euclidean distance from river mouths, hypothesizing a contribution to accumulation areas by litter items transported by rivers [25,27]. The position of harbors, accesses, and river mouths was manually digitalized and their Euclidean distance from each survey unit was calculated. All the predictors were rasterized at a spatial resolution of 2 m (i.e., the native spatial resolution of LIDAR imagery) and checked for their multicollinearity by posing a variance inflation factor ≤ 3 [57]. All the variable preparation and selection steps were carried out within the R environment [58].

2.4. Litter Distribution Models

We calibrated the LDMs by using the maximum entropy modelling algorithm implemented in MAXENT version 3.3.3k [59]. This algorithm compares covariate values at site localities to a sample of background points to create a map of probability of event occurrence ranging from 0 (i.e., no accumulation probability, in this case) to 1 (highest accumulation probability). Since MAXENT predictions are sensitive to initial modelling settings [60], we tested different MAXENT implementations through the ENMeval R package [58] to find the settings that optimize the trade-off between goodness-of-fit and overfitting [61]. In particular, we tested regularization values between 0.5 and 4, with 0.5 steps. Furthermore, we included the following feature classes: linear, linear + quadratic, hinge, linear + quadratic + hinge, linear + quadratic + hinge + product, and linear + quadratic + hinge + product + threshold [41,61]. Among the resulting 48 combinations, we chose the one reporting the lowest Akaike Information Criterion (AICc) [62]. To calibrate LDMs, a set of 10.000 background points was randomly placed within the six investigated protected areas to describe their environmental and geomorphological characteristics and to represent pseudo-absences. We evaluated LDMs through a spatial block cross-validation scheme relying on the inherent data partition structure due to the protected areas network. Specifically, we calibrated LDMs with data from five out of six protected areas, leaving the held-out data for evaluation purposes. We repeated the procedure by holding out in turn the data from each protected area. The block cross-validation scheme proved able to assess model transferability, i.e., the ability to extrapolate predictions into new areas [63] and to penalize models based on biologically meaningless predictors [64]. The predictive performance of the models was assessed by measuring the area under the receiver operating characteristic curve (AUC [65]), the difference between calibration and evaluation AUCs (AUCdiff [66]), and the true skill statistic (TSS [67]). AUC values range from 0 (models with no predictive ability), to 1 (perfect predictions [68]), while TSS values range from -1 (no predictive ability) to 1 (perfect prediction [67]). We predicted LDMs over the six protected areas included in the analysis and applied the "ExDet" approach [69] to assess the effect of model extrapolation on values of predictor variables lying outside the calibration range (i.e., negative values of the D metric indicate extrapolation). LDM projections were binarized according to three threshold approaches (i.e., "maximize TSS", "minimum training presence", and "10th percentile" [70]) to account for the effect of using different binarization schemes [71]. For each litter class, binary maps obtained under the three thresholding schemes were stacked and summed [72] to obtain spatially explicit predictions of litter accumulation hotspots.

2.5. Hotspot Analysis

The spatial association between litter accumulation hotspots, i.e., areas where multiple litter classes are predicted to occur, and the geographical covariates considered in LDMs was explored by calibrating a Random Forest (RF) algorithm, where the pixel-by-pixel count of each litter source/material obtained by summing binary maps was used as the response variable. We also included the different thresholding schemes as additional categorical covariate to account for the different hotspot maps generated by the three thresholds considered. As RF parameters, we set (i) 1000 decision trees, (ii) number of variables randomly selected at each node equal to two (i.e., the square root of the number of geographical covariates), and (iii) the Gini index as the split rule [73,74]. RF goodness-of-fit was assessed through the out-of-bag R^2 (i.e., the mean prediction error of each RF training sample x_i, using only the trees that did not include x_i in their bootstrap sample [75]). To ease evaluating the association strength between litter accumulation hotspots and geographical covariates, we followed the approach proposed by Igras and Biecek [76] and fitted spline functions through the RF marginal response for each covariate. Then, such splines were used as surrogate covariates in generalized linear models (GLMs) against litter pixel-by-pixel count (i.e., as done for RF models). The statistical significance of each surrogate covariate coefficient quantified in these GLMs allows us to evaluate if the association between litter hotspots and geographical covariates as predicted by RF is strongly supported by the data [76].

3. Results

3.1. Litter Distribution Models

LDMs achieved fair-to-good predictive performance (*sensu* Swets [68] and Landis and Koch [77]), showing AUC values ranging from 0.765 (SD = 0.203) for "packaging" source to 0.874 (SD = 0.130) for "aluminum" material type; AUCdiff values scored from 0.060 (SD = 0.178) for the "food and beverage" origin to 0.035 (SD = 0.150) for the "other" origin, and TSS values ranged from 0.551 (SD = 0.101) for "plastic" material type to 0.844 (SD = 0.085) for "aluminum" material type (see also Table S1). Although LDMs for both litter classification schemes yielded good predictive performances, models for litter classified by material type reported significantly higher TSS values than models for litter classified by origin (Figure 2; Wilcoxon test W = 583, $p < 0.05$). Negligible spatial autocorrelation was found in LDM residuals (Appendix A), and only ca. 11% (SD = 0.031%) of the study area reported a marginal extrapolation effect (ExDet D value = −0.034, SD = 0.013; Figures S1–S10).

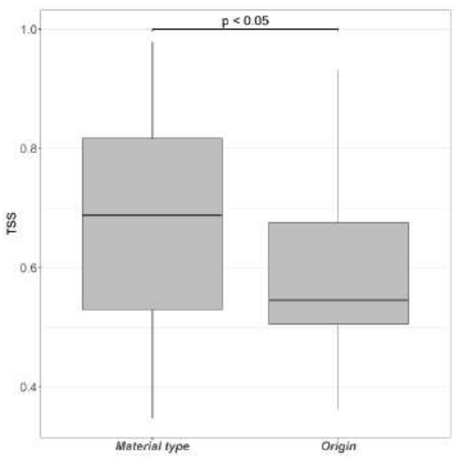

Figure 2. Predictive performances of LDMs for both litter classification schemes according to the True Skill Statistic (Wilcoxon test W = 583, $p < 0.05$).

Most of the litter classes based on both material type and origin classification exhibited similar responses to a specific pool of geographical covariates that reported a variable contribution >10% in almost each LDM. Among the most recurrent responses, litter accumulation probability increased toward low CHM values (e.g., containers, fishing, mixed materials, packaging), close to direct accesses to the beach (e.g., aluminum, plastic) and near to river mouths (e.g., polystyrene, glass), and far from artificial surfaces (e.g., food; Figures S11–S20).

3.2. Hotspot Analysis

Overall, accumulation hotspots hosting the highest number of litter classes (i.e., five) were mostly localized along the beach and embryonic dunes (Figure 3), as well as near direct accesses to the beach and to river mouths (Figure 3E,F; see also Figures S21–S25). Hotspot distribution was not homogeneous within each protected area. For instance, protected areas A and B were largely covered by continuous hotspots potentially hosting all the material types (Figure 3A,B). On the contrary, protected areas E and F include small and isolated hotspots (Figure 3E,F). Considering litter classes by material type, most of the study area was covered by hotspots hosting four litter classes (ca. 700 ha), while hotspots with five litter classes covered ca. 300 ha (Figure 4). Considering litter classes by origin, hotspots with five classes were the widest, covering more than 1000 ha in the study area, whereas the other hotspots occupied around 250 ha (Figure 4).

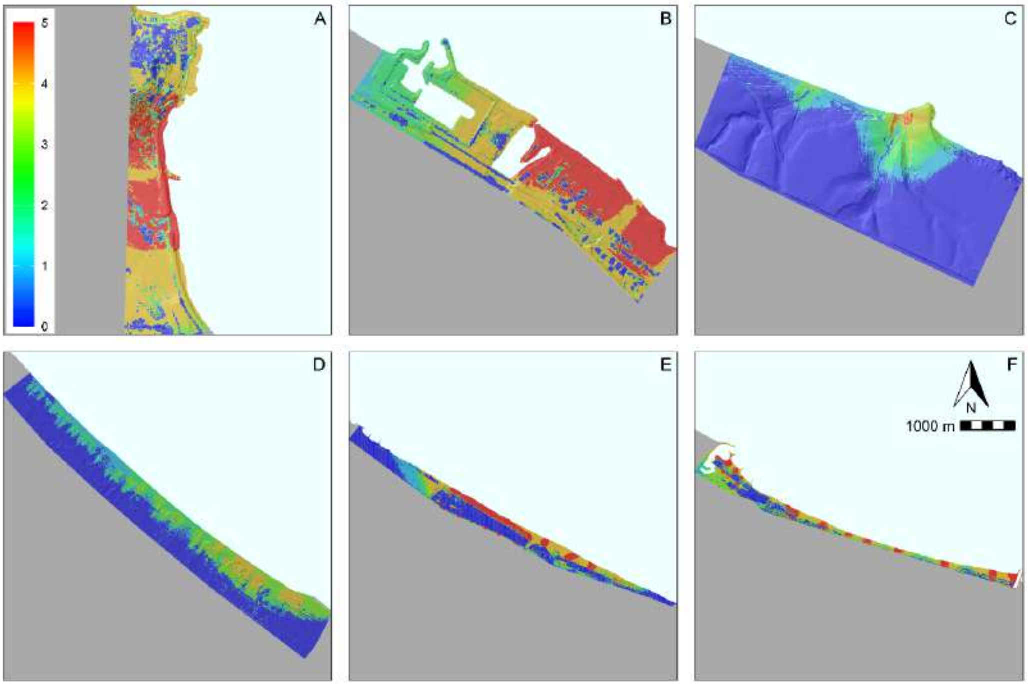

Figure 3. Distribution map of the accumulation litter material-type hotspots identified by LDM projections binarized by the "maximize TSS" threshold. Colors indicate the cumulative number of litter items (e.g., plastic, polystyrene, paper, glass, aluminum and mixed materials) according to the scale on the top left. (**A**) Punta dell'Acquabella Regional Natural Reserve; (**B**) Lecceta di Torino di Sangro–Foce del fiume Sangro (SAC IT7140107); (**C**) Punta Aderci–Punta della Penna (SAC site IT7140108) (**D**) Marina di Vasto (SAC IT7140109) (**E**) Foce fiume Trigno–Marina di Petacciato (SAC IT7228221); (**F**) Foce fiume Saccione–Bonifica Ramitelli (SAC IT7222217).

