Hyperspectral imaging applied to microplastic monitoring in marine sediments from a highly contaminated coastal site (Taranto, southern Italy)

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Introduction

In this study, an innovative methodological approach based on hyperspectral imaging (HSI), combined with machine learning approaches, is proposed to identify microplastics (MPs) in marine sediments collected in the Mar Piccolo basin (Gulf of Taranto, southern Italy) during a sampling campaign carried out in December 2022. The area of Taranto is characterized by the presence of high-density anthropogenic activities, including industrial districts, shipyards and arsenals, and intensive mussel aquaculture plants, which have led to relevant environmental modifications. The study area has been selected since, due to the high level of environmental risk, it is included in the Italian list of contaminated Sites of National Interest (SIN in Italian). Furthermore, this research represents the first attempt to monitor the MPs distribution in the investigated sites. The study area location is shown in Fig. 1. It is a semi-enclosed sheltered sea with very low water circulation and characterized by the presence of several submarine springs that recharge the basins with freshwater.

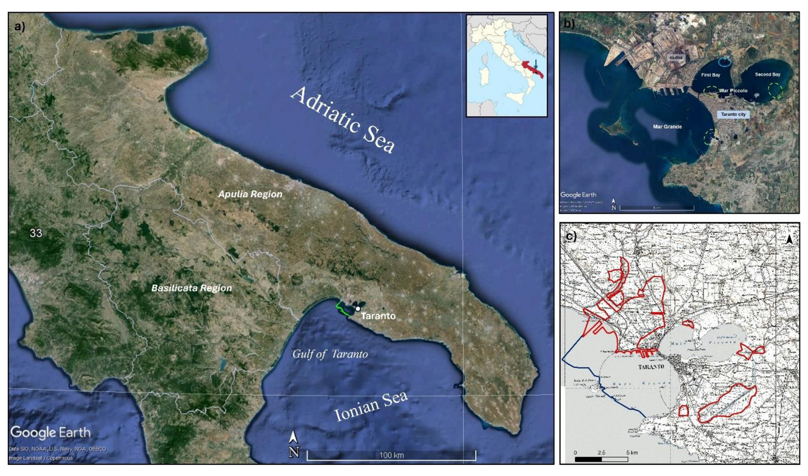


Fig. 1. a) Geographical location of the Taranto area (Apulia Region). b) The green dashed circles identify the shipyards and dockyards of the Italian Navy in the Mar Grande and Mar Piccolo (Bay I) and of the Italian Air Force in the Mar Piccolo (Bay II) whilst the blue circle identifies the ex-Tosi shipyard area. c) SIN_07 - Taranto perimeter. Blue lines identify the limit of the marine area included in the SIN perimeter whilst red lines identify the limit of the in-land areas. Background images in a) and b) were exported from Google Earth and modified by the authors whilst SIN perimeter in c) was provided by the Italian Ministry of the Environment.

Materials and methods

Marine sediments sampling and grain-size analysis

Marine sediments were collected using a grab sampler installed on board a ship. Sampling sites (MP_01 – MP_08) were defined in both sub-basins of the Mar Piccolo (the First Bay and the Second Bay – Fig. 2a) accounting for the distribution of the mussel farm facilities (Fig. 2b, c), that obstructed the passage of the ship, and the mouth of the main rivers that flow in the basins (Galeso and Cervaro). Grain-size analyses were carried out at the laboratory of the Department of Earth and Geoenvironmental Sciences of University of Bari Aldo Moro (Italy) by following international standard procedures. For the sieving, a set of ASTM sieves was used. Before sieving, the sediment samples were firstly weighed and then dried in the oven at a temperature between 30 and 50° C for at least three consecutive days. The sand sediments from 2.0 mm to 0.063 mm were sieved with the vibrating screen for a duration of 20 min. Grain-size analyses of the fraction <63 µm were conducted by the use of Coulter counter that works on dispersing samples. Each retained fraction was weighed, and the results were processed with a specific application for Microsoft Excel (Gradistat© v8) to evaluate the main textural parameters.

