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Microplastic pollution in the North-east Atlantic Ocean surface water: How the sampling approach influences the extent of the issue

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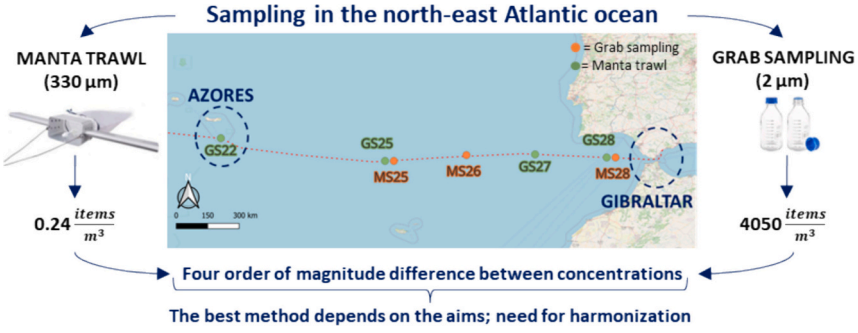
HIGHLIGHTS

- Manta net and grab sampling were compared for MP pollution in the NE Atlantic Ocean.
- Onshore processing using tape lifting avoids airborne contamination in MP analysis.
- Grab sampling yielded four orders of magnitude more MPs than manta trawl.
- No technique is inherently superior. Appropriateness depends on study goals.
- Combining methods often provides the most reliable MP quantification in water.

GRAPHICAL ABSTRACT



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ABSTRACT

A lack of standardization in monitoring protocols has hindered the accurate evaluation of microplastic (MP) pollution in the open sea and its potential impacts. As sampling techniques significantly influence the amounts of MPs contained in the sample, the aim of this study was to compare two sampling methods: Manta trawl (size selective approach) and grab sampling (volume selective approach). Both approaches were applied in the open sea surface waters of the North-east Atlantic Ocean. Onshore sample processing was carried out using the innovative tape lifting technique, which affords a series of advantages, including prevention of airborne contamination during analysis, without compromising integrity of the results. The results obtained indicated an MP concentration over four orders of magnitude higher using grab sampling compared to the Manta net approach (mean values equal to 0.24 and 4050 items/m^3, respectively). Consequently, the sole quantification of MPs using results obtained with the Manta trawl resulted in a marked underestimation of abundance. Nevertheless, the grab sampling technique is intricately linked to a risk of collecting non-representative water volumes, consequently leading to an overestimation of MPs abundance and a significant inter-sample variability. Moreover, the latter method is unsuitable for use in sampling larger MPs or in areas with low concentrations of MP pollution. The optimal sampling method therefore is dependent on the specific objectives of the study, often resulting in a combination of size and volume selective methods. The results of this study have the potential to contribute to the standardization of monitoring protocols for microplastics, both during the sampling phase and sample processing.

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1. Introduction

The excessive use of plastic and persistence in the environment has led to the pollution of seas and oceans worldwide: plastic waste constitutes 80 % of marine debris (GESAMP, 2019; Poli et al., 2023; UNEP, 2016), with an estimated 4.8 to 12.7 million tons of plastic ending up in the ocean annually (Barrows et al., 2017; Jambeck et al., 2015; UNEP, 2016). This issue is included in the United Nations' Sustainable Development Goals (SDGs), specifically SDG 14, which aims to "Conserve and sustainably use the oceans, seas and marine resources for sustainable development". The target is to prevent and significantly reduce all types of marine pollution by 2025 (Target 14.1), including marine plastic debris (Index 14.1.1) (United Nations, 2023).

Plastic litter contamination of aquatic ecosystems is recognized as one of the major causes of a wide range of harmful effects, including death of aquatic animals due to waste entrapment or ingestion, and bioaccumulation of MPs – small polymer fragments measuring less than five millimeters – in the food chain (Arcangeli et al., 2018; Cincinelli et al., 2019; GESAMP, 2019; Lavagnolo et al., 2023; Pasquier et al., 2022; Schmid et al., 2021a, 2021b; Shim et al., 2022; Zeri et al., 2018). The latter is particularly concerning in view of its potential to compromise aquatic biodiversity and adversely affect human health (Arcangeli et al., 2018; Jambeck et al., 2015; Poli et al., 2023; Schmid et al., 2021a; Simon-Sánchez et al., 2022). The identification of a reliable, established and standardized method for use in quantifying MP particles in the environment will play a key role in assessing the consequences of plastic debris in aquatic ecosystems (GESAMP, 2019).

The monitoring of MPs in an oceanic environment is particularly challenging, due to its vast surface and volume of water, dynamic nature, and intricate interactions with a multitude of physical, chemical, and biological phenomena. For these reasons, no standardized sampling methods or internationally recognized protocols are currently available; accordingly, a plethora of diverse methodologies have been formulated, encompassing both surface water and abyssal water (Du et al., 2022; Montoto-Martínez et al., 2022; Pasquier et al., 2022). Barrows et al. (2017) conducted an extensive examination of a series of established techniques used in the sampling of MPs in aquatic environments. This review distinguished between methods involving the collection of specific volumes of water, which were subsequently filtered (e.g., bottle grab samples or Niskin bottle samples), and the more widespread employment of net-based sampling surveys.

The specific used in MP sampling in an aquatic environment significantly affects the concentration of MPs present in the sample (Barrows et al., 2017; Green et al., 2018; Montoto-Martínez et al., 2022; Prata et al., 2019; Song et al., 2014; Vermaire et al., 2017). Using a size selective approach, i.e. net-based sampling methodologies such as Manta trawl, an extended volume of surface water can be sampled, although the effectiveness is limited by the net mesh size (Barrows et al., 2017; Du et al., 2022; Green et al., 2018; Gwinnett et al., 2021a; Karlsson et al., 2020; Prata et al., 2019; Schmid et al., 2021b; Song et al., 2014; Tamminga et al., 2019). This methodology therefore is unrepresentative of the quantity of MPs as it results in an underestimation of plastic concentration (Barrows et al., 2017; Du et al., 2022; Pasquier et al., 2022; Song et al., 2014), and is therefore not reliable for use in investigating the actual extent of MP pollution in aquatic environments (Du et al., 2022; Gwinnett et al., 2021a). In contrast, a volume selective approach, such as grab sampling, can be relied on to fully represent the quantity of MPs in water, with the risk however of introducing a large variability among samples when the water sample volume is not large enough (Barrows et al., 2017; Du et al., 2022; Prata et al., 2019; Schmid et al., 2021b). It may be possible to overcome this issue by increasing the volume of water or augmenting the quantity of grab samples (Barrows et al., 2017; Lusher et al., 2014; Prata et al., 2019; Schmid et al., 2021b; Song et al., 2014; Tamminga et al., 2019). Consequently, when an accurate and precise understanding of the abundance of MPs is required, a synergic utilization of multiple methodologies is recommended (Barrows et al., 2017; Song et al., 2014).

However, a lack of harmonization in MP sampling, processing and analysis methodologies has hindered the comparison of data deriving from different studies (Barrows et al., 2017; Du et al., 2022; GESAMP, 2019; Karlsson et al., 2020; Pasquier et al., 2022; Prata et al., 2019; Rivers et al., 2019; Schmid et al., 2021b; Shim et al., 2022; Simon-Sánchez et al., 2022). For example, different techniques yield different quantities of sampled material, different units may express different results, different net mesh sizes or filter pore sizes may be used, different particle size ranges may be investigated and different protocols may be used to reduce sample contamination (Barrows et al., 2017; Karlsson et al., 2020; Pasquier et al., 2022; Prata et al., 2019; Rivers et al., 2019). There is therefore an urgent need for standardization to enhance investigation of the true extent of plastic debris-related issues and assess the deriving potential impacts on the aquatic ecosystem and its biota (Barrows et al., 2017; Du et al., 2022; GESAMP, 2019; Green et al., 2018; Song et al., 2014).

The primary aim of this study was to elucidate the strengths and weaknesses of microplastic sampling methods, in order to understand their preferability based on the context and research objectives, with the ultimate goal of contributing to the standardization of monitoring protocols for microplastics. Another objective of this study was to elucidate the correlation between the abundance of collected MPs and the chosen sampling method, specifically exploring the relationship between MP concentration in the samples and lower dimensional cut-off selected for the analysis of polymeric particles. Two distinct sampling approaches were compared: a Manta trawl method, wherein the lower size threshold (300 μm) aligned with the mesh size of the net, and a grab sampling technique, whereby the dimensional limit (2 μm) approximately corresponded to the mesh size of the filter employed during the vacuum filtration processing step. The aim was to provide an approximation of the potential underestimation of MP concentrations yielded by means of size-selective methodologies. Sampling efforts were concentrated on surface waters of the North-east Atlantic Ocean, collected during a research expedition spanning from the Azores archipelago (Portugal) to Gibraltar (Spain). Samples were specifically obtained in open sea regions rather than coastal areas, which are characterized by limited available data in the existing literature.

2. Materials and methods

The project "A Sail for the Blue: Research for Oceans and Microplastics", promoted by the University of Padova, aimed to sample MPs in the Atlantic Ocean during the summer of 2022 in the context of a two-month sailboat cruise from Cape Canaveral (USA) to Gibraltar (Spain). In this article, only the results from the Azores archipelago to Gibraltar are presented.