RF models achieved an excellent goodness-of-fit for both material type and origin hotspots, reporting out-of-bag R^2 values of 0.974 and 0.965, respectively. For both litter classification criteria, RF models highlighted CHM and distance from river mouths and direct accesses to the beach as the most important covariates driving accumulation hotspot occurrence (Figure 5 and Figure S26). Specifically, the count of litter classes increased close to vegetation with a low canopy height (e.g., shrubs) and near to river mouths and direct accesses (Figure 5). GLM results indicated all the surrogate covariates fitted through the RF marginal response were significantly associated with the count of the litter classes (Tables S2 and S3), suggesting strong statistical support of the RF results.

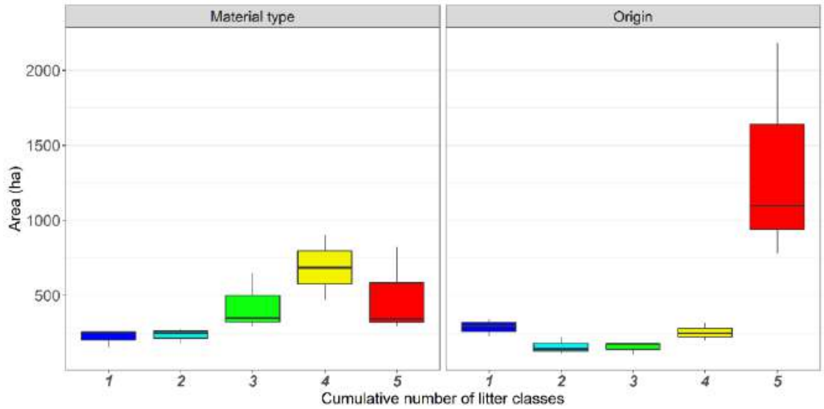

Figure 4. Extension in hectares of the mapped accumulation hotspots of litter classes. Numbers and colors indicate the cumulative number of litter classes (e.g., plastic, polystyrene, paper, glass, aluminum, and mixed materials). For hotspot distribution, see Figure 3.

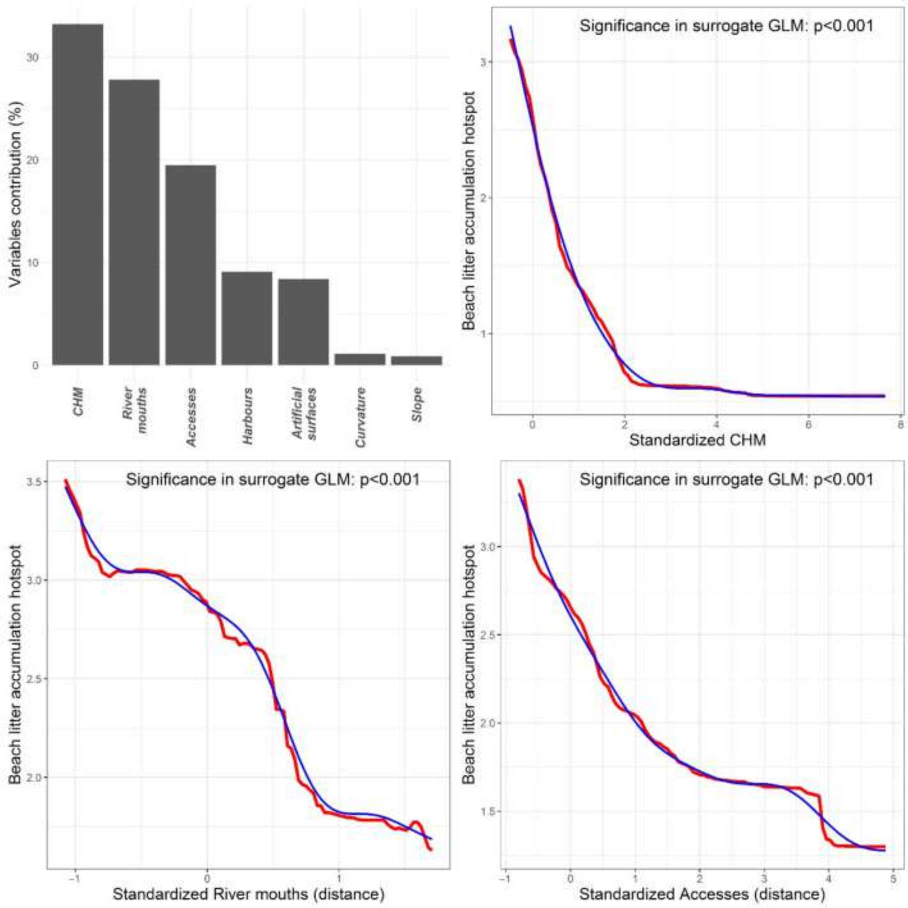

Figure 5. Bar plot depicts the relative contribution of the different covariates in driving the accumulation hotspots of litter material types. Line plots report the shape of the relationship between the count of litter classes in hotspots and the three most influential covariates. Red lines refer to the marginal response as predicted by RF models, while blue lines indicate the spline curves fitted through such responses and used as surrogate covariates in GLMs.

4. Discussion

In the present study, we showed how a modelling approach such as SDM can be effectively used to map beach litter accumulation patterns in a given area. In fact, LDMs achieved good accuracy levels in predicting beach litter occurrence along the Central Adriatic coastal dune ecosystems, explicitly integrating the influence of geomorphologic features, biotic conditions, and anthropogenic pressure in a single analytical framework. Furthermore, we showed that classifying litter items by their material types yielded more accurate LDMs than models calibrated from litter grouped by origins. Interestingly, our results highlighted that the influence of geographical variables on litter accumulation patterns varies across the coastal landscape. Specifically, debris occurrence probability increased toward dune vegetation with low canopy height values, close to direct accesses to the beach and to river mouths, and far from artificial surfaces, thus appearing jointly driven by context-specific biotic conditions, geomorphologic features, and anthropogenic factors. A similar effect was observed for plant canopy height and distance from river mouths and direct accesses in shaping litter accumulation hotspots.

4.1. Alternative Litter Classification Schemes Generate Different Predictive Accuracies

We found LDMs from litter classified by material type (e.g., plastic, polystyrene, glass, paper, aluminum, etc.) achieved good predictive performance according to both AUC [68] and TSS [77], suggesting their occurrence patterns are closely associated with the geographical covariates included in the models. This evidence might likely be related to the specific buoyancy and weight of each material type, which influence the transport dynamics mediated by wave and wind, resulting in similar accumulation patterns. Along the Mediterranean coasts, waves and tides usually drop off beach litter along the drifting line, embryonic, and mobile dunes [17,27]. These act as a sort of "source area" from where litter can be blown farther inland [7] until perennial herbaceous vegetation of shifting dunes and shrubs of fixed dunes stop and entangle the litter [26]. On the other hand, heavy materials tend to remain in place and be trapped in/under sand of the shifting dunes [78,79]. Geographical covariates, e.g., geomorphological features and plant canopy height, which we included in LDMs, were able to approximate such dynamics, therefore adequately explaining geographical variation in litter occurrence patterns and leading the models to perform well. While LDMs from litter classes by origin achieved overall acceptable performance (fair-to-good according to Swets [68] and Landis and Koch [77]), their accuracy was significantly lower than that for models for litter classes by material type. While the influence of the source on the litter accumulation pattern at the coarse scale is well documented in the literature [23,30], classifying litter items according to their origin puts a less direct focus on their buoyancy and weight, which likely represent the characteristics that mostly interact with geographical covariates. As a result, the modelled relation between the occurrence of litter classes by origin and such covariates was weaker. That said, we cannot entirely exclude that the lower accuracy of litter origin LDMs might rather depend on an improper split of the litter classes within the origin criterion.

4.2. Geographical Factors Affecting Litter Accumulation Hotspots

In the Mediterranean dunes system analyzed in this study, biotic elements such as vegetation height showed a predominant role in shaping litter accumulation patterns, confirming similar evidence documented by previous research [7,56]. Actually, litter accumulation patterns seem to follow the sea–inland gradient that characterizes the Mediterranean coastal dunes [17]. In particular, litter occurrence was strongly related to low–medium values of canopy height (e.g., containers, fishing, mixed materials, packaging), indicating a higher accumulation across the foredune vegetation zonation. Most of the heavy litter remains in the aphytic zone and on the strand line [80], as well as a small part of light litter that is trapped by organic waste as woody posts [7]. Moving toward inner sectors occupied by perennial herbaceous vegetation on embryonic and shifting dunes, litter items tend to be partly entangled by grasses and tufts, whose root structure is particularly able to fix and stabilize sediments [81,82], thus leading to litter accumulation and burial. Light litter items (e.g., polystyrene) can be further blown up toward fixed dunes, where most of them are curtailed by the pioneer scrubs of Mediterranean maquis [27]. Therefore, our results highlighted as a novel finding that not only the seashore and embryonic dunes but also foredunes, with perennial herbaceous and woody vegetation, are hotspots of litter accumulation across low-lying coastal areas. The perennial vegetation works as a "flying plastic litter sink" and a biological barrier that greatly reduces the accumulation of these items in the tall pine and evergreen oak woodlands growing in the adjacent inner sectors. Such an "uncommon" pattern is likely related to the dune characteristics in the study area. In fact, closed seas as the Adriatic are more protected from prevailing westerlies than open sea basins. Therefore, coastal dunes form "low-lying" systems [45] that are particularly prone to a sparse litter accumulation pattern, able to invade multiple dune sectors. Unfortunately, the litter removal in dense grasses and pioneer maquis is too hard to do and therefore polystyrene and plastic bags trapped under the branches become persistent and cumulative over time [16]. Further, the "flying plastic litter sink" vegetation is included in habitats of European concern (Directive 92/43/CEE) and is inhabited by several animal species included on the Red List such as the Kentish plover (*Charadrius alexandrinus* [83]) and the Hermann's tortoise (*Testudo hermanni* [84]).

