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A PLETHORA OF MICROPLASTIC POLLUTION STUDIES: THE NEED FOR A FORENSIC APPROACH

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ABSTRACT

Microplastic pollution has been under the magnifying glass for several years now. Existing data relating to microplastics on surface waters suggest that they are globally widespread, but there are several gaps of knowledge in relation to understand how many there are in different locations, what is their composition, where do they come from and where they are going. What we need is a global collaborative effort to collect this information on a large scale. To date, standardized methodologies for the sampling and analysis of microplastics are still lacking, which therefore hinders the comparison of the reported data. This review summarizes the currently used methodologies for sampling and identifying microplastics in surface water, with the intention of contributing to the establishment of standardized and harmonized protocols. In addition, we focus our attention on the great potential that environmental forensic sciences have to face the delicate and insidious challenge of microplastic pollution, urging future research to go in this direction, in order to develop a rigorous and robust forensic method for microplastics study.

1. INTRODUCTION

Plastic pollution is undoubtedly one of the greatest global concerns of the 21st century. Nowadays, it is recognized as a complex, multidimensional and multi-sectorial problem with economic, environmental, public health, food safety and even cultural implications (Llorca et al., 2020). Complying with the ever-increasing demand, world plastic production has grown dramatically from year to year, reaching as much as 370 million tons in 2019 (Plastics Europe, 2019). This massive production of plastic materials seems destined not to stop, in fact this value is estimated to quadruple within the next 30 years, accounting for 20% of the global oil consumption and 15% of the annual carbon emissions by 2050 (MacArthur & Waughray, 2016). As if that weren't enough, such predictions will likely be aggravated by the excessive use and consumption of single-use plastics (including personal protective equipment such as masks and gloves) due to the outbreak of COVID-19 pandemic. In a business as-usual scenario, this projected increase in plastic production will be accompanied by the resulting mismanagement of waste, which often ends up in the environment through a variety of pathways. Currently, it is estimated that at least 8 million tons of plastic end up in the oceans every year, so without significant action and continuing to business as usual, by 2050 we will have more plastic than fish (by weight) in the world oceans (Jambeck et al., 2015). Plastics can remain in the ocean for hundreds of years in their original form and even longer in small particles (MacArthur & Waughray, 2016). Although many plastics are remarkably persistent, they are not immune to degradation (by photochemical reaction or mechanical actions), which can lead to the formation of plastic particles smaller than 5 mm generally known as microplastics (MPs). These small plastic particles may either result from the breakdown of larger objects, or they can intentionally produced in small sizes. At present, almost all of the world's oceans and seas are contaminated with MPs. Substantial quantities of MPs have been found in the global marine ecosystem (Shahul Hamid et al., 2018; Suaria, Achtypi, et al., 2020), from the tropics to the poles, including Arctic and Antarctic sea (Ross et al., 2021; Suaria, Perold, et al., 2020; Waller et al., 2017). The recognition of the magnitude of the problem has given rise to a series of initiatives by different institutions and the scientific community, which has shown an ever increasing interest in this problem and consequently, the number of published studies on surface MPs in the marine environment has significantly increased, but the result is a wide range of sampling, sample processing, sample analysis, data analysis, and reporting methodologies, which complicate cross-study comparability and larger scale synthesis. We currently have the awareness that MPs are everywhere, but knowledge about the polymeric composition, the spatial and temporal distribution of these

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floating particles is still largely unknown. Environmental forensic approaches have the potential to tackle this ubiquitous challenge. Environmental forensics has emerged as an important growing area, which concerns the investigation of a diverse range of pollutants that have been, accidentally or deliberately, released into the environment (Philp, 2014). Environmental forensic analysis involves the application of defensible scientific methods to address questions related to understanding the extent, duration, and responsibility for environmental contamination sites in a regulatory and/or legal context. It generally involves the reconstruction of past environmental events, determining the timing, types, amounts and sources of chemical releases into the environment. Over the past decades, pollutants such as oil and heavy metals, have been the focus of investigation, however recently, emerging pollutants such as plastic waste have become of interest to environmental forensic scientists (S. K & Varghese, 2020). Although MPs are not a standard contaminant within the remit of an environmental forensic expert, the identification and characterization of pollutants and the consequent achievement of the pollution source is an integral part of any environmental forensic analysis. An effective forensic analysis involves the systematic and scientific evaluation of data obtained from field measurements or laboratory analysis, and historical information of the contaminated sites, in order to develop defensible scientific and legal conclusions regarding the release histories, age and source of a contaminant into the environment. In some cases, therefore, the forensic puzzle is resolved through such file review, while in others this guides the forensic strategy for testing and helps establish the most appropriate forensic tools. In the MPs case, for example, due to to the absence of standardized methodologies for the sampling and analysis of MPs, the comparison of the reported data may not provide any forensic answers. In order to manage environmental data and forensically interpret the results, specific approaches, commonly referred to as fingerprinting methods, should be carried out. "Fingerprinting" is a broad term that includes a variety of methods and techniques aimed to establish correlation and patterns of contamination that may be related to specific sources and/or time frames of contaminant release (I. Petrisor, 2005). Several main fingerprinting techniques are well established and their applicability proved (e.g., chemical fingerprinting, isotopic analysis, statistical methods). Methods and techniques for the study of MPs are continuously evolving with increased research in this field (Miller et al., 2021). Nevertheless, to date, the questions concerned with the level of MPs, the transport, eventual fate, and source identification are still to a large extent unanswered, mainly due to the fact that the forensic investigation of MPs is still in the infancy stages and the standards are not evolved enough.