2.1. Sampling area and strategy

Field sampling was conducted between 19 and 26 June 2022 onboard the *Jancris* sailing vessel, transiting from the Azores (Marina de Vila do Porto, Santa Maria island, 36°56′44.7″N, 25°08′45.3″W) to near Gibraltar (Puerto Deportivo Marina de Estepona, Spain, 36°24′54.6″N 5°09′28.2″W). Surface water from the North-east Atlantic Ocean was sampled by means of two different types of MPs sampling approaches, based on the amount of water to be sampled and the size fraction to be targeted:

- A volume reduction approach consisting in the towing of a net (Manta trawl) along the water surface. This method is size selective, meaning that only MPs larger than the net mesh size are retained.
- A grab sampling approach (aka bulk sampling), in which a known amount of surface water is collected with the use of glass containers. This technique is not size selective – all MPs present in the water grab sample are collected – but rather volume selective, with the

container volume limiting the volume of sampled water. Five-liter water samples were collected, conservative compared to the range of values suggested by Prata et al. (2020), to achieve representativeness of the volume for analysis of small MPs.

A total of 3 samples were collected via Manta trawl (MS25, MS26, MS28) and 4 via grab sampling (GS22, GS25, GS27, GS28), as shown in the graphical abstract and Fig. 1 C).

The Manta trawl used in this study (Hydro-Bios Apparatebau GmbH, catalogue number: 438217) had an aluminum frame with a rectangular mouth opening (15 cm height x 30 cm wide) and a 200 cm long polyamide net with a mesh size of 300 μm. At the end of the net, a detachable cod-end (diameter 11 cm) with a polyamide mesh size of 300 μm was set. Once positioned at the initial location of the transect, the Manta net was launched from the stern of the boat, towed horizontally at the water surface and deployed at a distance from the vessel of about 20 m, outside the vessel's wake. GPS location was recorded to calculate the towing distance (Table 1). The chosen towing duration was 30 min, which is slightly conservative compared to the GESAMP (2019) guidelines and findings presented in the review of Pasquier et al. (2022). When a trawl was completed, the net was rinsed from the outside with seawater, so that the material trapped in the length migrated to the cod-end. Upon completion of the net cleaning procedure, the distal end of the tube was detached, inverted upside down into a metallic bucket and meticulously rinsed externally to ensure complete transfer of its entire contents into the bucket, and later into glass jars for subsequent filtration and analysis in the onshore-laboratory. The determination of the volume of water sampled by the Manta trawl was accomplished according to the

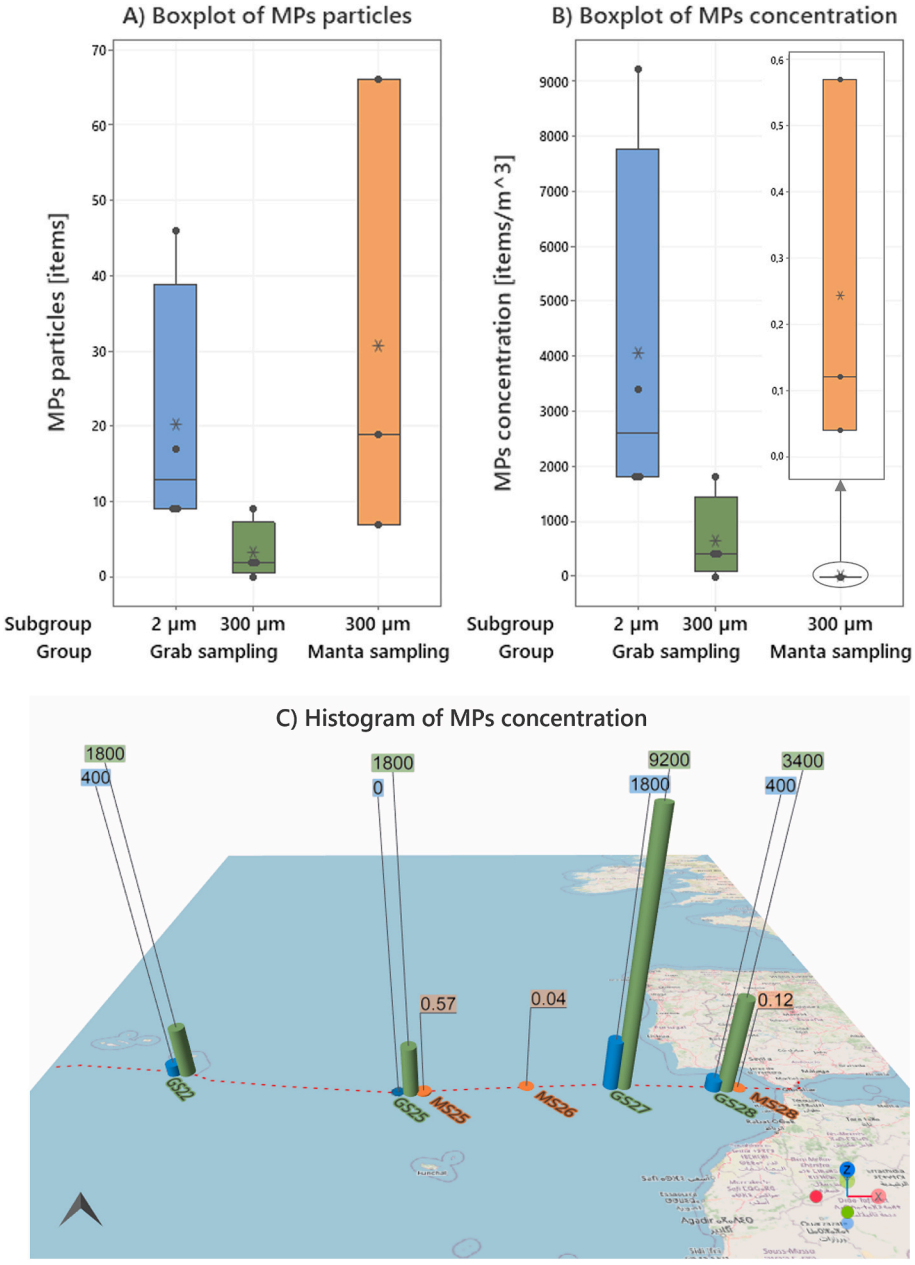

Fig. 1. Graphical representation of the results in terms of MPs abundance and concentration: A) boxplots representing the MP particles [items]; B) boxplots representing MP concentrations [items/m^3]; C) histograms representing MP concentrations [items/m^3] at the different sampling sites. Blue and green boxplots and histograms represent the abundance and concentration resulting from the grab sampling technique (dimensional cut-off utilized during the analysis phase equal to 2 and 300 μm, respectively), orange boxplots and histograms the abundance and concentration obtained with the Manta sampling technique. In A) and B) black dots represent the values of single measurements, the inner black line the median value and the stars the mean.

Table 1
Towing start and final coordinates and towing distance for the Manta trawl samples, sampling coordinates for the grab samples, volume of sampled water, number of MPs and MP concentrations found in the samples.

	A) Manta trawl approach (300 μm cut-off)		
	MS25	MS26	MS28
Towing start coordinates [°]	35.927633 N; 17.5933 W	36.18055 N; 14.0323 W	36.0634 N; 7.226417 W
Towing final coordinates [°]	35.935517 N; 17.537417 W	36.18705 N; 13.955383 W	36.061967 N; 7.149517 W
Towing distance [m]	5109	6943	6916
Volume of sampled water [m^3]	114.95	156.22	155.61
MP particles [items]	66	7	19
MP concentration [items/m^3]	0.57	0.04	0.12

	B) Grab sampling approach (2 μm cut-off)			
	GS22	GS25	GS27	GS28
Sampling coordinates [°]	36.945595 N; 25.145971 W	35.916133 N; 17.715467 W	36.21085 N; 10.9254 W	36.07195 N; 7.6859 W
Volume of sampled water [m^3]	0.005	0.005	0.005	0.005
MP particles [items]	9	9	46	17
MP concentration [items/m^3]	1800.00	1800.00	9200.00	3400.00

	C) Grab sampling approach (300 μm cut-off)			
	GS22	GS25	GS27	GS28
Sampling coordinates [°]	36.945595 N; 25.145971 W	35.916133 N; 17.715467 W	36.21085 N; 10.9254 W	36.07195 N; 7.6859 W
Volume of sampled water [m^3]	0.005	0.005	0.005	0.005
MP particles [items]	2	0	9	2
MP concentration [items/m^3]	400.00	0.00	1800.00	400.00

estimates of (Courtene-Jones et al., 2022) and (Montoto-Martínez et al., 2022): by multiplying the travelled distance by the area of the mouth of the Manta net (0.045 m^2) and subsequently dividing the result by two, as it was assumed that the net undergoes vertical oscillation and maintains a partially submerged state on average. The water volume (specified in Table 1) served as a normalization factor in quantifying the abundance of MPs in surface water samples (items/m^3).

The collection of bulk water samples from surface water was conducted using a metallic basket deployed from the vessel's side. The content was subsequently transferred into glass vials for further processing in the laboratory. Each sample, consisting of a volume of 5 l of surface water, was geographically tagged using GPS coordinates (Table 1). To determine MPs concentration (expressed as items/m^3), the number of MPs in each sample was divided by the corresponding sample volume.

In addition, several physical-chemical parameters of water were measured using probes at each MP sampling location. Temperature, conductivity and salinity were measured by means of the Multi Probe System YSI 556 MPS (YSI Incorporated), dissolved oxygen using an LDO101 probe (Hach Company) and pH a PHC101 probe (Hach Company). Although these data are not directly relevant to the current study, they are nonetheless included in the supporting information (Table S1) for the sake of completeness and to contribute to the database that may prove valuable for future research endeavors.

2.2. Sample processing

The collected samples were processed onshore and all treated according to the same processing protocol, regardless of whether the samples were obtained through employment of the Manta net or with the grab sampling approach. Samples were vacuum filtered through Whatman® glass microfiber filters (Grade GF/C, 1.2 μm, 47 mm), in order to separate plastic materials from water. No chemical digestion was performed, as the water appeared transparent on visual inspection and free from organic contamination. This choice was subsequently supported by the rapidity of the filtration process, which did not result in clogging of the filter due to organic matter.