As similarly observed along oceanic coasts [85], on Mediterranean seashores, the river discharge into the sea contributes to beach litter accumulation by providing a variety of litter items coming from inland alluvial plans. In fact, the prevalent occurrence of litter hotspots near river mouths evidenced by our results is coherent with well-established literature reporting litter accumulation to be favored near coastal rivers [19], which act as pathways of terrestrial mismanaged waste to the sea [25,85]. Litter items discharged by rivers can then be reshuffled and deposited on the seashore by sea currents and tides, therefore remaining mostly close to river mouths [34].

The direct accesses to beaches emerged as another important determinant of beach litter accumulation hotspots, as well as of several single litter items (e.g., aluminum, food, and plastic; see Figures S11, S14 and S18). Such rather intuitive evidence is supported by several studies showing how small roads and trails increase the potential number of visitors frequenting the seashore, which in turn increase litter droppings [86]. It is widely documented how recreational activities represent one of the main factors behind beach litter accumulation [25,30,87]. Their effects appear even worse in protected natural areas due to an overall poor ecological awareness of the tourists. Moreover, in such areas, it often necessary to walk long distances before reaching the closest cans to deposit garbage [29,88].

Although not resulting among the most relevant covariates, we reported an interesting effect of the distance from urban areas. While several studies in the Mediterranean basin reported a higher marine litter abundance close to populated and industrialized areas [89], as well as a clear relationship between growing populations in coastal cities and increasing coastal debris accumulation [90], our results showed an inverse relationship (see e.g., Figures S11–S20). Interestingly, more recent papers found similar evidence [25,87,91], interpreting the lower litter amount close to the urban areas as due to an efficient litter collection by the municipal litter agencies. Such evidence might be valid in our case, although we did not have detailed information about the cleaning effort in the study area to confirm this.

Contrary to other similar studies [92], we found no relevant effect played by geomorphological features (i.e., slope and curvature) in shaping litter accumulation patterns. Actually, this seems a reasonable outcome given the characteristics of the costal dune system analyzed in our study. Since such a system is mostly shaped by low-lying dunes [45], it is plausible that its mild geomorphologic features did not have a relevant effect in driving accumulation patterns. That said, we could not exclude *a priori* a role of geomorphology also in our peculiar context. That is the reason why we have included, and suggest to consider, dune morphology among the covariates in LDMs.

4.3. Management Implications

The integration of geomorphologic features, biotic elements, and anthropogenic pressure into LDMs allowed us to better investigate litter occurrence patterns and yielded accurate spatially-explicit predictions of litter accumulation hotspots along the analyzed coast. Incorporating ecological modelling tools into a coastal zone management context offered a new possibility of mapping areas with high pollution hazard, which can support site-specific management actions. In fact, a better understanding of the effect played by the individual factors behind litter accumulation, along with the production of debris accumulation maps, represent essential steps for developing effective strategies coherently to those committed to by the MSFD. From such a perspective, the strong geographical variability in litter accumulation patterns in the protected area network analyzed in this study supports the approach of prioritizing the implementation of tailor-made measures to control, monitor, and prevent the formation of litter hotspots, in order to achieve good coastal environmental status. For instance, in the analyzed low-lying coastal areas which are particularly prone to storm surges [45] that wash up important amounts of litter reaching embryonic dunes, periodic manual cleaning of the driftline is advisable. The observed hotspots of items blown by the wind to fixed dunes and trapped on perennial vegetation evidenced a paucity of such periodical cleaning campaigns. In such cases in which waste collection is occasionally possible, cleaning work must be preferentially done in litter hotspot areas.

5. Conclusions

The modelling framework implemented in this study represents a new, promising tool in the context of coastal zone management, able to explicitly integrate several factors notoriously involved in litter accumulation patterns. In keeping with an ever wider tendency of exporting SDM toward different fields, we explored if and how this statistical tool, which is highly popular among ecologists, might perform in predicting litter accumulation occurrence. We cannot exclude that adding some specific predictors (e.g., waves, tides or currents) involved in litter transport, especially in the water, might have improved the obtained predictions. Unfortunately, we did not have mapped information on such parameters at a sufficiently high detail along the study coastline to be included in our models. Therefore, we assumed a preponderant effect of dune morphology, vegetation pattern, and anthropic pressure on the modelled litter accumulation patterns. While we are aware of this basic assumption, we found it acceptable especially given our specific focus on terrestrial litter accumulation (i.e., dunes and river mouths). That said, it would surely be worthwhile to adapt our approach in future exercises focusing on marine accumulation. Despite such limitations, our study showed that, when fed with highly accurate litter occurrence data combined with geographical covariates at extremely high resolution (e.g., LIDAR products), LDMs are able to predict litter accumulation with good predictive performance. In addition, this approach allowed exploring the differential role in litter accumulation played by factors such as geomorphology, vegetation, and human infrastructures, in terms of their magnitude and shape. Different from other, very complex approaches relying on numerical modelling, the outputs of LDMs can widely support preventive and effective measures in litter hotspots to avoid further litter accumulation in coastal EU habitats, where it is detrimental for wild plants and animals and its removal is extremely difficult. In light of the promising results achieved by LDMs as described in this study, we conclude that this tool can be successfully extended to different coastal zone management applications. Notwithstanding, we stress the importance of replicating our approach in different coastal area contexts, in order to further test its reliability.

Supplementary Materials: The following are available online at https://www.mdpi.com/2073-445X/10/1/54/s1, Figure S1: Map of ExDet metric values showing possible extrapolation of LDM predictions for aluminum; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S2: Map of ExDet metric values showing possible extrapolation of

LDM predictions for containers; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S3: Map of ExDet metric values showing possible extrapolation of LDM predictions for fishing and boating; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S4: Map of ExDet metric values showing possible extrapolation of LDM predictions for food and beverage; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S5: Map of ExDet metric values showing possible extrapolation of LDM predictions for mixed materials; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S6: Map of ExDet metric values showing possible extrapolation of LDM predictions for other materials; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S7: Map of ExDet metric values showing possible extrapolation of LDM predictions for packaging; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S8: Map of ExDet metric values showing possible extrapolation of LDM predictions for plastic; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S9: Map of ExDet metric values showing possible extrapolation of LDM predictions for polystyrene; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S10: Map of ExDet metric values showing possible extrapolation of LDM predictions for glass; yellow to red tones indicate low to high extrapolation, while blue tones refer to no extrapolation. Figure S11: Variable importance (bar plot) and response curves describing the shape of the relationship between aluminum accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S12: Variable importance (bar plot) and response curves describing the shape of the relationship between container accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S13: Variable importance (bar plot) and response curves describing the shape of the relationship between fishing and boating accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S14: Variable importance (bar plot) and response curves describing the shape of the relationship between food and beverage accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S15: Variable importance (bar plot) and response curves describing the shape of the relationship between mixed material accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S16: Variable importance (bar plot) and response curves describing the shape of the relationship between other material accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S17: Variable importance (bar plot) and response curves describing the shape of the relationship between packaging accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S18: Variable importance (bar plot) and response curves describing the shape of the relationship between plastic accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S19: Variable importance (bar plot) and response curves describing the shape of the relationship between polystyrene accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S20: Variable importance (bar plot) and response curves describing the shape of the relationship between glass accumulation probability (y axis) and each explanatory variable (x axis); red curves refer to the mean response, while blue ribbons indicate ± 1 SD around the mean. Figure S21: Distribution map of the accumulation litter material-type hotspots identified by LDM projections binarized by the "minimum training presence" threshold. Numbers and colors indicate the cumulative number of litter classes (e.g., plastic, polystyrene, paper, glass, aluminum and mixed materials). Figure S22: Distribution map of the accumulation litter material-type hotspots identified by LDM projections binarized by the "10th percentile" threshold. Numbers and colors indicate the cumulative number of litter classes (e.g., plastic, polystyrene, paper, glass, aluminum and mixed materials). Figure S23: Distribution map of the accumulation litter origin hotspots identified by LDM projections binarized by the "maximize TSS" threshold. Numbers and colors indicate the cumulative number of litter classes (e.g., containers, fishing and boating, food and beverage, packaging, other materials). Figure S24: Distribution map of the accumulation litter origin hotspots identified by LDM projections binarized by the "minimum training presence" threshold. Numbers and colors indicate the cumulative number of litter classes (e.g., containers, fishing and boating, food and beverage, packaging, other materials). Figure S25: Distribution map of the accumulation litter origin hotspots identified by LDM projections binarized by the "10th percentile" threshold. Numbers and colors indicate the cumulative number of litter classes (e.g., containers, fishing and boating, food and beverage, packaging, other materials). Figure S26: Bar plot depicts the relative contribution of the different covariates in driving the accumulation hotspots of litter origin. Line plots report the shape of the relationship between the count of litter classes in hotspots and the three most influential covariates. Red lines refer to the marginal response as predicted by RF models, while blue lines indicate the spline curves fitted through such responses and used as surrogate covariates in GLMs. Table S1. Evaluation metrics reported by LDMs for each litter element. The table includes mean ± standard deviation values referring to cross-validation replicates. Table S2. Coefficients of generalized linear models including surrogate geographical covariates fitted through the random forest marginal response. The RF model was calibrated for litter material-type classification. Table S3. Coefficients of generalized linear models including surrogate geographical covariates fitted through the random forest marginal response. The RF model was calibrated for litter origin classification.