Fig. 2. a) Sampling sites in the Mar Piccolo Basin. b) Mussel farm facilities in the First Bay. c) Mussel farms in the Second Bay.

Hyperspectral imaging and datasets preparation

Hyperspectral image acquisitions in the short-wave infrared (SWIR: 1000-2500 nm) range were performed at the Raw Materials Laboratory (RawMaLab) of the Department of Chemical Engineering, Materials & Environment (DICMA) of Sapienza University of Rome (Italy), using the SISUChema XL™ Chemical Imaging Workstation (Specim Ltd., Oulu, Finland) (Fig. 3). The selected configuration uses a 31-mm lens with 50 mm field of view (FOV), a spatial resolution of 150 µm/pixel and a scanning speed of 17.35 mm/s.

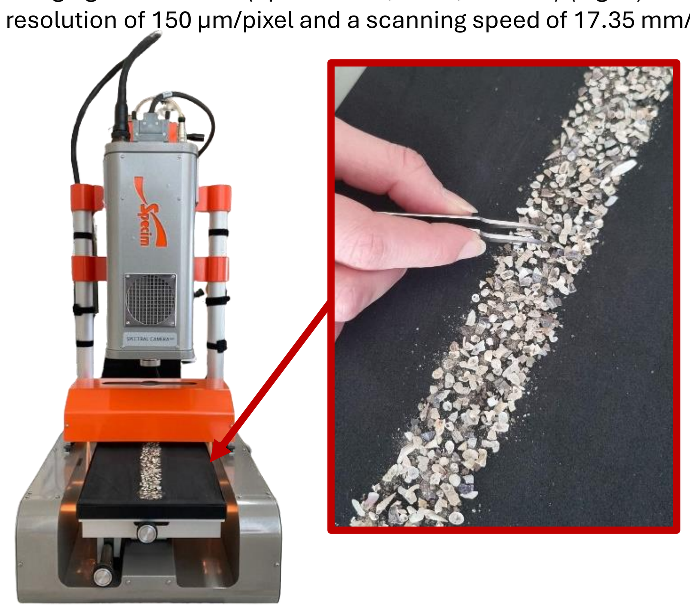


Fig. 3. HSI acquisition platform used to acquire the examined marine sediment samples.

Plastic waste flakes of 6 different polymers (Fig. 4a) and selected portions of marine sediment samples (Fig. 4b, 4c) collected from Mar Piccolo, subdivided in different size classes, were used as calibration (CAL) and validation (VAL) datasets to build and apply the classification model. The selected polymers are: polyamide (PA), polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS) and polyvinyl chloride (PVC). In addition, the considered 5 grain-size classes are: -4/+2.8mm, -2.8/+2mm, -2/+1.4mm, -1.4/+1mm, and -1mm/+710µm. Finally, the classification model was applied to the hyperspectral images of the real marine sediments from Mar Piccolo subdivided in the same 5 grain-size classes and placed in monolayers of 5 cm strips (Fig. 5).