At this point, the filter paper was removed from the filtration funnel and placed onto a clean ceramic surface and the MPs were recovered by tape lifting, according to the procedure described in Gwinnett et al. (2021b). Specifically, the adhesive surface of a forensic tape was gently and repeatedly stuck to the filter surface until all the debris had been lifted. The tape was consequently adhered to a glass microscope slide to immobilize MPs. The forensic tape used, Easylift®, is manufactured by Tecman Ltd. and is available from Staffordshire University. This technique offers several advantages, including protecting the sample from airborne contamination during analysis and eliminating the risk of accidental loss of MPs, which cannot be achieved using conventional MPs analysis directly from the filter. Furthermore, Gwinnett et al. (2021b) demonstrated that tape lifting is compatible with a diverse array of non-destructive analytical techniques, including Raman spectroscopy, employed in this study for the chemical identification of polymers and counting of related particles. Nevertheless, an assessment was conducted as part of this study to ascertain compatibility of the tape lifting technique with Raman spectroscopic analysis. To achieve this, a micro-Raman (*inVia* Renishaw) was utilized, interfaced with a Leica DM-LM microscope. A comprehensive mapping of an entire region (2500 × 2000 μm) within a microscope slide sample was conducted (Fig. S1), in order to evaluate the potential existence of background signals that might interfere with the identification of MPs, as well as to confirm the ability to recognize polymeric particles beneath the Easylift® tape.

2.3. Sample analysis

At this stage, a physical and chemical characterization of MPs was performed to evaluate size, concentration (items/m^3) and abundance by polymer type, using a micro-Raman spectrometer (*inVia* Renishaw) under 633 nm excitation, interfaced with a Leica DM-LM microscope (10× magnification and 0.25 NA). In contrast with numerous other studies, the entirety of particles was chemically analyzed and imaged (Piarulli et al., 2022), rather than just a subsample (Rosso et al., 2023).

Initially, each laboratory glass slide was mapped by dividing the glass slides into three square sections, each measuring approximately 2 cm on each side. Each section was subjected to microscope imaging under transmission white light. A pattern recognition Matlab algorithm was used to select particles exceeding a given threshold (300 μm for Manta trawl samples, 2 μm for grab samples). The selection of threshold for the Manta trawl methodology was based on alignment with mesh size of the Manta net. In contrast, the threshold for bulk sampling was determined in order to be larger than filter porosity utilized during the filtration process (1.2 μm), while at the same time sufficiently small to highlight contrasting levels of MP abundance observed between the two sampling techniques. However, in the case of grab sampling, the Matlab algorithm was also utilized for the selection of particles larger than 300 μm, in order to allow for a quantitative comparison between the two sampling techniques at an equal dimensional threshold.

Micro-Raman spectra were therefore acquired only over selected coordinates. Spectra were recorded using a 633 nm argon laser

excitation at ~3 mW, with 10× magnification objective and 10s acquisition time for each spectrum. Two windows were acquired in static mode, centered at 1500 and 3000 cm^{-1}, and about 600 cm^{-1} width. The spectra were baseline subtracted and automatically compared with SLoPP and SLoPP-E Raman Library (Munno et al., 2020).

Dimensional characterization of MPs was conducted utilizing the mean Feret diameter (μm). Feret diameter is a measurement commonly used in microscopy in assessing particle sizes, particularly on two-dimensional representations of three-dimensional objects. It is defined as the distance between two parallel lines tangential to MPs in a precisely defined orientation (Rosal, 2021). Mean Feret diameter was determined by calculating the average between the maximum and minimum Feret diameters for each individual MP.

Chemical characterization of MPs was accomplished using the Pearson correlation parameter, a widely employed analysis for assessing the degree of linear correspondence between an unknown spectrum (derived from the sample) and a reference spectrum (obtained from the abovementioned libraries). A Pearson correlation value equal to 1 signifies a perfect linear correlation between two spectra, whereas a Pearson correlation value equal to 0 indicates no correlation (Levermore et al., 2020). Attributions were only provided by Pearson's correlation >0.6, as this threshold was deemed a suitable trade-off for reliable attributions. In cases where lower Pearson correlation values were obtained, no attributions were provided, and the particle was classified as "unknown".

The decision to utilize micro-Raman spectroscopy for analysis of MPs was made based on the premise that the glass microscope slide (and adhesive tape) does not impede collection of Raman spectra, unlike IR spectra where radiations are absorbed. Furthermore, Micro-Raman spectroscopy has demonstrated reliability in detecting even the finest size fraction of MPs, including particles smaller than 10 μm, where FTIR spectroscopy has proved to be of limited efficacy (Luo et al., 2023).

2.4. Contamination prevention

Stringent precautions must be adopted throughout all stages of MP studies to mitigate potential procedural contamination of samples, i.e. "any anthropogenic microparticles that have entered the sample during sampling and processing that was not part of the original sample taken from the environment" (Gwinnett and Miller, 2021), such as contaminants from analysts' clothes, airborne sources, laboratory surfaces or from equipment being used. Protocols for the prevention of contamination described in (Gwinnett and Miller, 2021) and (Prata et al., 2019) were applied during sampling and sample processing.

In order to minimize procedural contamination while collecting samples in the Atlantic Ocean, cotton clothes made of non-synthetic polymers were worn by the sole person in charge of sampling throughout the entire process. Additionally, all potential sources of MP contamination were replaced with non-plastic alternatives: a metallic basket was employed for grab sampling and glass jars were used for storage of oceanic samples. Prior to usage, glass jars were triple rinsed thoroughly with freshwater, while the metallic basket and Manta net were rinsed with seawater. Before transferring the sampled water from the metallic bucket or cod-end of the Manta, the sampler ensured they were downwind of the water being transferred, while the glass jars and lids were kept face down until used to reduce air exposure time and potential airborne contamination.

During laboratory sample processing, the following actions were implemented to minimize the risk of procedural contamination:

- Cotton clothing and cotton lab coats were worn by the sole analyst in charge of all sample processing activities;
- All equipment and surfaces were cleaned prior to use with Milli-Q water and alcohol;
- Use of plastic equipment was avoided;
- The glass funnel was covered with a glass disc during the filtration process;
- Procedural blanks were run as negative controls by creating blank samples made up of milli-Q water in equivalent volumes to real samples (5 l for grab sampling and approximately 1 l for Manta sampling), as detailed in (Prata et al., 2021). The two blank samples – one for each sampling method – underwent identical processing procedures as samples (vacuum filtration and tape lifting) and identical analysis (micro-Raman spectroscopy with dimensional cut-off equal to 300 μm for Manta trawl, 2 μm for grab sampling);

After the filtration process, actions needed to immobilize MPs on the microscope slide using forensic tape were promptly executed to minimize the potential for sample contamination by airborne MPs. Subsequent to the successful execution of these procedural steps, the risk of contamination was effectively mitigated to zero, with the adhesive Easylift® tape acting as a barrier against airborne contamination.

2.5. Statistical analysis

The use of statistical analyses for the purpose of comparing MPs concentration data between sampling methods was deemed inappropriate due to the limited number of samples (7 in total, 4 relating to grab sampling and 3 Manta trawl approach), which would have compromised the reliability and robustness of any statistical evidence, also in light of the high variability of results (illustrated in Section §3.2). Furthermore, it should be underlined how the main focus of the article is on methodological emphasis rather than quantitative assessment of the abundance of microplastics in the aquatic environment.

Pearson's *t*-test was performed to examine the potential correlation between MP concentration data (expressed as items/m^3) for the two sampling methods and environmental parameters measured at the sampling sites, including temperature, conductivity, salinity, pH and dissolved oxygen. Statistical significance was set at 0.05. Normality was accessed using the Anderson-Darling test, which was verified (*p*-value >0.05).

Furthermore, concentration data obtained using the Manta trawl sampling approach were compared with concentration data from other studies conducted in the same macro-area (North Atlantic Ocean) using the same sampling technique (net-based methodology). The normality of data was assessed using the Anderson-Darling test, while the assumption of equal variances was evaluated using the Levene test. Significance was set at 0.05 for both cases. Data adhered to the assumption of homogeneity of variances (p-value = 0.592), but not to parametric assumption of normality (p-value <0.005). Therefore, for a dataset comparison, the non-parametric Mann-Whitney test was applied, with significance set at 0.05.

All statistical analyses were performed using Minitab statistical software v20.1.1.0.

3. Results

3.1. Compatibility of the Easylift® tape with Raman spectroscopy

The suitability of the forensic tape Easylift® for Raman spectroscopy analysis was previously demonstrated by (Gwinnett et al., 2021b). However, as part of this study, a validation experiment was conducted to assess compatibility of the tape lifting technique with this non-destructive analytical technique. A thorough mapping of an entire region (2500 × 2000 μm) on a microscope slide sample was performed (Fig. S1), with a micro-Raman (*inVia* Renishaw), to evaluate the presence of any background signals that could potentially interfere with the identification of MPs, as well as to verify the ability of recognizing polymer particles beneath the Easylift® tape. Micro-Raman spectra were acquired at a resolution of 50 μm over the selected area, compared with SLoPP and SLoPP-E Raman Library.

Background signals exhibited Pearson's Correlation values below the predefined acceptability threshold chosen for this study (0.6), indicating that the glass microscope slide and the Easylift® tape could not be identified as MPs within the sample analysis. Simultaneously, the MP fragments actually present in the mapped area displayed significantly high Pearson's correlation values (> 0.75), suggesting that MPs can be recognized with good accuracy under the Easylift® tape (Fig. S1 B and S1 C).