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Appendix A. Checking for Significant Spatial Autocorrelation in LDM Residuals

Appendix A.1. Method

We calculated the models' residuals as 1—predicted probability of the presence for each litter item record, also including the pseudo-absences. Moran's I was calculated considering multiple distances between points, ranging between a minimum distance with each point connected only to its nearest neighbour to a maximum with all points connected. Significance of Moran's I was calculated using a randomization test with 999 Monte Carlo permutations. All procedures were repeated 10 times, each time randomly choosing 1000 pseudo-absences.

Appendix A.2. Results and Conclusions

All of the litter items, classified either by origin or by material type, exhibited a significant but weak correlation. In particular, *Container* litter reported the highest correlation, showing a mean Moran's I value = 0.147 ± 0.501, which was statistically significant in ca. 90% of the replicates. By contrast, *Food and beverage* litter scored a mean Moran's I value = 0.006 ± 0.200, which was statistically significant in ca. 75% of the replicates. Averaged among all the analysed litter items, we reported a mean Moran's I value = 0.009 ± 0.259, which was statistically significant in ca. 70% of the replicates. Such low values allow considering as negligible the degree of correlation of LDM residuals.

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Challenges for Restoration of Coastal Marine Ecosystems in the Anthropocene

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Coastal marine ecosystems provide critical goods and services to humanity but many are experiencing rapid degradation. The need for effective restoration tools capable of promoting large-scale recovery of coastal ecosystems in the face of intensifying climatic stress has never been greater. We identify four major challenges for more effective implementation of coastal marine ecosystem restoration (MER): (1) development of effective, scalable restoration methods, (2) incorporation of innovative tools that promote climate adaptation, (3) integration of social and ecological restoration priorities, and (4) promotion of the perception and use of coastal MER as a scientifically credible management approach. Tackling these challenges should improve restoration success rates, heighten their recognition, and accelerate investment in and promotion of coastal MER. To reverse the accelerating decline of marine ecosystems, we discuss potential directions for meeting these challenges by applying coastal MER tools that are science-based and actionable. For coastal restoration to have a global impact, it must incorporate social science, technological and conceptual advances, and plan for future climate scenarios.

Keywords: coastal marine ecosystems, social-ecological restoration, coral reefs, seagrass, mangrove, oyster reefs, kelp, saltmarshes

BACKGROUND

Humanity is facing serious environmental challenges at the onset of the Anthropocene (Crutzen, 2002; Kareiva et al., 2011; He and Silliman, 2019). The swift decay of natural ecosystems, their biodiversity, and services to humans presents a global challenge (Dobson et al., 2006; Dirzo et al., 2014; Hautier et al., 2015). Coastal marine ecosystems are immensely important for human well-being (Barbier, 2012; Duarte et al., 2013), and they are among those facing the most rapid ecological degradation (Lotze et al., 2006; Duke et al., 2007; Waycott et al., 2009; Beck et al., 2011; Burke et al., 2011; Bugnot et al., 2020), resulting in declines in the goods and services that they provide to society (Cesar, 2000; Barbier, 2012; Costanza et al., 2014).

The decline of many coastal ecosystems and current lack of effective solutions for reversing this trend have triggered growing interest in developing tools for the restoration of degraded marine environments (Edwards, 1999; Elliott et al., 2007; Borja, 2014; Possingham et al., 2015; Kienker et al., 2018; Airoldi et al., 2020). For example, recovering ecosystem structure and function through restoration has recently been identified as one of eight "grand challenges" in marine ecosystems ecology (Borja, 2014). Although significant progress has been made in some coastal systems, notably mangroves, kelp forests, wetlands, seagrass meadows, oyster reefs, and to some extent, coral reefs (Hashim et al., 2010; Beck et al., 2011; Roman and Burdick, 2012; Campbell et al., 2014; van Katwijk et al., 2016; Boström-Einarsson et al., 2020; Eger et al., 2020), restoration science of coastal marine ecosystems lags behind terrestrial and freshwater counterparts (Craig, 2002; Suding, 2011).

Restoration has been defined in multiple ways (Elliott et al., 2007). Here, we use the common definition, "the process of assisting the recovery of damaged, degraded, or destroyed ecosystems" (Hobbs et al., 2004; SER, 2004), which views restoration as a broad term that spans from preventative management aimed at stress relief to full habitat reconstruction. We consider restoration to be an integral part of conservation management (Abelson et al., 2015; Possingham et al., 2015), but the full recognition of ecological restoration as an essential element of coastal marine management (Murcia et al., 2014) will require well-defined and achievable objectives, and reliable cost-effective restoration tools (Bayraktarov et al., 2016). While we acknowledge that progress has been made in developing novel tools for marine ecosystem restoration (MER; e.g., eco-engineering or nature-based solutions; Morris et al., 2019), the increasing rate of degradation of coastal environments emphasizes the need for rapid development of integrative approaches to science-based restoration of marine ecosystems (e.g., Elliott et al., 2007; Abelson et al., 2015; Possingham et al., 2015; Airoldi et al., 2020). An important first step in this process is to identify major scientific, societal and operational gaps in coastal MER, which should help to accelerate the development of more effective, scalable tools and practical approaches for coastal MER. Overall, our goal is to build an effective framework for enhancing the multidisciplinary science of coastal MER via the following objectives: (1) development of cost-effective, scalable restoration tools, (2) use of these tools to promote adaptation of coastal marine ecosystems to cope with climate change and global stressors, (3) integration of social and ecological restoration priorities, and (4) fostering the acceptance and routine consideration of coastal MER as a scientifically credible management tool (**Figure 1**).

DEVELOPMENT OF EFFECTIVE, SCALABLE RESTORATION TOOLS

Many current coastal MER tools (techniques and methodologies) have been criticized for high costs that exceed perceived benefits, often with superficial treatment of symptoms rather than the causes of degradation (Elliott et al., 2007; Mumby and Steneck, 2008; van Katwijk et al., 2016; but see Lefcheck et al., 2018; Reguero et al., 2018). Four common and potentially inter-related methodological problems that can result in coastal MER failure are: (1) lack of clear criteria for success, (2) challenging site selection, (3) inadequate or inappropriate tool selection/availability including scalability commensurate with the scale of degraded habitats, and (4) poorly designed assessment protocols (Suding, 2011; Abelson et al., 2015; Bayraktarov et al., 2016).

Lack of Clear Criteria for Success

The implementation of clear, measurable restoration goals requires quantifiable benchmarks for determining whether or not the stated restoration goals are achieved (SER, 2004; Suding, 2011). Specific criteria used to measure success (such as resilience indicators; Maynard et al., 2015) will by necessity vary depending on project goals and stakeholder interests, and should be specified at the outset. The inclusion of key stakeholders and the institutions they represent is crucial in the framing of restoration strategies and related expectations of the outcomes of the MER effort. Projects are likely to gain wider acceptance if their goals are broadened to include ecosystem services such as coastal protection and job creation (Temmerman et al., 2013; Kittinger et al., 2016; World Bank, 2016) that benefit a variety of aware and connected stakeholders (Coen and Luckenbach, 2000; Abelson et al., 2015; Strain et al., 2019).

Site Selection Issues

Appropriate site selection, especially complicated in increasingly urbanized and fragmented systems, is a major determinant of restoration success (Suding, 2011; Bayraktarov et al., 2016). The selection of sites to be restored should be done carefully with consideration of both ecological (e.g., connectivity among populations) and social (e.g., business plan for long-term stewardship) objectives that can reduce the risk of restoration failures (Abelson et al., 2015; Bayraktarov et al., 2016). Also, restoration should be prioritized in areas where the local stressors responsible for the initial degradation of the site are known and can be reduced to levels compatible with the long-term sustainability of the intervention. In cases of non-manageable stressors, e.g., climate-change effects or heavy boat traffic, a different restoration approach should be applied, which promotes adaptation to cope with climate-change conditions (see section "Promoted adaptation"), or eco-engineering techniques, such as living breakwaters, to insulate against stressors (see New tools, approaches, and conceptual framework, below). If multiple candidate sites are available, then these should be compared by relating past, present and predicted future community states using information on environmental conditions, ambient stressors, risks, biodiversity values, and ecosystem services (e.g., Game et al., 2008; Abelson et al., 2015, 2016a).