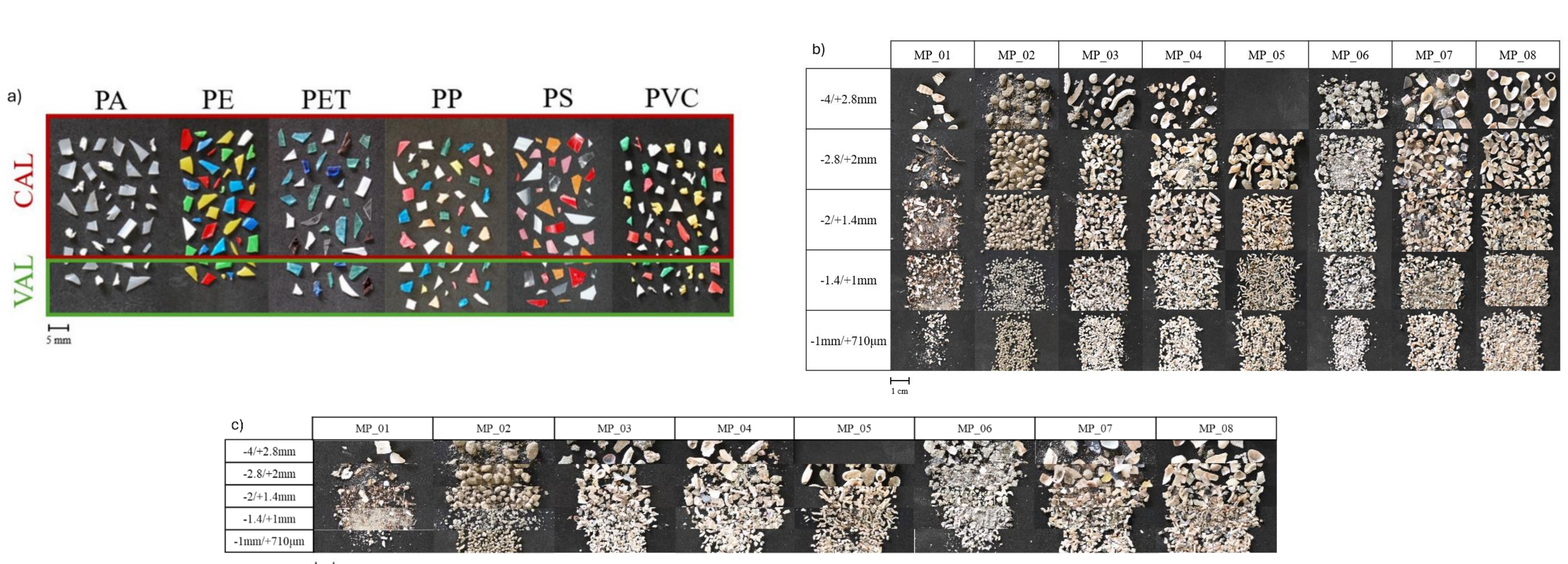


Fig. 4. Source images of the samples used to build the calibration and validation datasets of the classification model. a) Polymer waste flakes selected for the calibration (red) and validation datasets (green). b) Marine sediments without MPs selected for the calibration dataset. c) Marine sediments without MPs selected for the validation dataset.

(MP_05)

Fig. 5. Source images of representative samples of real marine sediments from Mar Piccolo basin, used to evaluate the presence of MPs by applying the classification model.

Hyperspectral data processing

Data processing was carried out by PLS_toolbox (Eigenvector Research, Inc.) running in MATLAB® (The Mathworks, Inc.). Different pre-processing algorithms were applied to data, highlighting the spectral differences of the 7 studied classes, (i.e., PA, PE, PET, PP, PS, PVC and Sediment) eliminating undesirable phenomena and reducing noise, such as light scattering. Principal Component Analysis (PCA) was applied to spectral data for exploratory purposes. Starting from the information obtained by PCA and given the complexity of the spectral data, a hierarchical classification approach (Hi-PLS-DA) was adopted based on 6 different PLS-DA (Rule 1, 2, 3, 4, 5 and 6), constituting a single classification model. Contiguous Block method was applied as cross-validation method for each rule.

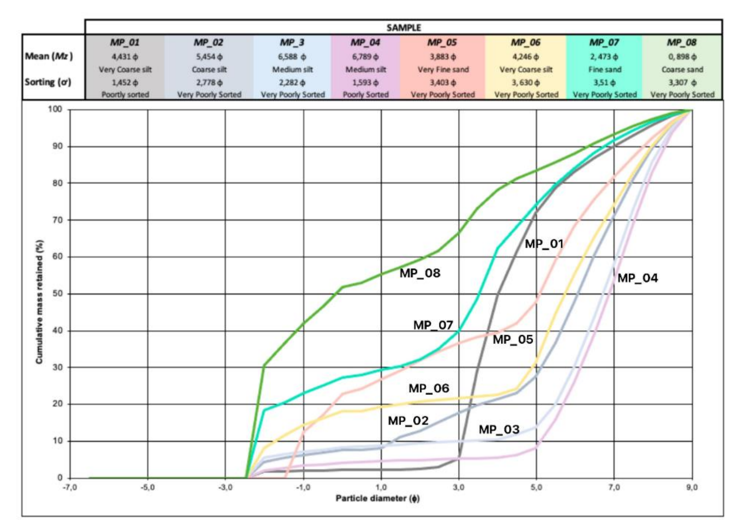
The particles classified by Hi-PLS-DA as MPs were subsequently validated using Fourier-Transform Infrared spectroscopy with Attenuated Total Reflection (FTIR-ATR).