3.2. Abundance

The cumulative water volume filtered using the grab sampling method amounted to 20 L, which translates to 5 L per sample. Conversely, the estimated volume sampled using the Manta trawl was calculated based on dimensions of the mouth opening and the distance covered during towing, divided by two, resulting in a total of 426,780.00 l, with an average of 142,260.00 ± 23,650.00 l per sample (Table 1).

As evident from Table 1 A and B, all collected samples were found to contain MPs, with a total of 173 particles identified. Out of these, 92 particles (53.18 %) were captured through the Manta trawl, while 81 particles (46.82 %) were collected via grab sampling. Specifically, the volume reduction approach retrieved an average of 30.67 particles (min. = 7; max. = 66; SD = 31.18), against the 20.25 (min. = 9; max. = 46; SD = 17.58) collected with the grab sampling approach.

The choice of sampling approach exerts a substantial influence on the concentration of MPs in the sample, as highlighted in previous studies (Barrows et al., 2017; Montoto-Martínez et al., 2022; Prata et al., 2019; Song et al., 2014). In the present study, the findings revealed a significantly higher concentration of MPs larger than 2 μm when employing the grab sampling approach, in contrast to MPs larger than 300 μm obtained through the Manta trawl approach (Table 1 A and B, Fig. 1 B and C – orange and green colored boxplots and histograms). On converting the data to represent the concentration per volume of water sampled, the average concentrations observed were 0.24 items/m^3 (min. = 0.04; max. = 0.57; SD = 0.29) for the Manta net method and 4050 items/m^3 (min. = 1800; max. = 9200; SD = 3515) for the grab sampling approach (Table 1 A and B, Fig. 1 B and C – orange and green colored boxplots and histograms).

In order to compare the two different sampling methods at an equal mesh size, data relating to grab sampling were reprocessed with a dimensional cut-off of 300 μm, equivalent to the mesh size of the Manta net. A total of 13 MPs were found in the samples with a 300 μm threshold, compared to the 81 MPs found in the same samples with a 2 μm threshold. This translates to approximately six times fewer particles as the used dimensional threshold increased. The mean number of MPs found in the 300 μm threshold grab samples was 3.25 (min. = 0; max. = 9; SD = 3.95), corresponding to an average concentration of 650 items/m^3 (min. = 0, max. = 1800, SD = 790) (Table 1 C, Fig. 1 B and C - blue colored boxplots and histograms). At an equal mesh size, the average concentration of MPs derived from Manta sampling is significantly lower than that derived from grab sampling (0.24 vs. 650 items/m^3). However, the difference in concentration between the two methods is even more pronounced when the same mesh size is not ensured (0.24 vs. 4050 items/m^3).

Two procedural blanks, one for each of the two different sampling methods, were run in parallel to samples to investigate potential cross-contamination from MPs during the processing phase. Consequently, volumes of the samples forwarded to the laboratory after oceanic sampling were reproduced using MilliQ water: 5 l for grab sampling and 1 l for Manta sampling (obtained from the onboard net cleaning procedure). A total of 2 MPs were detected on the two procedural blanks: specifically, 2 particles were found on the control filter representing the grab sampling technique, while no particles were observed on the control filter associated with the Manta trawl approach. The discrepancy in number of MPs detected between the simulated grab sample and Manta sample can be attributed to two factors. Firstly, the difference in processed volume, with grab sampling processing a larger volume (5 l) compared to the volume processed in Manta sampling (1 l). Additionally, the applied cut-off size for detecting plastic particles varied between the two sampling approaches: 2 μm for grab sampling and 300 μm for Manta sampling. As a result, the wider dimensional range targeted for MP detection in the grab sampling technique increases the probability of finding more particles compared to Manta sampling. From a purely speculative perspective, if a 300 μm cut-off was applied to the procedural blank representing the grab sampling technique, the absence of particles resulting from procedural contamination during the processing phase could be observed. This observation is in line with considerations that will be made in Section §4, indicating a higher likelihood of detecting a greater number of MPs as the dimensional threshold used in the analysis phase decreases.

In general, the low number of MPs found in the procedural blanks confirms the effectiveness of the contamination prevention measures implemented during the processing phase. Additionally, it is important to note that the potential contamination of MPs during the analysis phase is eliminated through use of the tape lifting technique, which allows the sample to be "frozen" as is after processing. Indeed, application of the barrier provided by the Easylift® tape prevents the sample from being contaminated with airborne particles during the analysis phase.

A Pearson's correlation *t*-test was performed to explore the correlation between MPs concentration (items/m^3) for the two sampling methods and the measured environmental parameters (temperature, conductivity, salinity, pH, and dissolved oxygen). For the grab sampling technique the correlation was investigated for both the 2 μm and the 300 μm dimensional cut-off applied in the sample analysis phase. No physical-chemical parameters demonstrated a statistically significant correlation with MP concentration for the two performed sampling approaches (*p*-values always >0.05). The results are presented in Table S2.

3.3. Size

By analyzing the results based on size distribution of particles collected with the Manta trawl, we observed a prevalence of particles ranging from 500 to 1000 μm in sample MS25 and from 350 to 500 μm in sample MS28, while sample MS26 exhibited only 7 particles, distributed approximately equally within the 400–700 μm range (Fig. 2, first row). In contrast, when examining the size of MPs obtained from grab sampling, the majority were found to be within the 50–150 μm range in sample GS22, 100–200 μm in samples GS27 and GS28, while sample GS25 only showed the presence of 9 particles, distributed nearly equally across the 2–300 μm range (Fig. 2, second row). However, analysis of the size of MPs from grab sampling by examining the presence of fragments smaller than 300 μm is particularly relevant as it emphasizes the distinction between the two sampling approaches. As expected, the significant majority of MPs fell within the 2–300 μm range: 77.78 % for GS22, 100 % for GS25, 76.09 % for GS27, and 88.24 % for GS29, with an overall mean of 85.53 % (± 11.04 % SD). Consequently, both these data and those relating to MPs concentration, underline the greater reliability of volume selective sampling approaches in accurately quantifying the true extent of the issue of MPs pollution in water compared to size selective approaches.

3.4. Polymer types

A total of 11 polymers were identified in surface water (Fig. 3). In the three samples collected using the Manta trawl approach, only four distinct polymer types were identified: polyester (63 %), polycarbonate (24 %), cellulose acetate (12 %), and polystyrene (1 %) (Fig. 3, first row). In contrast, analysis of the composition resulting from the grab sampling approach revealed a much broader range of polymer types, including cellulose acetate (42 %), polycarbonate (24 %), polypropylene

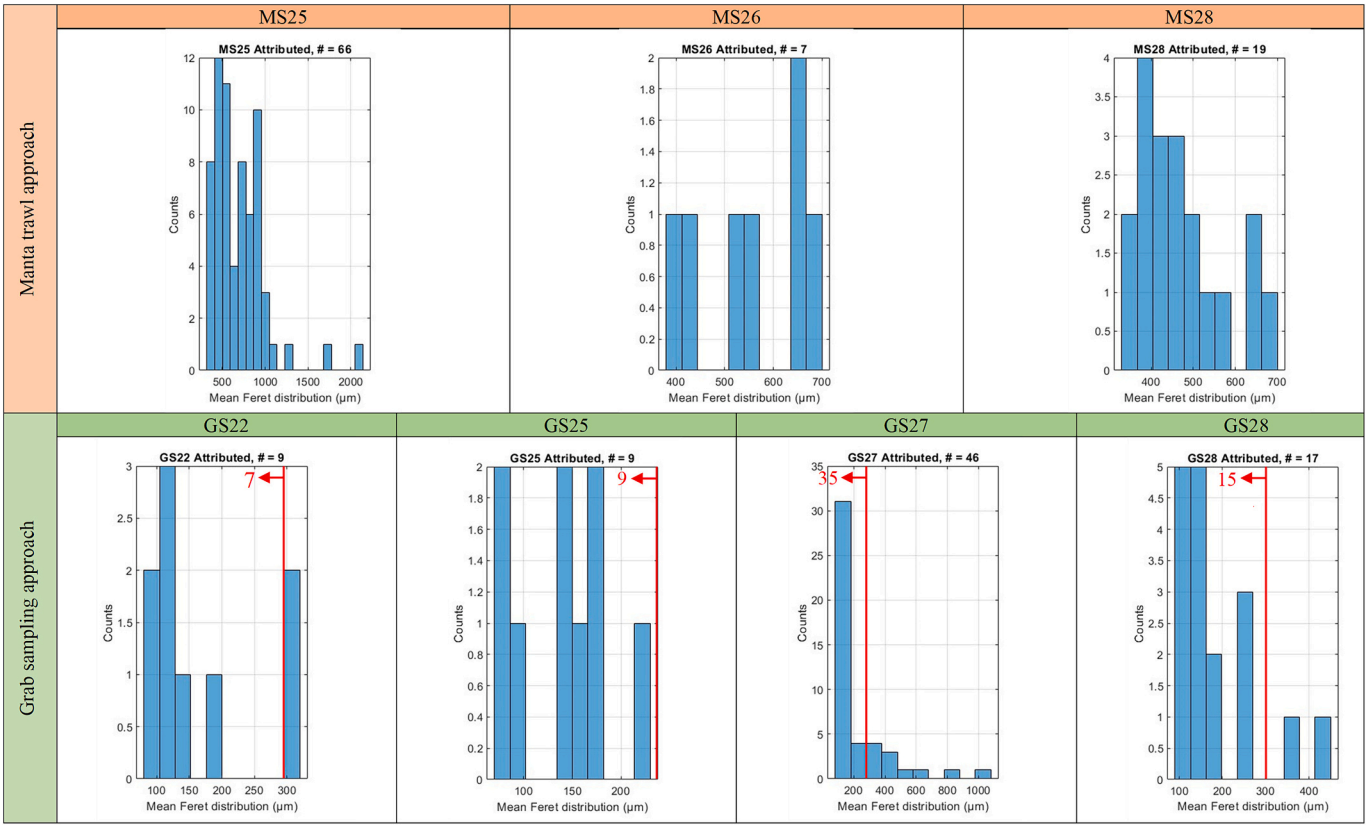

Fig. 2. Histograms representing MPs size distribution in the different samples. Only MPs with a mean Feret diameter bigger than 300 µm (Manta sampling) and 2 µm (grab sampling) were analyzed. The vertical red lines for grab sampling approach represent the 300 µm threshold, useful for comparison between the two sampling approaches, while the red numbers represent the total particles smaller than that threshold.