Assessment of Achievements

Inadequate funding for well-designed monitoring aimed at evaluating the success of a project in meeting its objectives is

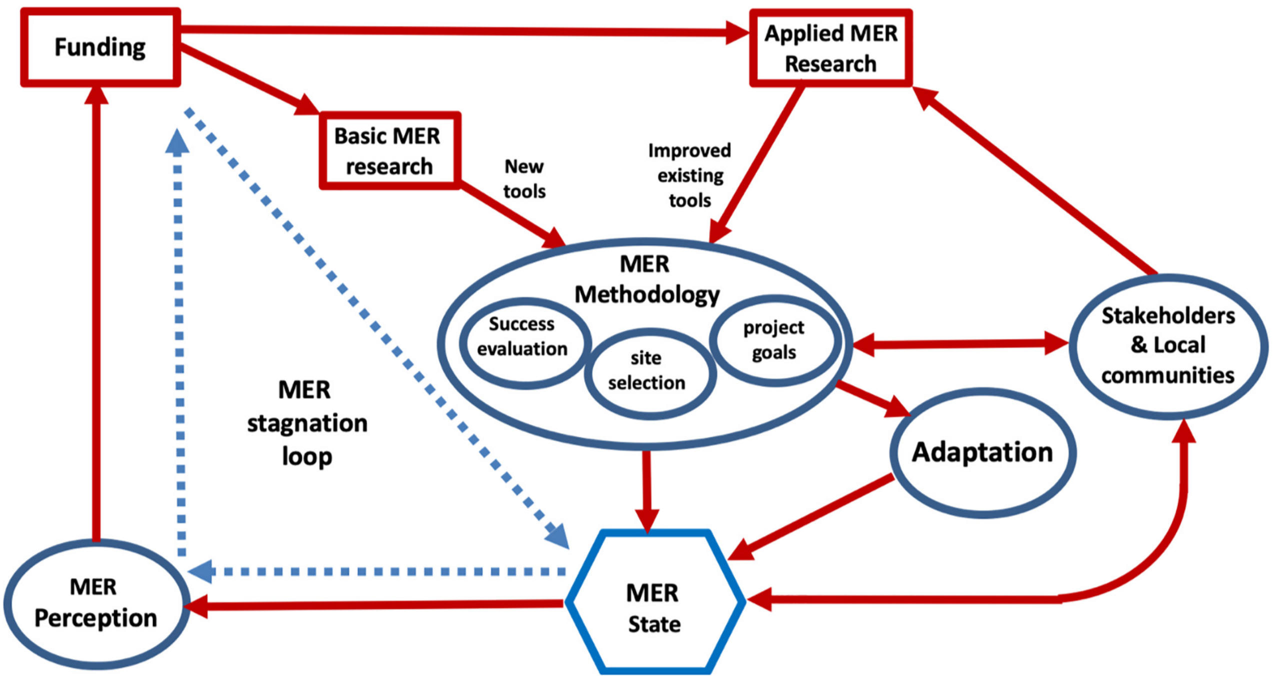

FIGURE 1 | Conceptual framework of the four major coastal Marine Ecosystem Restoration (MER) challenges (i.e., development of tools, adaptation to global stressors, social-ecological integration, and promotion of MER perception; blue circles), and their governing and interacting factors (red rectangles), all of which influence the state of coastal MER (i.e., the success and progress of MER projects; hexagon). Solid arrows indicate flow of knowledge, labor and other monetary values. Broken arrows indicate interactions among the coastal MER positive-feedback loop ("stagnation loop").

another major drawback of restoration efforts in general (Palmer and Filoso, 2009; Suding, 2011), and in coastal marine ecosystems in particular (Bayraktarov et al., 2016). Even in cases where monitoring and evaluation is planned, it is often funded for a short period of time, not allowing for proper assessment of the outcome of the project over time (Statton et al., 2012, 2018). In other cases, monitoring is overlooked and considered to be an unnecessary additional cost of restoration (Bayraktarov et al., 2016). However, information gained from monitoring (ecological and social parameters) is not only necessary for determining whether the restoration goals are being met, but is essential in determining the reasons for failures, which are critically important for informing future restoration and conservation efforts. Such information is also essential for evaluating the long-term resilience of MER interventions in the face of changing climatic and societal pressures such as land use that results in continued degradation of water quality and habitat destruction (Bouma et al., 2014). The length of monitoring will depend on the stated restoration goals and performance criteria, and on the ecology of the system being restored, which influences rates of recovery. Additionally, the timescales of recovery periods may be related to the life-history characteristics of the key species targeted for restoration (e.g., ecosystem engineer species, Montero-Serra et al., 2018). Therefore, the design of monitoring programs should include relevant ecological (e.g., demographic knowledge) and social performance metrics and governance indicators (e.g., fish functional diversity, fish catch yields, coastal erosion rates, level of conflict among stakeholders), with the cost of developing and implementing a monitoring plan included as a prerequisite for all restoration projects.

New Tools, Approaches, and Conceptual Frameworks

Advancements in restoration tools and approaches that optimize success and cost-effectiveness of coastal MER may take several directions. First, indirect restoration tools can revitalize damaged ecosystems by alleviating physical stressors or improving local conditions (also termed passive restoration; Perrow and Davy, 2002). For example, improving the quality of coastal waters by restoring terrestrial ecosystems within the relevant watershed area (e.g., by re-forestation, retention ponds and constructed wetlands; Bartley et al., 2014; Abelson et al., 2016a; Roque et al., 2016; Lefcheck et al., 2018). The restoration of the hydrological conditions in mangrove rehabilitation areas provides another example, including dismantling weirs and removing dikes and dams to reduce the duration of inundation with polluted water. This in turn may enhance the dispersal and successful colonization of propagules, and promote the chances of natural regeneration (Van Loon et al., 2016). The implementation of indirect tools that have the potential to accelerate recovery and enhance resilience of restored systems should be considered in combination with direct approaches (e.g., planting and seeding) to achieve restoration goals (e.g., Lefcheck et al., 2018).

Second, technological advances can lead to efficiencies of scale and drastic reductions in cost. For instance, restoring corals through large-scale capture and release of coral larvae on decayed reefs is predicted to be much cheaper than restoring the same amount of area with garden-grown adult corals (Doropoulos et al., 2019). Likewise, restoring marsh grasses and seagrasses is sometimes more successful when they are outplanted with biodegradable structures that protect them from wave action and sediment erosion (Temmink et al., 2020, but see Orth et al., 1999; Statton et al., 2018). Another potential direction for optimizing restoration success is the development of relatively low-cost restoration tools that can be effectively scaled to different sized projects (Spurgeon, 1999; Spurgeon and Lindahl, 2000; Bayraktarov et al., 2016). An example of one such restoration approach involves restocking of key consumers (also termed "biomanipulation;" Lindegren et al., 2010). For instance, depleted herbivorous fish populations on degraded coral reefs can lead to undesirable algal-dominated phase shifts following natural disturbance events (e.g., Bellwood et al., 2006). However, in many cases the recovery of fish populations under strict fishery management and fishing bans may take many years (up to several decades; e.g., MacNeil et al., 2015). Therefore, restocking of herbivorous fish populations (accepting the prerequisite of protection in the restored site) may prevent the excessive proliferation of macroalgae, or accelerate their eradication and aid in the recovery of degraded reefs that have undergone a phase shift to an undesirable macroalgal-dominated state (Abelson et al., 2016b; Obolski et al., 2016). Under certain circumstances, eradication or culling of, for example, herbivores may be included in the restoration, mainly in temperate ecosystems (Piazzi and Ceccherelli, 2019; Guarnieri et al., 2020; Medrano et al., 2020).

Third, to improve outplanting yields, the paradigm in restoration ecology can be expanded from one frameworks that systematically identifies and reduces physical stressors, to one that also systematically harnesses positive species interactions at all levels of biological organization. This paradigm change was first proposed by Halpern et al. (2007) and Gedan and Silliman (2009) and received the first experimental support by Silliman et al. (2015), who found that planting marsh plants in clumps rather than in dispersed patterns as the paradigm called for resulted in a 100–200% increase in plant yields at no extra cost. Importantly, this study did not add extra resources to the restoration project; instead a simple design change in planting arrangement allowed for naturally occurring positive interactions to occur, as plants in clumps interacted to resist erosion and oxygen stress in the soil (Silliman et al., 2015).

Recent conceptual papers highlight that inserting positive species interactions into restoration of corals, seagrasses and mangroves, as well as into eco-engineered structures, can have beneficial outcomes and need not be limited to just intraspecific facilitation (Shaver and Silliman, 2017; Bulleri et al., 2018; Renzi et al., 2019; Smith et al., 2020; Valdez et al., 2020). Interspecific facilitation and mutualism could be equally or more important. For example, manipulation of the bacterial community is likely to enhance settlement and establishment of foundation species (e.g., corals, seaweeds and mangroves; Holguin et al., 2001; Kelly et al., 2014; Qiu et al., 2019); waterborne chemicals from various species could be mimicked at scale to induce coral settlement and fish grazing behavior (Dixson and Hay, 2012; Dixson et al., 2014); key autogenic ecosystem engineer species can enhance stress tolerance for associated organisms (i.e., "human-assisted evolution;" *sensu* Palumbi et al., 2014; van Oppen et al., 2015; see: section "Promoted adaptation"); predators can facilitate regrowth of seagrass systems and increase their tolerance to nutrient stress by promoting populations of algal grazing sea slugs (Hughes, 2014); and positive landscape-scale interactions involving fluxes of energy, materials and organisms among ecosystems can facilitate the establishment and persistence of foundation species (Gillis et al., 2014; van de Koppel et al., 2015). While incorporating positive species interactions into restoration designs holds great promise, a recent review unfortunately found that only 3% of over 600 studies investigating coastal restoration actually tested for the effects of inserting facilitation by design (Zhang et al., 2018).

Finally, management concepts should be implemented that combine restoration efforts with protection. Currently, protection and restoration are rarely integrated into management programs. Protection from anthropogenic stressors is generally not a prerequisite for MER projects, and restoration is often disregarded as a tool in MPA (marine protected area) management plans (Abelson et al., 2016a). We believe that including protection (MPAs) and stress relief in restoration projects as part of ecosystem-based management may be highly effective in conservation and the recovery of coastal marine ecosystems, and therefore, should be a normative baseline.