The present review evaluates the current knowledge on the occurrence and abundance of floating MPs in the marine environment, providing a snapshot of the global MP contamination of surface waters and underlining the methodological differences of the various studies present in the literature. In addition, we focus our attention on the great potential that environmental forensic sciences have to face the delicate and insidious challenge of MP pollution, urging future research to go in this direction, in order to develop a rigorous and robust forensic method for MPs study.

2. ANALYSIS OF MICROPLASTICS IN SURFACE WATER

During the last years, several studies have evaluated the abundance, distribution and composition of floating MPs in marine environment. Table 1 summarizes some of the most recent works reporting MP concentrations found together with investigated areas, year of sampling, sam-

TABLE 1: Microplastics concentrations and methods used in some recent and representative works.

Study Area	Year of sampling	Sampling nets	Net mesh (µm)	Mean density	Analysis tool	Reference
Whole Mediterranean	2013	Neuston net	200	0.243 items/m²	Stereomicroscope	(Cózar et al., 2015)
Western Mediterranean	2013	Neuston net	200	0.40 ± 0.74 items/m²	Stereomicroscope/ FTIR	(Suaria et al., 2016)
Central-Western Mediterranean	2011-13	Manta trawl	333	0.147 items/m²	Optical microscope	(Ruiz-Orejón et al., 2016)
Central Mediterranen	2020	Manta trawl	333	0.13 ± 0.194 items/m²	Stereomicroscope/ FTIR	(Marrone, La Russa, et al., 2021)
Eastern Mediterranean	2018	Manta trawl	52	4.3 ± 2.2 items/m³	Stereomicroscope/ Raman	(Kazour et al., 2019)
Eastern Mediterranean	2013-15	Manta trawl	333	7.68 ± 2.38 items/m³	Stereomicroscope/ FTIR	(van der Hal et al., 2017)
Mid-west Pacific Ocean	2017	Manta trawl	333	34,04 ± 25,1 items/km²	Stereomicroscope/ µRaman	(S. Wang et al., 2020)
Eastern Indian Ocean	2019	Manta trawl	330	0.34 ± 0.80 items/m²	Stereomicroscope/ µFTIR	(C. Li et al., 2021)
Northwest Pacific Ocean	2017	Manta trawl	330	10^4 items/km²	Stereomicroscope/ µRaman/SEM	(Pan et al., 2019)
Southern Ocean	2016-17	Neuston net	200	188 ± 589 items/km²	µFTIR	(Suaria, Perold, et al., 2020)
Arctic Ocean	2016	/	63	40.5 ± 4.4 items/m³.	µFTIR	(Ross et al., 2021)

pling net and instrumental methods used for detection of MPs. Clearly the comparison between the different studies is very difficult due to the absence of a standardized sampling protocol and the lack of homogeneity both in the identification methods and in the expression of the results. In the following sections we summarize the sampling and analysis methods used so far aiming to promote homogeneous monitoring programs for MPs in aquatic environments.