(21 %), polymethyl methacrylate (8 %), acrylonitrile butadiene styrene (1 %), nylon (1 %), polyacrylonitrile (1 %), polyvinylchloride (1 %), and styrene/isoprene (1 %) (Fig. 3, second row).

However, distribution of the polymers was highly heterogeneous across the sampling locations (Fig. 3, third row). The only polymer observed in all samples was cellulose acetate, ranging from 5 % to 86 %. Polyester was only found in samples collected using the Manta trawl approach. It is interesting to note that in two pairs of successively collected samples (GS25 and MS25, GS28 and MS28), their compositions in terms of polymer types differed significantly, particularly in the latter pair. This is due to the high variability in spatial distribution of MPs in aquatic environments (Pasquier et al., 2022; Tamminga et al., 2019), subsequently reflected in the samples, particularly when sampled volumes are too small to be considered representative. To overcome this issue, sampled water volume or the number of samples collected at each sampling location should be increased (Barrows et al., 2017; Pasquier et al., 2022; Prata et al., 2019; Schmid et al., 2021b; Song et al., 2014; Tamminga et al., 2019).

4. Discussion

This study demonstrates the widespread occurrence of MPs in the surface waters of the North-east Atlantic Ocean. Regardless of the sampling method employed, MP particles were detected in all samples, exhibiting concentrations ranging from 0.04 to 9200 items/m^3.

Although the main objective of the study was to demonstrate the benefits and limitations of two different sampling methodologies, the possibility of quantifying MPs in the open ocean along routes not normally monitored for this purpose provided the opportunity to create a baseline of information on MPs pollution in the North-East Atlantic Ocean, from the Azores to Gibraltar. Furthermore, given the prevalence of the size selective method for MPs sampling in surface water in the context of a range of sampling techniques (GESAMP, 2019; Karlsson et al., 2020; Lenz and Labrenz, 2018; Lindeque et al., 2020; Montoto-Martínez et al., 2022; Pasquier et al., 2022; Shim et al., 2022), data relating to MPs obtained in this study using the Manta trawl were compared with those deriving from other studies conducted in the same macro-area (North Atlantic Ocean) based on the same sampling principle (net-based approaches), in order to investigate their consistency. Therefore, a systematic literature review was conducted on October 31, 2023, consulting two scientific databases (Web of Science, www.webofknowledge.com; SCOPUS, www.scopus.com) and using the following construction for the three keyword strings formulation (Eq. (1)):

$$NA + MP + SM \quad (1)$$

where:

NA = "North Atlantic" or its synonyms, such as "Northern Atlantic Ocean", "North-east Atlantic", "North-west Atlantic", etc.

MP = "microplastic" or its synonyms. Subsequently, for a more precise research, the names of the most widely-used polymers (polyamide, polyethylene terephthalate, polyester, polypropylene, polystyrene, polyvinyl chloride, polyethylene) were included.

SM = "sampling method" or its synonyms such as "sample collection", "sampling approach", etc. The terms "grab sampling", "Manta trawl", "neuston net" and their synonyms were also used.

The search yielded 384 articles (excluding duplicates), which underwent two consecutive screenings, the first focused on the exclusion or acceptance of an article based on analysis of title and abstract, the second centered on the ultimate acceptance of articles selected in the first screening and reading of the entire article text. Review articles were excluded from the analysis, as well as studies pertaining to MPs in media

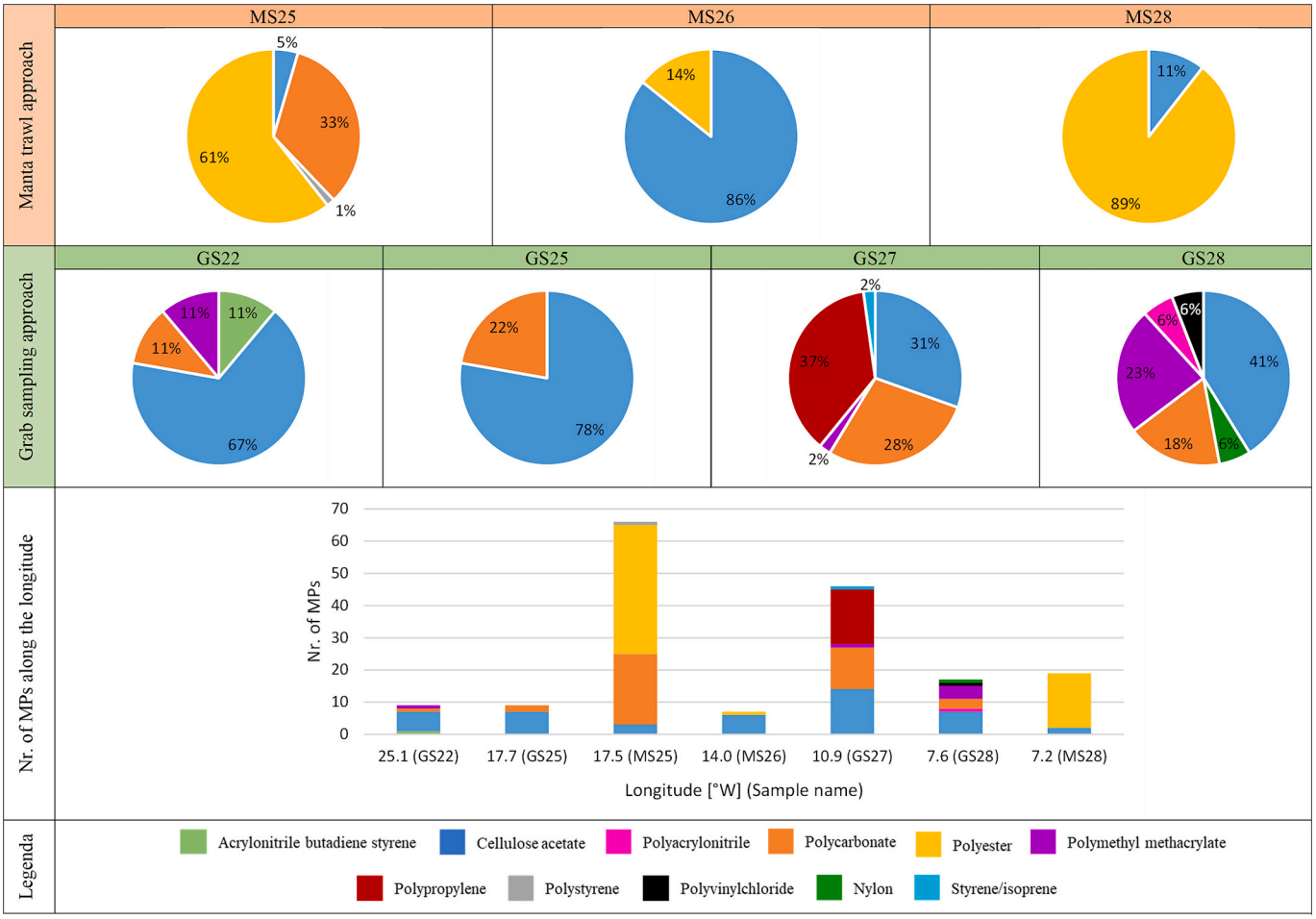

Fig. 3. Composition by polymer type of MPs collected with the Manta trawl approach (first row) and the grab sampling approach (second row). MPs composition by polymer type in the different samples, ordered by longitude from the Azores (leftmost) to Gibraltar (rightmost) (third row).

other than surface water (such as sediments and subsurface water) or ingested by biota, or freshwater-related articles. Moreover, any articles focused on sampling sites outside of the North Atlantic Ocean, including the Gulf of Mexico, Caribbean Sea, North Sea, Baltic Sea, Wedden Sea, Arctic Sea, Mediterranean Sea, Russian Sea, Iceland Sea, and Scotia Sea, were also excluded. Finally, only articles in which the concentration of MPs was expressed as items/m^3 were considered, thus avoiding studies reporting the concentration on an areal basis (items/km^2), in order to facilitate comparison between data from the review and the present study. As shown in Fig. 4A, 355 articles were excluded on initial screening, while 19 articles were excluded during the second screening, focusing the literature review on the analysis of 10 texts. The reasons for exclusion for both screening phases are illustrated in Fig. 4B. The primary reasons for exclusion during the first screening were the presence of articles not directly related to MPs (at times dealing with marine waste litter in general, plastic additives, bacterial biofilm formed around polymer fragments, etc.), articles not focusing on the North Atlantic Ocean, and articles encompassing both of the aforementioned exclusion criteria. Exclusion percentages based on these reasons were 55 %, 12 %, and 16 %, respectively. On the other hand, the main reasons for exclusion during the second screening were the presence of articles discussing MPs abundance in non-surface water (usually subsurface water, but also sediments or beaches), the use of measurement units other than volumetric particle concentration (typically items/km^2 instead of items/m^3), and articles not pertaining to the North Atlantic Ocean. The exclusion percentages for these reasons were 53 %, 16 %, and 11 %, respectively.