PROMOTED ADAPTATION

At present, coastal MER tools rarely enhance ecosystem resistance to climate-change related stressors such as ocean warming, sea-level rise and acidification (but see Shaver et al., 2018; He and Silliman, 2019). However, restoration of coastal vegetation-based ecosystems, which are major carbon sinks (i.e., saltmarshes, mangrove forests and seagrass meadows) can help mitigate climate change over large scales (Gattuso et al., 2018). When combined with other local-management actions, they can also help buffer global climatic impacts and compensate for critical ecosystem services that are impaired (Duarte et al., 2013; Possingham et al., 2015; Abelson et al., 2016a; Anthony et al., 2017; Darling and Côté, 2018; He and Silliman, 2019). Nevertheless, as climate-change mitigation (reduction of greenhouse gases emission) can take at least decades to affect the Earth's climate (Solomon et al., 2009), there is a growing recognition of the need to identify practical tools to promote adaptation to climate change, so that coastal marine ecosystems can continue to function and provide ecosystem services under a range of future environmental conditions (Webster et al., 2017; Darling and Côté, 2018; Abelson, 2020). We suggest that beyond fostering the services and ecosystem health of degraded coastal marine ecosystems, restoration tools be used to promote adaptation management to cope with future climate-change conditions. We further argue that under the reality of climate-change conditions, practices that promote adaptation should be included in coastal MER projects to improve their long-term success.

Promoted adaptation can be implemented via two potential directions: "Predict-and-Prescribe" approaches (e.g., "assisted evolution" and "designer reefs;" Mascarelli, 2014; Webster et al., 2017; Darling and Côté, 2018), which attempt to foresee future conditions; and the "Portfolio" approach, which considers the range of uncertainty of future conditions (Schindler et al., 2015; Webster et al., 2017; **Figure 2**). Although the two strategies are distinct, they may serve as complementary tools. That is, even though their applications depend on specific circumstances, both strategies can be simultaneously applied to increase the likelihood of recovery as well as helping to cope with future unpredictable conditions.

Predict-and-Prescribe

Predict and prescribe approaches are based on the notion that future environmental conditions can be predicted to some extent. Promoting adaptation of coastal marine ecosystems to predicted plausible climate change scenarios can be achieved by increasing either the intrinsic or extrinsic resistance of a system (Darling and Côté, 2018). Adaptation, in the context of "intrinsic resistance," often involves manipulating species or genotypes of ecosystem engineers (e.g., coral, mangrove, and seagrass species) to make the system better equipped to contend with changing conditions (e.g., elevated temperature and acidification), and to better resist climate change and other global stressors. Restoration employing "intrinsic resistance" approaches involves identifying or developing resistant genotypes or species, stockpiling them in sufficient quantity (e.g., via culture), and transplanting, re-introducing, or restocking them in areas most influenced by changing conditions – a process termed "assisted colonization" or "assisted migration" (Hoegh-Guldberg et al., 2008; Palumbi et al., 2014; van Oppen et al., 2015; Darling and Côté, 2018; Coleman and Goold, 2019).

Restoration employing "extrinsic resistance" approaches involves identifying and ensuring spatial refuge sites (i.e., "Resistance and Refuge;" Darling and Côté, 2018). Existing "no-take" MPAs tend to support high fish biomass, but typically provide little resistance to large-scale disturbances (Bates et al., 2019; but see Bates et al., 2014), which suggests a need for management to identify and protect regional refugia (Graham et al., 2008). Suitable refugia may include locations that are less vulnerable to climate disturbances (e.g., cool currents and deeper sites; Darling and Côté, 2018), or stressful or frequently disturbed habitats (e.g., high sedimentation, elevated temperature, acidified waters) whose constituent species are locally adapted to tolerate exposure to chronic stressors (Fabricius, 2005; Palumbi et al., 2014; Shamberger et al., 2014; Webster et al., 2017). Such habitats could serve as potential refugia due to their future resistance potential (e.g., Palumbi et al., 2014). Local refugia have the potential to drive cascading processes of large-scale recovery ("robust source sites;" Hock et al., 2017) by possessing high connectivity with the wider ecosystem network, and a low risk of exposure or sensitivity to disturbances. They serve as a source of replenishment when other sites are depleted, and promote the recovery of desirable species (Hock et al., 2017). Sites identified as potential local refugia need to be protected and the recovery of degraded sites of potentially high extrinsic resistance ("potential refugia") should be promoted by relevant restoration interventions. That is, sites can play a role as potential refugia thanks to favorable environmental conditions dictated by their location. However, if these sites are in a degraded state due to local anthropogenic stressors, they cannot serve as effective refugia, unless those local stressors have been eliminated or reduced and these systems have recovered. Also, for effective restoration and the selection of potential refugia, empirical genetic information is required to assess diversity and the potential adaptive capacity to cope with future conditions (Coleman et al., 2020). This is particularly pertinent for species that exhibit limited dispersal and are therefore susceptible to reduced gene flow (e.g., Buonomo et al., 2017).

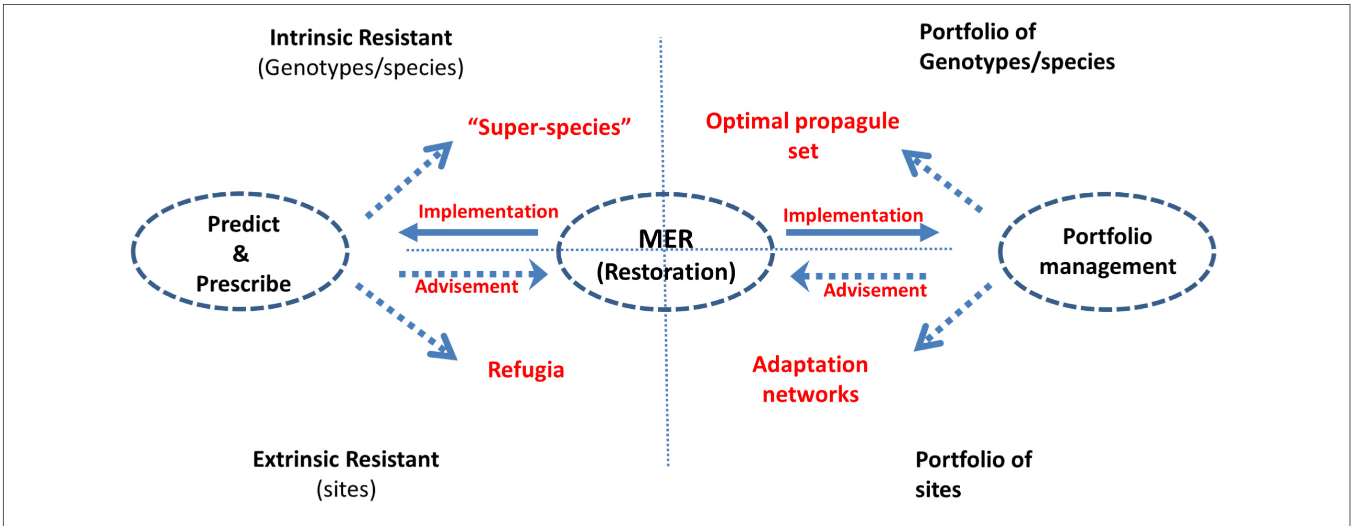

FIGURE 2 | Applying coastal Marine Ecosystem Restoration (MER) to build adaptation via two directions: "Predict and Prescribe" and "Portfolio management" approaches. Restoration tools can serve as implementation vectors of promoted adaptation. Alternatively, adaptation concepts can serve as considerations in planning sound restoration interventions.

The "Portfolio Management" Approach

The "Portfolio" approach is a risk management tool adopted from financial portfolio theory, which exploits information about spatial covariances in future ecological conditions and applies that tool to spatial targeting of conservation and restoration investments (Schindler et al., 2015; Webster et al., 2017). Recent research in fisheries and terrestrial ecosystems suggests that the portfolio theory can be applied as a potential approach to promote adaptation, while taking into account our inability to fully understand or predict the impacts of large-scale stressors (Crowe and Parker, 2008; Ando and Mallory, 2012; Schindler et al., 2015; Webster et al., 2017). The portfolio approach can be applied in coastal marine ecosystem management via two operational routes: portfolio of sites (adaptation networks of management units; Webster et al., 2017), and portfolio of genotypes and species (optimal sets of propagules; Crowe and Parker, 2008).

Portfolio of Sites

This approach is applied via adaptation networks, which are regional systems of managed areas (i.e., "management units") with attributes that promote adaptation (i.e., managed areas of high diversity, connectivity, and spatial risk mitigation; Webster et al., 2017). The management units should comprise sites of different states, depths and locations, and under diverse environmental conditions, but which are connected physically (horizontally and/or vertically) or demographically (via passive dispersal or active movement) to form networks. To maximize the ecological outcomes of each "management unit," adequate investment in protection features (planning and maintenance), notably staff capacity, fishery governance, effective enforcement, and MPA area size, has to be ensured (Edgar et al., 2014; Cinner et al., 2016; Gill et al., 2017). However, as most coastal marine ecosystems experience some extent of degradation, protection alone is insufficient and should therefore be integrated with restoration (Possingham et al., 2015; Abelson et al., 2016a). This requires investment in the exploration, examination, and development of restoration tools (e.g., Rogers et al., 2015; Abelson et al., 2016a; Anthony et al., 2017), the aim of which is to improve the recovery of each management unit.

Even if a minimum viable fraction of a given ecosystem can be protected, isolated sites may substantially weaken connectivity among the management units within the potential adaptation network (Green et al., 2015), which may in turn compromise ecosystem functioning and neutralize the effectiveness of the network (Gaines et al., 2010; Berglund et al., 2012; Green et al., 2015). Thus, the restoration of degraded coastal marine ecosystems can promote the recovery of otherwise low-quality management units and subsequently improve the connectivity (e.g., Abelson et al., 2016a; Bayraktarov et al., 2016) and the effectiveness of the "adaptation networks."