The performances of the Hi-PLS-DA model applied to the validation dataset were evaluated in terms of statistical parameters (pixel-based) in prediction, i.e., recall (or sensitivity in binary classification), and specificity (Eq. 1 and Eq. 2).

$$Recall = \frac{TruePositive}{TruePositive + FalseNegative} \quad (1) \qquad Specificity = \frac{TrueNegative}{TrueNegative + FalsePositive} \quad (2)$$

Experimental results

Sediments characterization



The sediments of the Mar Piccolo basin show a large variety of granulometric classes, being mainly composed of silt (ranging from very coarse to medium silt) and sand (from very fine to coarse sand). In fact, sample sediments are characterized by a range of mean size limited between 0,898 phi (coarse sand) and 6,588 phi (medium silt) (Fig.6).

Fig. 6. Mar Piccolo sediments characterization. The values of the main granulometric parameters (mean size and sorting) are indicated in the upper part of the figure and showed using the cumulative curves. Each color in the table and graph correspond to a specific sediment sample.

Reflectance spectra and PCA

The average raw reflectance spectra of the 7 studied classes for the calibration dataset in the SWIR range are reported in Fig. 7. Sediment and polymer classes show different spectral absorption bands in the SWIR range. The results of PCA in terms of score plots, for each rule of the Hi-PLS-DA, are shown in Fig. 8.

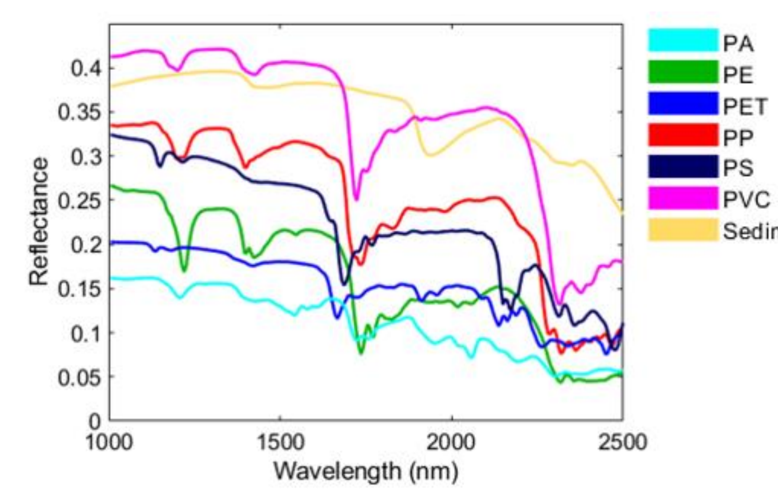


Fig. 7. Average raw reflectance spectra of the 7 selected classes of materials (i.e.: 6 polymers and sediment matrix) in the SWIR range.