Table S3 illustrates the ten articles that successfully passed both selection stages and were thus utilized to compare the concentration of MPs in surface water in the North Atlantic Ocean with the data obtained from this study. For each included study, the following information was extracted: year of sampling, sampling location, sampling method used and characteristics (number of samples, trawling speed and time, transect length, sampled volume, sampling depth, mouth opening dimensions of the net and net mesh size), method used for calculation of the sampled water volume, MP concentration and unit of measurement used.

A comparison of data relating to MPs concentration from the review and those obtained in the present study using the Manta trawl sampling approach was performed by means of the non-parametric Mann-Whitney statistical test with a significance level set at 0.05. The assumption of equal variances was verified, while normality was not met. No statistically significant differences were observed between the two datasets, as indicated by a *p*-value of 0.08. Consequently, MPs concentration data obtained in the present study align with those derived from similar studies conducted in surface water in the North Atlantic Ocean. Fig. 5 A depicts the boxplot of MPs concentration data obtained from the literature review and its zoomed-in version. The two outliers in the graph (empty rhombuses) correspond to an MPs concentration of 8.03 and 66.68 items/m^3, obtained from the studies of Lindeque et al. (2020) and Montoto-Martínez et al. (2022), respectively. In these investigations, oceanic surface water was sampled using devices with a mesh size of 100 and 50 μm, i.e. a smaller dimensional threshold compared to those commonly used for the net-based sampling approach. The fact that a smaller sampling mesh size is associated with higher MPs concentration is not surprising and will be extensively discussed later in this Section. The mean MPs concentration per volume obtained using the Manta

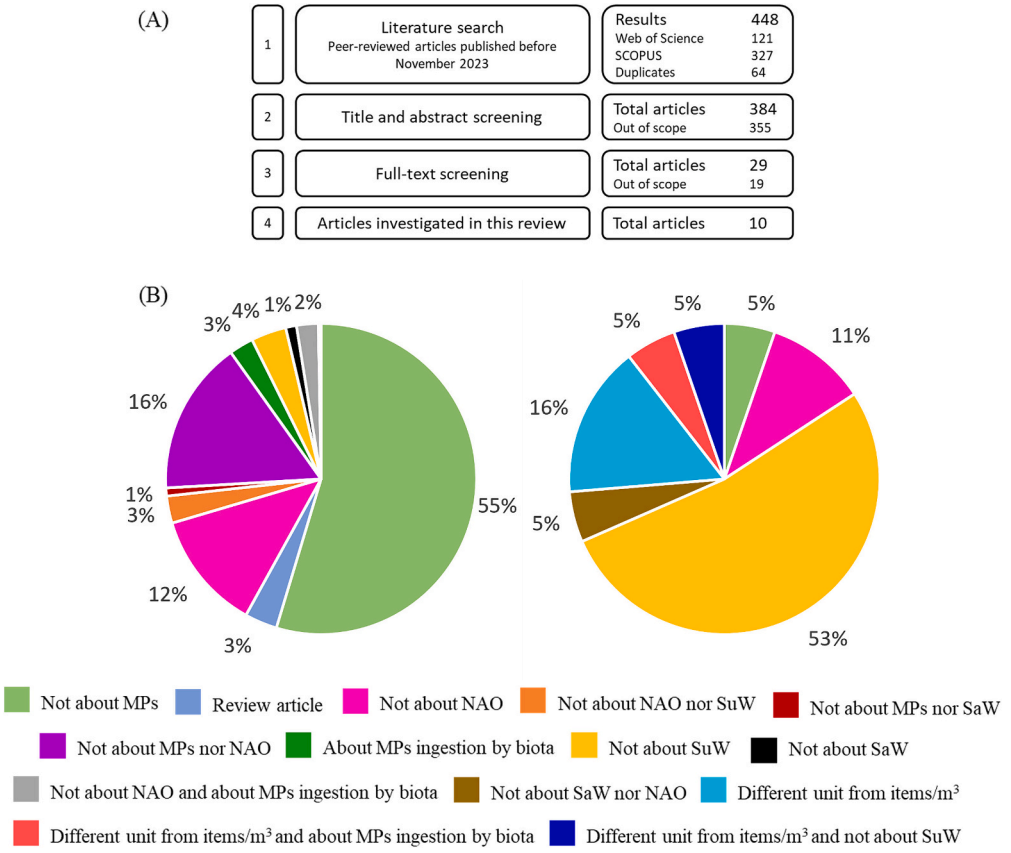

Fig. 4. Flow diagram showing the selection process applied in the literature review (A) and reasons for exclusion (B) applied in the first screening (left) and in the second screening (right) of the literature review. Legend: MPs = MPs; NAO = North Atlantic Ocean; SuW = Surface Water; SaW = Salt Water.

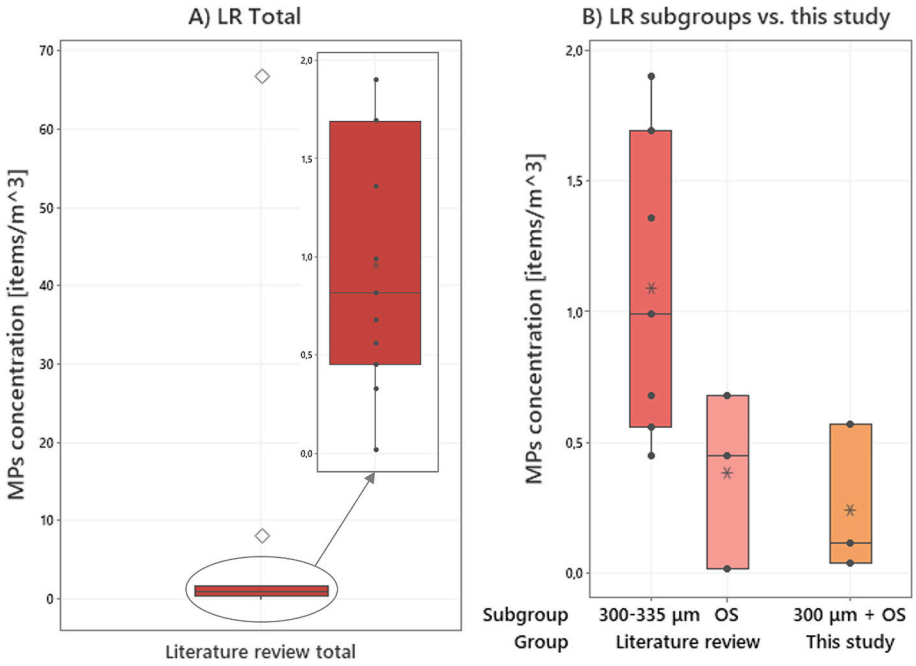

Fig. 5. Boxplots representing MPs concentration [items/m3] obtained from A) the literature review when all selected studies were considered and B) from the literature review when limited to 300–335 µm net mesh size (left-most), from the literature review when limited to open sea waters (central), from the present study with the Manta trawl sampling approach (right-most). The empty rhombuses represent the outlier values, the black dots values of the single measurements, the inner black lines the median value and the stars the mean. Legenda: LR = literature review; OS = open sea.

trawl approach in the present study corresponded to 0.24 items/m^3. This value falls within the lower whisker of the boxplot of Fig. 5 A and is therefore comparable with the other literature values, particularly considering the inherent high variability in these measurements. This variability can be attributed, among other factors, to differences in mesh size used in the various studies (varying between 50 and 500 μm) and uncertainties associated with measurement of the sampled water volume. It is not surprising that the average concentration obtained in the present study is slightly lower than the mean concentration obtained in the literature review, since sampling was conducted in the open ocean rather than in coastal areas, as was the case for the majority of studies considered in the literature review, thus resulting in a reduced abundance of plastic particles.

To achieve a more accurate and reliable comparison, analysis was subsequently limited to the results from the literature review that corresponded to a sampling net mesh size comparable to the size used in the present study. Specifically, only studies employing a net-based sampling methodology with a mesh size ranging from 300 μm to 335 μm were included (Fig. 5 B, the left-most boxplot), which represents a frequently employed cut-off interval in numerous investigations focusing on MPs (GESAMP, 2019; Karlsson et al., 2020; Lenz and Labrenz, 2018; Pasquier et al., 2022; Prata et al., 2020). The concentration values obtained in this study (represented by the right-most boxplot in Fig. 5 B) are in line with those obtained from the literature review, particularly when considering studies that focused on open ocean sampling rather than those conducted along the coastlines bordering the Northern Atlantic Ocean (graphically represented by the central boxplot in Fig. 5 B). In particular, the studies of Courtene-Jones et al. (2022), Kooi et al. (2016) and Silvestrova and Stepanova (2021) reported average concentration values for surface water in open Northern Atlantic Ocean equal to 0.45, 0.68 and 0,02 items/m^3, respectively, very close to the mean value of 0.24 particles per cubic meter obtained in the present study.

In Fig. 6, MPs concentration values obtained from the literature review (red dots) and from the current study using the Manta trawl (orange rhombuses) were plotted against the mesh size used for sampling. This representation of the results further emphasizes the consistency of the data obtained in the present study with those from previous investigations. Additionally, on the same graph, concentration values obtained in the present research by means of the grab sampling technique (green squares) were plotted to further give supplementary prominence to the indirect proportionality between abundance of MPs and dimensional threshold used. This relationship will be further demonstrated later in this Section.