Portfolio of Genotypes and Species

Another application of portfolio theory is to select an optimal set of propagule sources ("propagule portfolio;" i.e., larvae, seeds, seedlings, and fragments) to be used to restore sites in environments of multiple plausible future climates, based on the results of a climate change impact model (e.g., Crowe and Parker, 2008). This approach combines the "intrinsic resistance" and the portfolio approaches, by applying the restoration tools required for the former with the concept of the portfolio of genotypes and species, which expands the set of propagules by a wide range of source sites under diverse environmental conditions.

To apply the "propagule portfolio," consideration should be given to selecting and culturing propagules comprising an optimal set of genotypes (i.e., a set that minimizes risk of maladaptation across a variety of future plausible climates, while meeting targets on mean adaptive suitability; Crowe and Parker, 2008), collected from populations that experience different environmental conditions, to use in the restoration of a target site via transplantation or restocking. This approach requires two data sources: (1) provenance trial data derived from multiple common culture trials of multiple propagule sources collected from populations located at various environmental conditions ("geographic points") within a region (e.g., genotypes adapted to pollution; Whitehead et al., 2017); and (2) environmental data for those geographic points (Crowe and Parker, 2008).

The portfolio approach is still largely theoretical with regard to the marine realm (but see Beyer et al., 2018). However, there is a growing array of models and proposed implementation methods that support its high potential as a management approach to cope with climate change and other unpredictable effects (e.g., Aplet and McKinley, 2017; Holsman et al., 2019; Walsworth et al., 2019). Moreover, some studies, from terrestrial and aquatic ecosystems, provide encouraging support for its applicability (e.g., Crowe and Parker, 2008; Penaluna et al., 2018; Eaton et al., 2019).

INTEGRATED SOCIAL-ECOLOGICAL RESTORATION

A major question related to ecosystem restoration in the Anthropocene is whether we can devise and implement restoration practices that service both the needs of society and promote sustained ecological functions and values (i.e., social-ecological restoration). The concept of "social-ecological restoration" extends beyond the usual scientific scope of "ecological restoration," to include reciprocal relationships between ecosystems and humans (Geist and Galatowitsch, 1999). We give this concept particular attention as restoration is a fundamentally human endeavor and social processes have been historically understudied (Wortley et al., 2013), despite the fact that they can be integral to project success (Bernhardt et al., 2007; Druschke and Hychka, 2015). Social-ecological restoration is not meant to replace ecological restoration and the consideration of natural heritage or biodiversity values, but rather to complement, as they are both nested subsets within the overall definition of restoration. Here, we highlight a few key ways that MER may benefit from the inclusion of social priorities as restoration goals and via the broadened participation of society.

The adoption of a social-ecological approach to restoration can help delineate clearer goals and aid in evaluating project achievements through performance criteria that go beyond just habitat creation (e.g., Palmer and Filoso, 2009) and contribute to the "blue economy" (World Bank United Nations Department of Economic and Social Affairs, 2017). Practically, this can be implemented by prioritizing targeted ecological and social restoration goals (e.g., conservation value, job creation, flood risk reduction; Abelson et al., 2015) that are valued by relevant stakeholders. For example, Stone et al. (2008) found that different resource user-groups were willing to contribute time and money to mangrove restoration in India, but the motivations and level of support were not consistent across groups and related to different perceived ecosystem services (i.e., fisherman supported restoration because they believed mangroves were good fish nurseries whereas rice farmers believed mangroves would control erosion). Accordingly, understanding local motivations for restoration and using that information to set and communicate clear and relevant restoration goals may enhance community buy-in and ongoing support for restoration initiatives. Furthermore, increasing societal understanding of and connection to restoration projects may facilitate more widespread support of ecological restoration as an effective management tool (Challenge 3; e.g., Edwards et al., 2013; NOAA SAB, 2014; World Bank, 2016; Strain et al., 2019).

With emerging threats from climate change and coastal urbanization, we can expect heightened conflict between MER, the propagation of new development and infrastructure, and shifting ecosystems that may impede MER efforts (e.g., mussel restocking in the wake of ocean acidification). Rising to this challenge, the field of eco-engineering has emerged with the goal of restoring ecosystems in a way that maximizes services that are desired by humans (e.g., coastal protection, waste-water treatment), rather than restoring to a previous state. These "designer ecosystems" are unlikely to deliver on all restoration goals (e.g., maximizing the restoration of biodiversity), but they are nevertheless likely to become a vital component of future coastal conservation plans for several reasons (Airoldi et al., 2020). First, eco-engineering projects that combine habitat restoration with infrastructure may be applicable in highly urbanized marine environments where large-scale restoration projects are infeasible or undesirable (Sutton-Grier et al., 2015; Morris et al., 2019). Recent research suggests that perceptions about what is desirable and acceptable in the marine environment seem to be normalizing toward degraded and artificial states (Strain et al., 2019); in these cases, eco-engineering projects can act as demonstration sites exhibiting some of the benefits of restoration within communities that are otherwise disconnected from nature. Second, eco-engineering projects may be able to provide a direct substitute for gray infrastructure that individuals and municipalities are already accustomed to paying for, and thus we may be able to redirect funding that has typically been spent to build and repair expensive gray infrastructure toward restoration (McCreless and Beck, 2016; Sutton-Grier et al., 2018; Airoldi et al., 2020). Finally, by diversifying the goals and motivations behind coastal MER projects, away from purely ecological priorities, it is likely that a larger sector of society will be engaged, restoration will be possible in a greater variety of environments, and highly urbanized areas will be able to contribute toward global restoration goals (e.g., The Bonn Challenge).

Societal involvement in the planning, implementation, and monitoring of restoration projects can play an important role in restoration success. Past experience suggests that integrated coastal MER projects that include consensus among different stakeholder groups are likely to be the most successful and cost-effective, especially in developing countries (Bayraktarov et al., 2016). Moreover, awareness of and connectedness to the marine environment can strongly predict social support for projects aimed at coastal rehabilitation (Strain et al., 2019). Therefore, the early and continuous engagement of key stakeholders (on multiple levels) should be integrated into restoration plans (**Figure 1**; Abelson et al., 2015; Zhang et al., 2018; Gann et al., 2019). Potential applications for such integration include "Marine Spatial Planning" (MSP; Tallis et al., 2012), marine protected area planning (Giakoumi et al., 2018) and other quantitative frameworks (Samhouri et al., 2012; Tallis et al., 2012). Furthermore, it has become increasingly popular to involve volunteers and citizen scientists in restoration practice and monitoring (Huddart et al., 2016), which can lower project costs (Bayraktarov et al., 2016), confer benefits to the participants including greater life satisfaction (Miles et al., 1998), and foster a stronger environmental ethos (Leigh, 2005). This in turn could help to raise support for other restoration initiatives that volunteers are not directly involved with, and potentially increase the social acceptability of projects. This mirrors the common notion that local communities are responsible for granting (or withholding) social license for a restoration effort, as these will be felt locally. Yet in practice, the dynamics of social acceptance frequently extend beyond local regions and can include stakeholders that are based far from the site in question. As Moffat et al. (2016) argue, restricting social license to local communities "neglects the organizational reality in a modern globalized world"; social license cannot therefore be restricted to "the exclusive domain of fence-line community members and operational managers." Nevertheless, volunteer efforts may not be feasible or cost-effective in certain contexts or at large scales, in which case it may be more efficient to employ local professionals.

Currently, in many conservation and restoration projects, high paying jobs and management positions go to outside professional experts and significant benefits do not reach local communities (e.g., Blue economy; World Bank United Nations Department of Economic and Social Affairs, 2017). Training and incorporating community-based professionals (e.g., Australia's Vocational Education Training programs in "Natural Area Restoration" and "Marine Habitats Conservation and Restoration") as active participants in all project stages will increase societal benefits as well as reduce potential tensions.

The value of implementing a social-ecological restoration approach in management frameworks is gaining traction. This is partly due to the ongoing degradation of coastal marine ecosystem services and the failure of traditional management practices to halt this decline (Possingham et al., 2015; Golden et al., 2016). Incorporating a social-ecological restoration component that focuses on ecosystem service outcomes, rather than exclusively relying on outcomes like biodiversity, may help compensate for decreasing ecosystem services, which now lie well below historical levels in many regions due to misuse, over-exploitation and the emerging threats of climate change (e.g., Golden et al., 2016 and citations therein). Expanding coastal MER to an integrated social-ecological system will increase the scope and complexity of restoration science and governance, and therefore demands expanded investments in development, implementation and maintenance.

PROMOTING THE PERCEPTION OF MER AS A SCIENTIFICALLY CREDIBLE MANAGEMENT APPROACH

The end of the 20th and early years of the 21st century yielded several key studies that raised the scientific background and awareness of ecological restoration, including coastal MER (e.g., Dobson et al., 1997; Edwards, 1999; Jaap, 2000; Young, 2000; Palmer et al., 2004).

In a literature search (Google Scholar) of the terms (restor* or rehabilitat*) and (marine ecosystem*, coral, mangrove, oyster, saltmarsh, kelp, or seagrass) in the title, we found relatively few restoration papers published prior to 2000 (**Figure 3**). The trend changed significantly circa 2000 with an order of magnitude increase in the number of restoration studies in six major coastal ecosystems (**Figure 3**). However, the total number of coastal MER studies remains negligible relative to restoration studies in terrestrial (e.g., forests) and freshwater (e.g., rivers and lakes) ecosystems. We recognize that our figures may be underestimates of the actual numbers of restoration studies. However, figures obtained by our search should provide a reasonable indication of the general trend of restoration ecology as a field of science, and the relative fraction of each sub-field for different ecosystems.