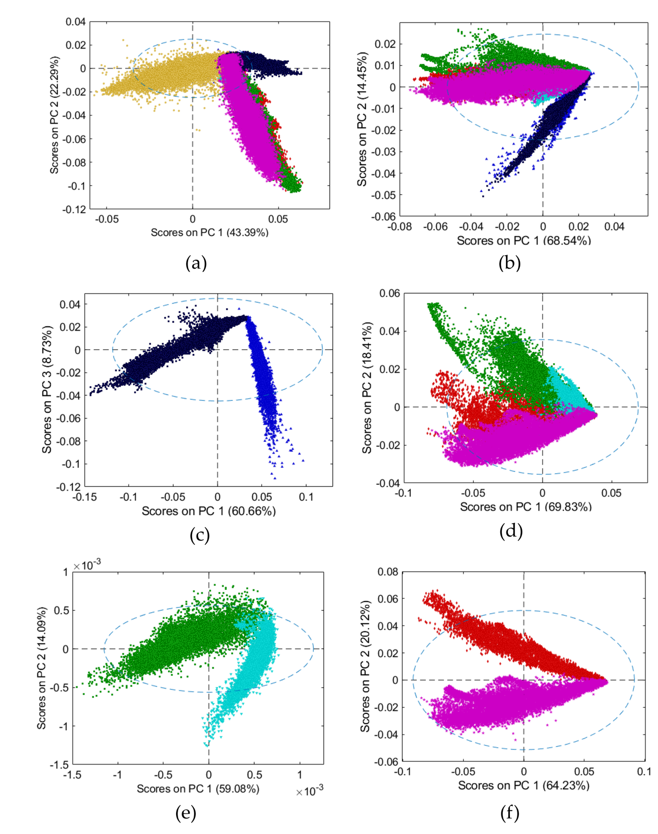


Fig. 8. PCA score plots related to the 6 different rules used to build the Hi-PLS-DA model.

Hierarchical classification model results

The structure of the hierarchical model is shown in Fig. 9, displaying the relationships between the different rules. The classification results of the validation dataset in terms of false color predicted images are shown in Fig. 10 while the classification results of validation dataset in terms of statistical parameters in prediction phase are shown in Table 1. A satisfactory classification was achieved for each class by Hi-PLS-DA model, considering the complex spectral scenario, except for some pixels attributed to the PA class at the edges of some sediment particles.

The classification results in terms of predicted images of the real marine sediments and the corresponding source images are shown in Fig. 11. The classification model successfully identified a total of 11 real MPs, with PP being the most abundant polymer (36.4%), followed by PS (27.3%), PE (18.2%), PVC (18.2%). The abundances of polymers (number of particles and number %) constituting the MPs identified in the real marine sediment samples by Hi-PLS-DA model are shown in Fig. 12.

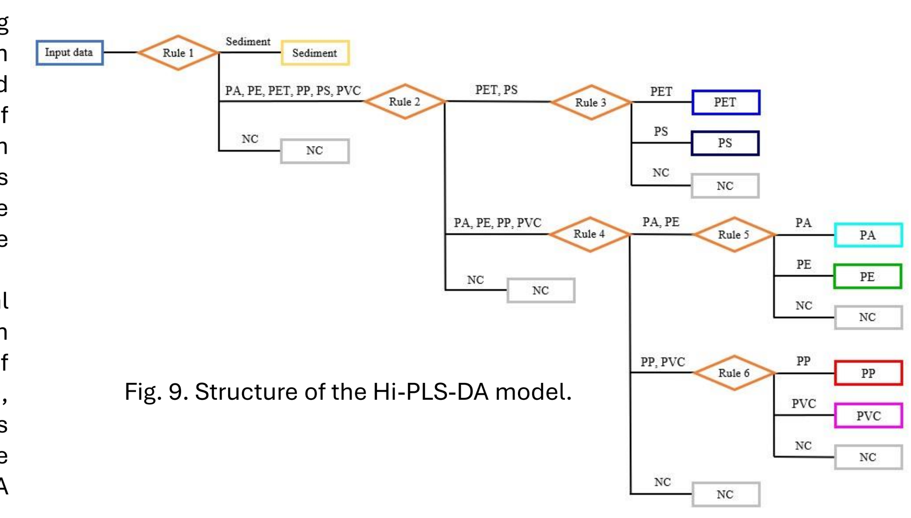


Fig. 9. Structure of the Hi-PLS-DA model.