2.1 Sample collection and preparation

The selection of sample collection methods is critical and will dramatically influence the study results (Miller et al., 2021). Most of the studies carried out so far used different versions of the Neuston and Manta trawl nets to sample sea surface MPs. Although either of these two trawling devices are recommended, a limiting factor is the net mesh sizes that can vary widely, strongly influencing the size spectrum and the abundance of collected particles (Gago et al., 2019; Lindeque et al., 2020). For instance, MP concentration using a 100 µm net is 10-fold greater than a 500 µm net. A nylon net (100 µm) revealed concentrations almost a hundred times higher than a manta net (333 µm) (Vermaire et al., 2017). Another study showed that an 80 µm mesh could retain up to 250 times higher concentration of plastic fibers than that of a 330 µm mesh (Dris et al., 2018). Despite these evidences, it should be noted that most of the currently used sampling techniques are only applicable to collection of MPs with certain size ranges. The Marine Strategy Framework Directive (MSFD) guidelines recommends a mesh size of 333 µm, and as can be seen from the table, the most usual mesh size ranging from 300 to 390 µm, which seems a reasonable cut-off to yield acceptable and meaningful samples under open net sampling conditions. However, the above examples suggest underestimation of smaller plastics based on traditional sampling, thus it remains of fundamental importance to adequately cover the lower MP size ranges in surveys that estimate current pollution levels. The quantity of MPs sampled has been expressed in different units, divided by the towed area (e.g., items/m^2) or volume of sampled water (items/m^3) or weight by sampled area (g dry weight/km^2). It is clear that having three different units of measurement to express the same quantity does nothing but complicate or sometimes make it impossible to compare the different studies. An interesting study showed that of the two most commonly used methods for calculating MP concentration (flowmeter and ship's log), ship's log provided consistently smaller abundances, with the exception of one sample, calling for a standardisation in the techniques and measurements used to quantify floating microplastics (Rivers et al., 2019). In addition, it has been proven that the concentration of MPs in the sea can be strongly affected by the presence of wind, therefore recent studies, taking into account this factor, corrected MPs concentration using a widely used theoretical model (Kukulka et al., 2012). Once collected, separation and extraction of MPs from samples are performed by sieving, density floatation and filtration, whose effectiveness depends on the particle size and MP typology (Martellini et al., 2018). Clearly it is important to adopt the most appropriate strategy based on the starting sample, also because the subtraction of even a single particular type of MP to the analysis can be costly, especially in investigations with forensic purpose (Kumar et al., 2021).

2.2 Microplastics identification and characterization

After field collection and laboratory preparation of the samples, MPs must be accurately identified from the remaining matrix and suitably characterized (W. Wang & Wang, 2018). Methods for characterizing MPs are rapidly evolving. Many older publications only visually identified likely plastic particles, the so called microparticles, while today the standard has shifted to a more accurate MPs identification (Miller et al., 2021). Particle analysis, or characterization, involves two steps. Morphological or physical categorization which take into account sizes, shape and color of potential MPs, followed by chemical characterization for definitive confirmation. This framework is based on the idea of characterizing samples initially using basic techniques and then progressively using more complex techniques in order to obtain a comprehensive picture of the composition of the basic material and its possible relationship to the origin source. In addition to identifying the basic material, tracing back the exact source requires rigorous efforts and the use of advanced technologies. Therefore a standalone method may not provide useful insights, and it is more useful to adopt several independent methods which would lead to identical conclusions (Cattle et al., 2010; Haddad, 2004; I. G. Petrisor, 2014). Analysis of MPs in environmental samples also requires an experienced laboratory due to the prevalence of background contamination sources.

2.2.1 Morphological characterization

In order to describe the morphological and physical properties that characterize MPs, the most common method involves the visual inspection of suspected MPs on optical microscope (typically a stereomicroscope). Magnified images using microscopy provide detailed surface texture and structural information of objects, which is essential for identifying ambiguous, plastic-like particles (Shim et al., 2017). To improve the accuracy of identification results, a series of previously established criteria must be taken into consideration during MPs visual inspection: absence of cellular or organic structures; fibers should have consistent thickness and color along the entire length and particles should be uniformly colored (Hidalgo-Ruz et al., 2012). Transparent and white particles should be further confirmed under a high-magnification microscope or a fluorescence microscope after Nile Red staining (Hidalgo-Ruz et al., 2012). Although high magnification optical microscopy, with optimized illumination methods, allows high resolution visual inspection, previous studies have shown that false identification of plastic-like particles using microscopy was often over 20%, and over 70% for transparent particles (Hidalgo-Ruz et al., 2012); highlighting the limits of visual characterization, which is subjective and prone to errors. In addition, this procedure, although it is the most widely used, is strongly influenced by several factors such as the sensitivity of the examiner, the particle shape and