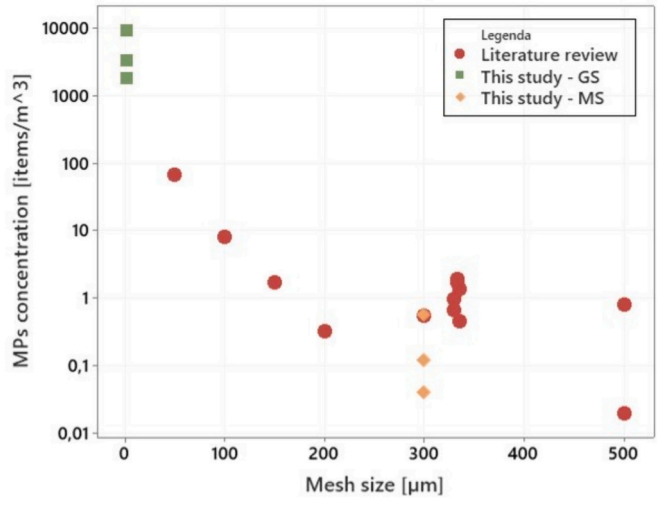

Fig. 6. Scatterplot of MPs concentration values [items/m^3] obtained from the literature review (red dots) and from the present study through the Manta trawl (orange rhombuses) and the grab sampling approach (green squares) against mesh size [μm] employed for sampling. Legenda: GS = grab sampling; MS = Manta sampling.

Similarly, concentration values obtained with grab sampling (with an average value of 4050 items/m^3) align with the extrapolation made by Lindeque et al. (2020), suggesting that for a mesh size of 1 μm the concentration would exceed 3700 particles per cubic meter.

To conclude, MPs concentration values obtained in this study using two different sampling approaches are consistent with those obtained in other studies in the same area. The main methodological innovation adopted in this investigation, compared to others, is the use of the tape lifting technique during the sample processing phase. By employing this approach, the direct analysis of MPs from the filter paper is avoided, thereby preventing accidental loss of polymeric particles and potential contamination from airborne MPs. Other advantages of this methodology are its cost-effectiveness and the ease of transportability of tape lifts which can then be preserved for future studies. Moreover, the technique is compatible with a wide range of non-destructive analytical techniques, including Raman spectroscopy – as demonstrated in Gwinnett et al. (2021b) and here in Section §3.1 –, used in this study for the chemical identification of polymers. The ability to recover MPs from filter papers was confirmed by the studies of Schotman and van der Weerd (2015) and Gwinnett et al. (2021b), in which the mean percentage recovery rate corresponded to 94.5 % and to 96.55 % (when using a glass funnel type and a glass fiber filter tape, as in the case of the present application), respectively.

The choice of sampling methodology exerted a notable impact on the estimated concentration of MPs (Shim et al., 2022), with the grab sampling approach used in this study recording concentrations up to five orders of magnitude higher than those obtained with the Manta net. Numerous investigations have undertaken a comparison of sampling methods using different mesh sizes for MPs (Barrows et al., 2017; Di Mauro et al., 2017; Green et al., 2018; Lindeque et al., 2020; Montoto-Martínez et al., 2022; Song et al., 2014). In general terms, this study lent further support to the assertion made by Miller et al. (2021), Barrows et al. (2017) and Green et al. (2018) that the abundance of MPs in the marine environment is contingent upon the specific sampling methodology employed. Indeed, their findings demonstrated that the grab sampling method revealed a concentration of MPs per water volume up to two, three or four orders of magnitude higher compared to that obtained with the commonly used zooplankton methods (Manta, bongo, neuston and plankton nets). The same considerations can be extrapolated from the study conducted by Di Mauro et al. (2017), which confirmed the presence of up to four orders of magnitude more MPs in Niskin bottle samples (followed by a filtration to 0.7 μm) compared to net-based approaches (such as bongo and neuston net, both with a net mesh size of 335 μm). Likewise, the study conducted by Song et al. (2014) confirmed a greater abundance of MPs in 100 l surface bulk water compared to that obtained with Manta net (330 μm), with an approximately four-fold, or order of magnitude, difference. The order of magnitude difference in MPs concentration between different sampling methodologies may also be expressed as an indirect proportionality between the abundance of MPs and mesh size used in the investigation (Karlsson et al., 2020; Pasquier et al., 2022; Shim et al., 2022; Simon-Sánchez et al., 2022). In other terms, utilizing smaller meshed nets when sampling translates into a considerably higher concentration of MPs compared to larger sized mesh nets (Du et al., 2022; Karlsson et al., 2020; Montoto-Martínez et al., 2022; Shim et al., 2022; Simon-Sánchez et al., 2022). In this regard, Lindeque et al. (2020) demonstrated that using nets with a 100 μm mesh size yielded 2.5 and 10 times more MPs than adopting nets with mesh sizes of 333 and 500 μm, respectively. Similarly, Kang et al. (2015) identified two orders of magnitude more MPs with a 50 μm mesh sized hand-net than with a 330 μm mesh Manta trawl, while Vermaire et al. (2017) found one hundred times more MPs using a 100 μm nylon net than a 333 μm Manta net. In addition, Song et al. (2014) demonstrated that the abundance of MPs obtained from bulk water samples and hand-net samples (mesh size of 50 μm) did not

result in significant differences, indicating how as the sampling size limit decreases, the number of captured MPs per unit volume increases, providing more representative outcomes of the true extent of the marine MP issue. As a result, direct comparisons between different studies that adopt varying minimum cut-offs in terms of mesh size of nets or filters, may lead to potentially significant errors in the evaluation of MPs pollution, with a tenfold increase in the number of MPs for each 121-μm decrease in the minimum net mesh size (Shim et al., 2022). In light of the aforementioned considerations, it is not surprising that the two outliers observed in Fig. 5 A (empty rhombuses) correspond to the concentration data obtained using a mesh size of 50 μm – 66.68 items/m^3 as reported by Montoto-Martínez et al. (2022) – and 100 μm – 8.03 items/m^3 as documented by Lindeque et al. (2020). These mesh sizes were the smallest employed among the articles selected for review. This evidence further reinforces the notion that as the mesh size used for sampling decreases, the concentration of MPs in the sample increases.

In the current case study, for the purpose of comparing the two sampling methods at the same mesh size, the grab samples were further analyzed imposing a hypothetical cut-off at 300 μm in the Matlab algorithm during the analysis phase, matching the mesh size of the Manta. The results, both in terms of MPs abundance and concentration, were downscaled by one order of magnitude compared to grab sampling with a 2 μm cut-off (average values of 3.25 items and 650 items/m^3 in the former case, 20.25 items and 4050 items/m^3 in the latter case), as expected therefore based on the considerations made up to this point. However, comparison of the two distinct sampling methods at the 300 μm cut-off revealed a significantly higher concentration of MPs for grab sampling versa Manta (650 ± 790 items/m^3 versus 0.24 ± 0.29 items/m^3). This result may appear inconsistent, as a similar order of magnitude in MPs concentration for the two different sampling methods at the same mesh size would have been expected. However, this discrepancy is easily explained by the observation that even a single MP in a grab sample would yield a concentration of 200 items/m^3 in a 5 l sample, thus representing a concentration three orders of magnitude higher than that obtained in Manta samples. In other words, the minimum possible concentration above zero, is 200 particles per cubic meter for 5-liter samples, equating to 1 particle. To achieve comparable results, it would have been necessary to grab sample volumes three or four orders of magnitude larger. This means that the sampled volume cannot be considered representative for the application of a 300 μm cut-off, or alternatively, grab sampling is not a suitable sampling method for larger MPs. However, the results obtained from the comparison of different sampling methods at the same mesh size in this study align with findings from the studies of Montoto-Martínez et al. (2022) and Du et al. (2022). Montoto-Martínez et al. (2022) compared the concentration obtained using a Manta net with a 200 μm mesh size with yield from an innovative MPs sampler (MuMi) incorporating a filter with a mesh size of 200 μm. A concentration two orders of magnitude lower was observed for the Manta compared to MuMi (0.3 ± 0.2 items/m^3 versus 23.3 ± 42.7 items/m^3), within a sampled volume three orders of magnitude larger (138.90 m^3 compared to 0.37 m^3). Du et al. (2022) compared the concentration of MPs obtained with two different pump filtration systems, both with a minimum filter mesh size of 100 μm. Specifically, they employed a trawl-underway pump in conjunction with an in-situ filtration device (Type I) and a stationary onboard pumping system coupled with an in-situ filtration device (Type II). For Type I, the researchers obtained an average concentration approximately three times lower than that achieved with Type II (2.56 ± 1.01 items/m^3 compared to 7.48 ± 6.92 items/m^3), within an average sampled volume roughly four times greater (17.21 m^3 versus 4.16 m^3). In a broader context, on comparing the concentration of MPs at the same mesh size for two distinct systems, it is evident that as the sampled volume decreases the concentration tends to increase, suggesting potential overestimation when dealing with small sample volumes (Lusher et al., 2014; Tamminga et al., 2019; Vermaire et al., 2017). Simultaneously, variance markedly diminishes with the increase in sampled volumes (Lusher et al., 2014; Tamminga et al., 2019). This phenomenon may stem from the fact that very low sampled volumes can lead to significant statistical uncertainties and serious overestimations when scaling up the results (Tamminga et al., 2019). To overcome this impasse, either the sampled volume could be increased or sampling methods unsuitable for the collection of large volumes could be used only in areas in which higher concentration levels are expected.

The enormous difference in the concentration of MPs obtained using the grab sampling approach and size-selective approaches can be easily explained by the theory of the size spectrum for particulate matter in the sea, as elucidated by Sheldon and Parsons (1967), which states that the abundance of particles is inversely proportional to their size in the aquatic environment, due to the fragmentation of larger plastic objects into smaller fragments.

The abovementioned studies concurred to consolidate the theory of a substantial underestimation of the quantity of MPs present in the aquatic environment when relying on net-based methodologies (Barrows et al., 2017; Du et al., 2022; Lindeque et al., 2020; Shim et al., 2022; Simon-Sánchez et al., 2022; Song et al., 2014), a consideration further supported by the results of the present study. This evidence however represents a particularly thorny issue when considering that 75.9 % of MPs studies are based on net tow methods (Shim et al., 2022) and 80 % of MPs sampling campaigns only accounted for polymers >300 μm in diameter (Conkle et al., 2018).