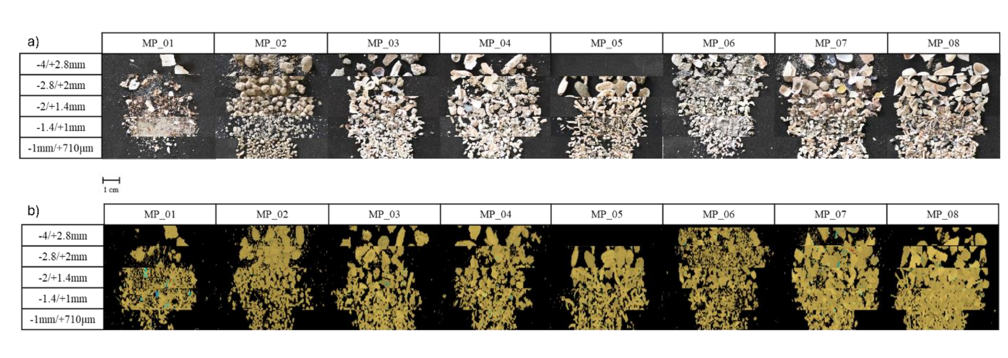


Fig. 10. a) Source images of validation dataset. b) Hyperspectral images of the validation dataset showing the classes predicted by Hi-PLS-DA.

The analyses carried out by FTIR-ATR, confirmed the correct recognition obtained by the developed HSI-based strategy.

The results show a wide variability in MPs distribution with respect to grain-size classes. Therefore, no correlations are highlighted between the presence of MPs and grain-size classes.

Concerning the MPs distribution with respect to sampling points in the Mar Piccolo basin, the results show a higher number of MPs particles in the MP_07 sample (4 particles), followed by MP_01 (3 particles). In the other sampling sites, i.e. MP_02, MP_03, MP_05 and MP_08, only 1 MP particle is recognized for each point. On the contrary, in the MP_04 and MP_06 sampling points no MPs particles are found.

Table 1. Recall and specificity values in prediction of validation dataset obtained by Hi-PLS-DA model.

Class	Recall (Pred)	Specificity (Pred)
PA	0.99	0.99
PE	0.99	0.99
PET	1.00	0.99
PP	1.00	0.99
PS	1.00	0.99
PVC	0.99	0.99
Sediment	0.99	0.99