size, the sample matrix and the microscope used, which could introduce potential bias affecting the final results (W. Wang & Wang, 2018). Moreover, visual inspection becomes more challenging as the size of the particles under consideration decreases, greatly increasing the possibility of misidentification and underestimation of these smaller elements, which are the most dangerous, since it has been suggested that smaller the particle size is, the higher are the chance of ingestion and retention rate by organisms (Gray & Weinstein, 2017; Kögel et al., 2020). For all these reasons, performing spectroscopic and chemical analysis to confirm the polymeric nature of the suspected MPs and to allow the specific identification of the different plastic types, avoiding MPs overestimation due to the presence of non-plastic items, isessential, especially for the smaller items (de Haan et al., 2019). Various techniques are feasible for MPs identification, such as scanning electron microscope (SEM) which allow to obtain high resolution images of a sample by firing a high-intensity electron beam at the sample surface and scanning it in a raster scan pattern (Crawford & Quinn, 2017). Surface details (<0.5 nm resolution) of the sample are imaged by the electrons at very high magnifications, thus differentiating the MPs from other organic or inorganic residues (Crawford & Quinn, 2017). Moreover, the combined use of SEM and energy dispersive X-ray spectroscopy (SEM-EDS) is able to provide detailed information about the MPs elemental composition and the inorganic additives they contain (Crawford & Quinn, 2017; Fries et al., 2013). SEM-EDS helps to further differentiate MPs from natural materials, but this technique requires considerable time and effort for sample preparation and therefore is hardly applicable for routine analysis of large numbers of samples.

2.2.2 Chemical characterization

Chemical characterizationis a final step to confirm the polymeric nature of MPs and distinguish them from other natural materials, when visual and microscopic observation is not enough to confirm particle nature. Additionally, this step allows for specific identification of different plastic types, which can be helpful in better understanding their parent materials, possible sources and input pathways, as well as the toxic chemicals associated with plastics with further instrumental analysis. The most common method in the chemical characterization of MP particles is spectroscopy, both Fourier-Transform Infrared (FTIR) and Raman. FTIR spectroscopy provides information on the specific chemical bonds and functional groups of each plastic polymer. The different chemical compositions of each materials produce unique infrared spectra, making it possible to identify an unknown substance by comparing its spectrum with the spectra of known materials. Raman spectroscopy, on the other hand, involves irradiating a suspected sample with a monochromatic laser beam, which results in a varying wave length of a backscattered light due to absorption, reflection, or scattering of the specific molecular structure and atomic composition (Crawford & Quinn, 2017). This so-called Raman shift can produce a unique spectrum for each polymer (Huppertsberg & Knepper, 2018). Thanks to their extreme efficacy and high degree of reliability, both of these are the most widely employed techniques in the chemical characterization of MPs from different environmental samples (Araujo et al., 2018; Shim et al., 2017; Silva et al., 2018). FTIR and Raman spectroscopy are both non-destructive methods that allow high throughput screening, requiring low sample quantities and respecting the environment (Araujo et al., 2018). In addition to the advantages, these two methods also share some disadvantages, in fact both require expensive instrumentation and are time-consuming when a large number of MPs need to be analyzed. FTIR, unlike Raman spectroscopy, can not only accurately identify the polymeric composition of MPs, but also provide further information about their physiochemical weathering by analyzing their oxidation intensity (Corcoran et al., 2009). Raman spectroscopy, on the other hand, is advantageous in terms of higher spatial resolution, wider spectral range, tighter spectral bonds and lower sensitivity to water interference than FTIR techniques (Käppler et al., 2016). At the same time, the main drawback of Raman spectroscopy is that it can be easily interfered by the presence of pigments, additives, or chemicals associated with MPs, which may adversely affect the accuracy of identification (Huppertsberg & Knepper, 2018). The application power of both of these methods is significantly increased in combination with microscopy. Micro-FTIR (μFTIR) and micro-Raman (μRaman) in fact allow the detection and identification of MPs with dimensions in the range between 10 and 20 μm (Lenz et al., 2015). All this is possible through the use of a single tool, by switching between the object lens and beam, whether it be IR or laser, thus allowing the chemical and physical characterization of the analyzed particles simultaneously (Elert et al., 2017).

2.3 Quality assurance and quality control (QA/QC)

It is standard practice in forensic investigations to apply strict anti-contamination protocols (Kumar et al., 2021). Forensic examinations are under scrutiny by the criminal justice system, therefore rigor and validity of approach are critical to their design (Woodall et al., 2015). The same principles should be applied when conducting monitoring programs on MPs, during which it is essential and crucial to adopt strict quality assurance and quality control (QA/QC) measures throughout the methodological process, in order to improve data quality (W. Wang & Wang, 2018). Background contamination can in fact cause a significant overestimation, negatively affecting the accurate assessment of MP abundance in the studied area. Despite the high potential for sample contamination, many studies still only report crude or limited procedures far less comprehensive than those that would be used in criminal investigations (Kumar et al., 2021). A recent study analyzed the problem of contamination in the analysis of MPs, proposing a protocol to be adopted during all analytical phases, from field sampling to the laboratory analysis (Prata et al., 2021). First of all, to check for background contamination, during sampling and laboratory handling process, a series of procedural blank tests should be conducted and processed in the same way as the real samples. During laboratory analyzes, as general control rules, some preventive measures