If the number of peer-reviewed publications serves as a proxy of investment in science (Ebadi and Schiffauerova, 2016), then it can be argued that, despite the growing research in coastal MER, investment is still relatively low, lagging behind restoration research of non-marine environments. A possible reason for this lagging behind of restoration of marine ecosystems is that their restoration projects are still undervalued (Gordon et al., 2020), mainly due to criticism about their limited spatial scale and high costs, which are too expensive to combat the extent of anthropogenic threats driving habitat loss (Gordon et al., 2020). The consequence is that major gaps remain in the applicability (e.g., cost-effectiveness) and relevance (i.e., goals detached from the definition of ecological restoration) of many coastal MER projects and practices, which may explain the current poor perception of coastal MER among many marine scientists (e.g., Adger et al., 2005; Mumby and Steneck, 2008: Bayraktarov et al., 2020). Although large-scale successful and relatively low-cost projects exist (notably large-scale mangrove forest, oyster reef and salt-marsh restoration projects; e.g., Beck et al., 2011; Bayraktarov et al., 2016; Friess et al., 2016; Duarte et al., 2020) many restoration projects are costly, conducted at small scales, and with narrow goals that do not benefit a diverse stakeholder group (including the majority of coral reef restoration projects; Bayraktarov et al., 2016). At present, a widespread goal of coastal MER projects is to achieve "item-based success" (i.e., survival of planted transplants, seedlings, or spat; *sensu* Bayraktarov et al., 2016), which in part reflects a common expectation for quick, measurable results, and a general assumption that associated ecosystem services will follow. The consequence is that basic science and "non-simplistic" applied research projects are missing,

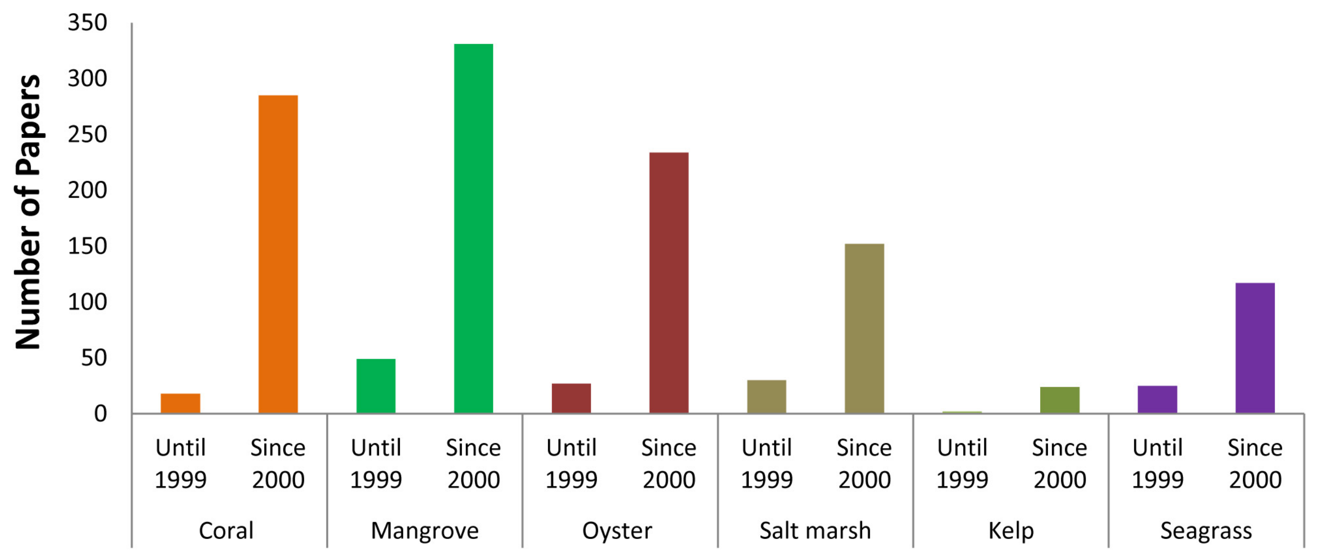

FIGURE 3 | The number of coastal Marine Ecosystem Restoration (MER) papers: the general term "marine ecosystems" and each of the six major coastal ecosystems: coral reefs, mangrove forests, oyster reefs, salt marshes, kelp forests and seagrass meadows, pre- and post-2000.

but are needed to promote tools, practices and scaling up of coastal MER (Bayraktarov et al., 2020). Moreover, MER is seen as a "risky choice" for resource managers and science policy-makers. Basic science is an important source of new ideas that figure prominently into developing solutions for many of society's needs (Remedios, 2000). Therefore, support for basic long-term research is crucial for the development and implementation of coastal MER. However, at present the development and implementation of most coastal MER sectors suffer from the effects of a "performance-perception-funding" cycle ("stagnation loop;" **Figure 1**), in which poorly performing restoration projects lead to poor images of coastal MER, and therefore hinder adequate investment in development of coastal MER science and practice despite general recognition of ecosystem decline. Breaking out of this "stagnation loop" requires major achievements by restoration projects in the relevant ecosystems.

Potential advantages of coastal MER compared with conservation-based management approaches reliant on area protection are best highlighted by successful restoration projects involving mangrove, oyster reefs (Beck et al., 2011; Bayraktarov et al., 2016; Friess et al., 2016) and seagrass meadows (Orth et al., 2012). However, although the list of successful large-scale MER projects continues to increase over time, modeling studies that compare the expected ecological and socio-economic benefits of different management approaches through time should be encouraged to demonstrate the economic benefits of restoration. Results from such studies done to date suggest that restoration-based conservation programs in coral reefs and large-scale efforts in seagrass-based restoration, despite the costly investment, may prove to be worthwhile due to the faster recovery and enhanced ecosystem services (Obolski et al., 2016; van Katwijk et al., 2016).

Targeted restoration projects with realistic ecological and socio-economic goals should help identify important knowledge gaps in coastal MER (i.e., SER, 2004 definition). Such goals include ecosystem-level parameters (e.g., fish species diversity and biomass) and upgraded ecosystem services, rather than "item-based success" indicators (e.g., survival of planted ecosystem engineer species). Likewise, coastal MER projects should be scaled up, beyond the usual but limited experimental scales, provided that the stressors that led to the degradation have been eliminated or minimized, or new tools, which help overcome the still existing stressors, are applied. The current proliferation of small-scale, item-based, trial projects, with no stakeholder involvement (Bayraktarov et al., 2016), is unlikely to fill the gaps and needs of realistic coastal MER. Hence, a shift toward realistic coastal MER interventions (i.e., feasible interventions of ecological and socio-economic benefits) is critically needed for coastal MER to gain wider acceptance. We believe that combining coastal MER and coastal ecosystem conservation into a single social-ecological framework (Possingham et al., 2015) has great potential to provide significant socially relevant gains in conserving and restoring highly valued coastal ecosystems. Such integration may further help to increase the traction of coastal MER and improve its perception and acceptance as an effective management strategy.

RECOMMENDATIONS

In view of the ongoing degradation of coastal marine ecosystems, restoration is an inevitable component of conservation management. Successful coastal MER offers great promise for accelerating the recovery of collapsed populations (including globally threatened species), destroyed habitats, and impaired ecosystem services, which may otherwise take much longer to recover (years to decades), if at all. To this end, effective implementation of coastal MER will benefit from incorporation of socio-economic elements, a wider portfolio of methodological tools, more focused post-restoration assessment, climate-change considerations, and wider stakeholder acceptance and engagement. We note that policy and legislation to enable this approach is critical, and notable efforts are being made, including, for example, the United Nations Decade of Ocean Science for Sustainable Development (2021–2030), the United Nations Decade of Ecosystem Restoration (2021–2030), and the European Green Deal, which makes restoration one of the key objectives. We encourage the development of specific recommendations in this field to further support restoration as a fundamental strategy in the race to reverse the decline of coastal marine ecosystems.

We Conclude

– Indirect tools that remove or modulate stressors, accelerate recovery and enhance the resilience of restored systems should be used in combination with direct approaches (e.g., planting and seeding) to achieve restoration goals. Basic scientific research will contribute to identification of such indirect tools.
– The growing need for large-scale restoration interventions, notably projects that combine remediation of degraded ecosystems due to past impacts and adaptation to cope with future threats, requires refinement of existing methods scaled to address the extent of degraded habitat, and support for multidisciplinary research that explores and identifies new tools and approaches. Such research requires adequate funding and a substantial breadth of skills; however, inadequacies in both have hampered the advancement of coastal MER. Therefore, concept promotion and education by ecological restoration proponents is essential for fundamental breakthroughs and coastal MER progress.
– Improved identification and understanding of social processes, drivers and priorities is needed to ensure broad public support and the long-term success of restoration efforts. Ideally, restoration and conservation approaches should be integrated with marine and coastal management. Under this umbrella, engaging local communities in the planning and monitoring of MER projects and designing projects with them to deliver specific socio-economic benefits will greatly enhance the long-term success of both conservation and restoration activities.
– Beyond fostering the ecosystem health and services of degraded coastal marine ecosystems, restoration tools can be used to promote adaptation to cope with

climate-change. Promoted adaptation can be implemented via two potential directions: the "Predict-and-Prescribe" approaches (e.g., "assisted evolution" and "designer reefs"), which attempt to foresee future conditions; and the "Portfolio" approach, which considers the range in uncertainty of future conditions. We argue that MER-based practices that can promote adaptation should be included in coastal zone management plans to improve their long-term success.

AUTHOR CONTRIBUTIONS

All authors have conceived the study. AA led the writing of the manuscript and project coordination. All co-authors contributed to the draft and gave final approval for publication.

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