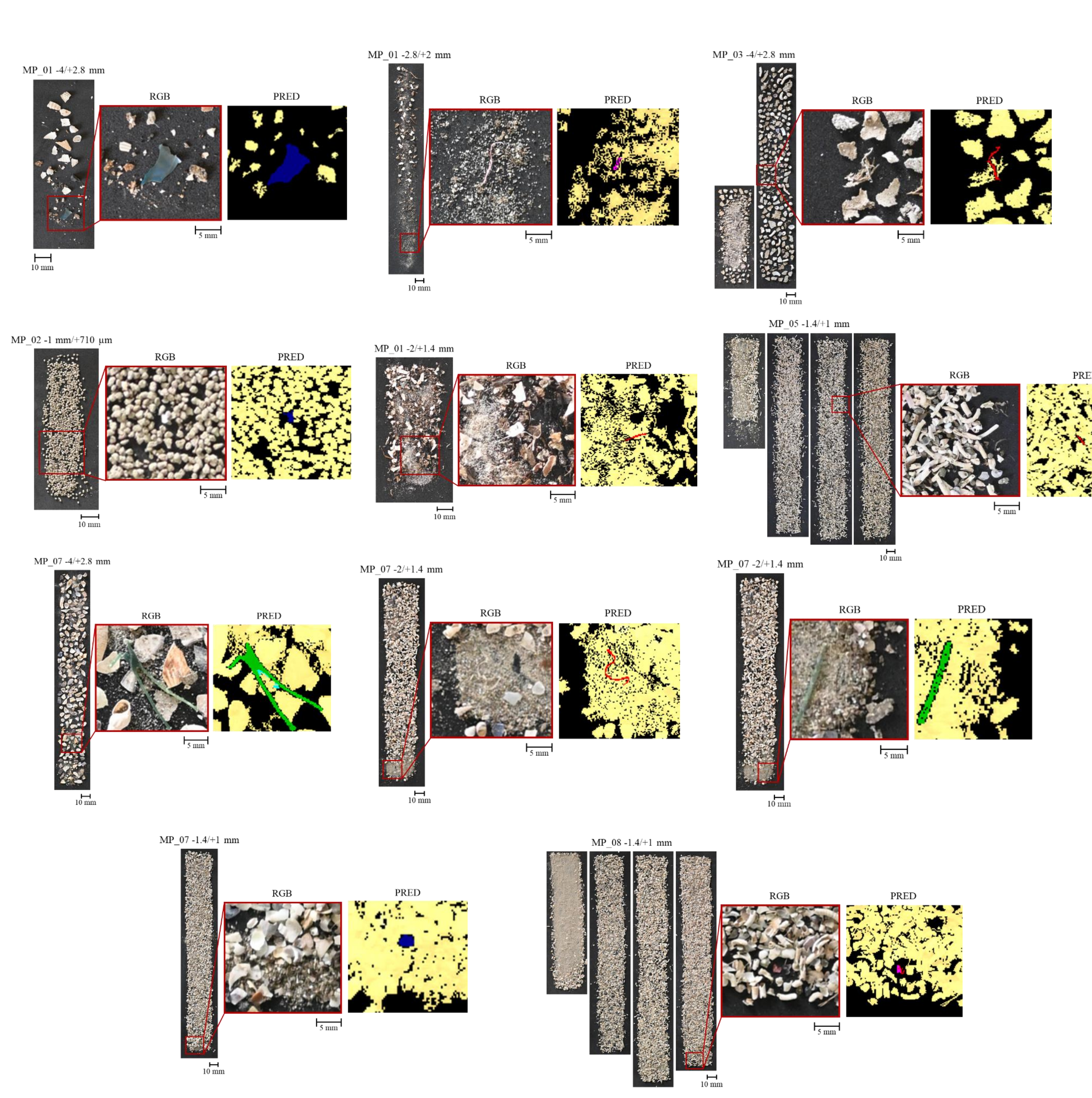


Fig. 11. Real marine sediment samples investigated by the Hi-PLS-DA model. Details of both source images and predicted hyperspectral images in which the presence of MPs is highlighted.

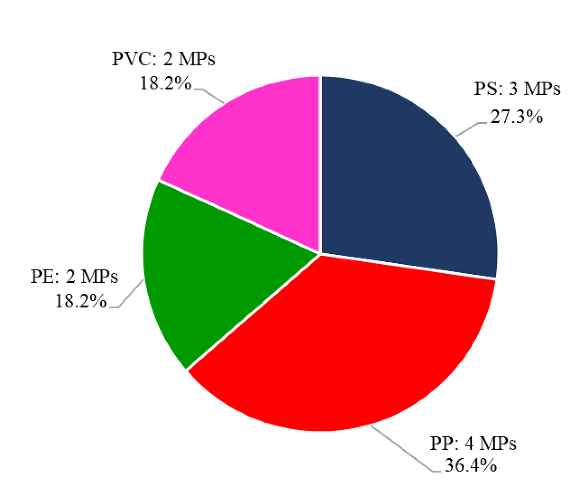


Fig. 12. Abundance (number of particles and number %) of polymers identified by the Hi-PLS-DA model in the real marine sediment samples.

Conclusions

In this work an efficient approach that reduces analysis time and facilitates environmental monitoring efforts was proposed. The results demonstrated the efficacy of the developed strategy, combining HSI with machine learning, for the rapid and automated detection of MPs >710 µm in marine sediments. Future research will investigate marine sediments of smaller sizes (diameter <710 µm) requiring a different HSI architecture set up, for acquisitions with a smaller spatial resolution, and the building of a new classification model. The outcomes of this research are aimed at supporting the geo-environmental characterization of highly contaminated coastal sites.

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