Another concern associated with the use of net-based methodologies stems from the amount of water sampled, with regard to both exact quantification and representativeness of the sampled volume (Montoto-Martínez et al., 2022). The inherent challenge related to the certainty of ascertaining the volume of sampled water consists in accurately quantifying submersion percentage of the net mouth owing to water turbulence due to waves, wind and boat movement (Green et al., 2018; Karlsson et al., 2020; Montoto-Martínez et al., 2022; Shim et al., 2022). Consequently, when dealing with size-selective MPs approaches, a weak point is represented by the exact estimate of the sampled volume, which may lead to huge uncertainties in volume estimates (Montoto-Martínez et al., 2022). This issue can be easily overcome by the use of flowmeters (Montoto-Martínez et al., 2022; Pasquier et al., 2022; Shim et al., 2022; Simon-Sánchez et al., 2022), although uncertainties may still arise based on its position in the net frame, often leading to markedly disparate volumes between replicates and consequent difficulty in repeatability (Karlsson et al., 2020; Montoto-Martínez et al., 2022; Pasquier et al., 2022). Furthermore, as indicated by the review conducted by Shim et al. (2022), only 47.5 % of studies on marine MPs pollution used a flow meter to accurately calculate sampled water volume. It is evident therefore that in the case of grab sampling, knowledge of the exact volume of water is a clear advantage of this methodology over Manta (Du et al., 2022; Shim et al., 2022). With regard to representativeness of the sampled water volume, it is evident that if the focus of a study is on larger MPs (sampled by means of net-based approaches) featuring lower concentrations, i.e. less than one particle per cubic meter (Tamminga et al., 2019), a considerable water volume is required for representativeness. Conversely, when targeting the collection of smaller MPs using grab sampling techniques, which tend to be more abundant, a considerably smaller sampled volume is sufficient without compromising the effectiveness of sampling (Lenz and Labrenz, 2018; Prata et al., 2020). In our case, the mean number of particles per cubic meter was 0.24 (SD = 0.29) for Manta sampling, compared to 4050 (SD = 3515) for grab sampling. It is obvious, therefore, that the volume required using the latter technique was significantly smaller compared to the volume needed for Manta sampling. In the present study, the reference volume for the two different sampling methods was determined following the GESAMP (2019) guidelines and the outcomes of (Pasquier et al., 2022) for the Manta case and in accordance with the findings of Prata et al. (2020) for the grab sampling case. The reference volume of net-based methodologies is typically calculated starting from the trawling time of the net, subsequently becoming a direct consequence of the trawling

duration itself. The GESAMP (2019) guidelines suggested trawling the net for approximately 15–30 min at a speed of approximately two knots. According to the review by Pasquier et al. (2022), the towing time used in studies up to 2021 varied between 5 and 90 min, with an average of 20 ± 5 min observed in 54 % of studies. In the present study, a conservative decision was made to use a trawling time of 30 min, considered to be a good trade-off between sampled volume and potential clogging risk. On the other hand, in determining reference volume for the grab sampling methodology, Prata et al. (2020) established the minimum water volume to be sampled in order to obtain reliable results, and thus to correctly quantify small MPs, as being in the range 0.5–1 l. In this study, a conservative safety factor was applied to mitigate variability among samples, and the reference volume for grab sampling was set at 5 l. Nonetheless, given the considerable variability in terms of MPs concentration found herein, future studies should focus on identifying water volumes that are representative for each sampling technique. This volume should preferably vary based on whether sampling occurs in open sea or in locations with high MPs pollution. This approach would facilitate the standardization of various techniques, enhancing comparability of different studies.

Additionally, the likelihood of sample contamination is considerably higher in the case of size selective approaches (Barrows et al., 2017; Prata et al., 2019): generally, the nets and ropes used for towing are made of plastic (typically polyamide and polyester, respectively) that may shed and contaminate the samples (Lenz and Labrenz, 2018; Lusher et al., 2014; Pasquier et al., 2022), while the outside of the Manta is rinsed with unfiltered water, which could contain MPs (Barrows et al., 2017). On the contrary, it is easier to reduce contamination when performing grab sampling: in this study, non-plastic alternatives were used for the collection and storage of grab samples that were triple rinsed and kept face down until use in order to reduce airborne contamination.

From a dimensional standpoint, the results obtained in this study are consistent with those of Barrows et al. (2017) and Di Mauro et al. (2017), with MPs obtained through grab sampling being smaller in size compared to those obtained using net-based sampling methodologies. The outcomes obtained by Kang et al. (2015) are aligned with the previously mentioned studies, with the size of MPs captured with the 50 μm hand-net being smaller than MPs collected with the 330 μm meshed Manta. A study conducted by Lusher et al. (2014) likewise demonstrated that the frequency distribution of MPs was skewed towards the smaller particles.

Accordingly, the optimal sampling method will vary to fit the specific context and objectives of the study. If the goal is to capture *meso*- or relatively large MPs in situ without the aid of microscope analysis, size-selective methods would prove more suitable as they allow significantly larger water volumes to be sampled, thus increasing the potential for capture of larger fragments (Barrows et al., 2017; Green et al., 2018; Shim et al., 2022). On the other hand, the use of volume-selective methods allow the abundance of MPs to be better quantified, in turn enhancing a more precise and reliable assessment of the risks posed to biota and ecosystems (Lindeque et al., 2020; Shim et al., 2022; Simon-Sánchez et al., 2022); it should however be underlined that the potentially small amount of water collected with this approach could lead to distribution anomalies at a local level and high variance among samples (Barrows et al., 2017; Du et al., 2022; Montoto-Martínez et al., 2022; Simon-Sánchez et al., 2022). An increase in sampled volume, number of samples collected or coupling this approach with volume reduction techniques may contribute towards solving this issue (Barrows et al., 2017; Prata et al., 2019; Schmid et al., 2021b; Song et al., 2014). The two analyzed methods can be considered complementary, as they investigate two different facets of the entire spectrum of MP pollution (Tamminga et al., 2019). Table 2 succinctly summarizes the advantages and disadvantages of the two sampling methodologies, bearing in mind that the pros and cons are contingent upon the context and sampling objectives.

The present study did not incorporate the collection of sample replicates due to logistical challenges. Future studies should focus on determining the appropriate number of replicates to be sampled, enabling the investigation of spatial and/or temporal variations in MPs concentrations.

Table 2
Advantages and disadvantages of volume selective and size selective sampling approaches.

Method	Advantages	Disadvantages
Volume selective approach (grab sampling)	- Captures all size fractions - Representative of the abundance of MPs in water - Can be easily conducted by non-specialized personnel. - No uncertainties in the knowledge of exact sampled water volume - When anticontamination protocols are applied, procedural contamination during the sampling phase can be limited - The collected samples can be stored for subsequent analyses of additional pollutants	- Does not allow for collection of a large volume of water, leading to large variability among samples - Large amounts of water can take a long time to be processed
Size selective approach (net-based sampling)	- Allows for collection of a large volume of water over a short period of time - Quick to process for meso or large MPs without microscope - Due to the large volumes of water collected, it can be used for phytoplankton monitoring	- Does not capture all size fractions, its effectiveness is limited by net mesh size - Unrepresentative of the abundance of MPs in water, leading to an underestimation of their concentration - A boat is needed for trawling - Mainly limited to surface water sampling. - Uncertainties in the estimates of sampled water volume, even with flowmeters - Procedural contamination during the sampling phase is difficult to control (from the Manta itself and tow ropes)

5. Conclusions

This study provides a comprehensive examination of two distinct sampling techniques employed in MPs field research, highlighting both strengths and limitations. In studies focusing on the collection and sorting of larger MP particles without the aid of a microscope, the Manta tow method emerges as the preferred choice due to its ability to efficiently sample larger volumes of water, thereby increasing the likelihood of capturing larger plastic fragments. Conversely, our findings corroborate the efficacy of the grab sampling method in capturing a higher density and more diverse range of MP samples, while also minimizing contamination through meticulous adherence to proper laboratory and field protocols. Addressing the critical challenge of accurately estimating quantities of MPs, particularly with regard to global projections, remains paramount within the field. In this regard, grab sampling, either independently or in conjunction with a size selective approach, offers a valuable approach for data synthesis and comparison. This technique demonstrates enhanced accuracy and flexibility in MPs sampling, capable of capturing plastics at both the micro- and nano-scale in a wide array of challenging environmental settings that are not easily accessible using tow nets. A pivotal aspect inherent to both sampling methodologies resides in the delineation of the representative sampling volume, in order to reduce variability among samples and mitigate the risk of overestimation when scaling up the results.

Existing guidelines proffer insights solely into sampling duration for net-based methodologies, with no explicit reference to a minimum sampling volume in the context of grab sampling. Furthermore, the dichotomy between coastal and open-sea environments is not addressed, despite their substantial discrepancy in MP concentrations, necessitating distinctive considerations for representative volumes. A prospective endeavor should be undertaken to assimilate this crucial information within the guidelines.

CRediT authorship contribution statement

Valentina Poli: Writing – original draft, Visualization, Investigation, Formal analysis, Data curation. **Lucio Litti:** Writing – review & editing, Methodology, Formal analysis, Data curation. **Maria Cristina Lavagnolo:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Data curation, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Maria Cristina Lavagnolo reports financial support was provided by National Biodiversity Future Center-Next Generation EU. Maria Cristina Lavagnolo reports equipment, drugs, or supplies and travel were provided by AcegasApsAmga S.p.A - Aliplast. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.174561.

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