should be adopted, such as wearing 100% cotton laboratory coats (avoiding synthetic clothing) and nitrile gloves. In addition, all work surfaces should be thoroughly cleaned and all laboratory equipment (such as sieves, tweezers and glassware) should be rinsed with bi-distilled water. Besides, a special mention goes to the potential airborne contamination by fibers. The presence of high background levels of fibers in a working laboratory was demonstrated by Nuelle and coworkers (Nuelle et al., 2014) and confirmed by Woodall and coworkers (Woodall et al., 2015), which in their study developed a protocol based on the recommendations and procedures made for the forensic investigation of fibers, therefore it follows that are likely to be the most effective as they must stand up to the scrutiny of the courts. However, a very recent study has verified that, despite the application of the most rigid and severe anti-contamination protocols of a forensic nature, fiber contamination can be reduced (in this specific case by 36.9%), but cannot be completely avoided (Kumar et al., 2021). To overcome these problems, the authors therefore suggest protocols which, in addition to minimizing contamination, also allow monitoring it, for example outfitting the whole team in the same thin garments in unusual colors, ideally also with unusual fiber morphology. The authors therefore state that using forensic analysis techniques, which aim to completely profile a particle, including its morphological, optical and chemical characteristics, enable to obtain information that allows much more confident conclusions to be drawn as to whether it comes from the environment or procedural contamination.

2.4 Source identification

One of the biggest challenges in environmental forensic investigations is determining the source of the contaminant. Identifying the sources of pollutants and the extent of their contribution is one of the first steps in their management (Kumar et al., 2021). Before continuing, however, it is necessary to to keep in mind a concept: it is very important to make very clear that source refers to the point of release, not the manufacture of a particular product. This is very important since there were papers published many years ago that tried to use certain characteristics of products made by certain manufacturers and use those properties to determine whether a particular released product was produced by that particular company (Philp, 2014). Having made this important premise, we can continue by saying that establishing provenance is not easy when the materials are microscopic in nature. Fortunately, as we have seen previously, MPs have tell-tale chemical fingerprints of their origin that can be recognized using modern analytical chemistry techniques. The first step towards the identification of the origin of microplastics starts with their complete and detailed characterization. As in any forensic context, the complete labeling of the samples provides invaluable information. For instance, fibers may result from washings of textile products, whereas regular shapes like spheres, cylinders, etc., are indicative of specific sources (i.e. personal hygiene products) and the detailed labeling cannot be missed when the purpose of analysis is source identification (Farooq et al., 2022; Kumar et al., 2021). If the MP is secondarily sourced from a larger plastic product, the debris will be irregularly fragmented. Observing MPs degradation patterns under an optical microscope can provide a basic idea on the resident time of MPs in the environment (Kumar & Varghese, 2021). Such observation will help in answering the questions if the MPs are recently formed or are quite old. Sharp edges of a fragment indicate freshly formed MP when compared to a MP with blunt edges. Also, crack formation, loss of material from the surface, etc. are indications of longer residence time in the environment (Kumar et al., 2021). In aquatic environments MPs age can be a determining factor also in relation to their origin, for example, a smooth MP suggests a local source, while a worn-out MP with a biofilm may suggest a distant source (Farooq et al., 2022). Moreover, studies have also shown that there is a significant effect for the shape on the MPs transport in the environment (Harrison et al., 2018; Jahnke et al., 2017). Unfortunately, many MPs studies limit their observations to colour, size and classification of the sample set as a whole rather than fully characterizing each microplastic as seen in forensic examinations (C. M. B. Gwinnett et al., 2021). In this regard, much can be learnt from forensic fibre analysis, where polymer fibres are examined for their colour, width, cross-sectional shape, presence of inclusions and optical properties such as its birefringence and sign of elongation (Robertson et al., 2017). This is the direction taken by Gwinnet and coworkers, who in a recent study have proposed a new workflow for the recovery and analysis of MPs, particularly fibres, which allows greater differentiation between samples and aids in source identification (C. M. B. Gwinnett et al., 2021). Obviously, the increased information of spectral techniques allows polymers and sometimes additives to be clearly identified, which can be utilized for chemical identification and MPs characterization (Primpke et al., 2020). Moreover, in recent years the forensic analysis of polymers has been improved by the application of advanced statistical analysis techniques (Cowger et al., 2020; Fang et al., 2022). In fact, several studies have processed the large data sets produced by the various spectral imaging techniques used, in analysis and identification groups, thanks to principal component analysis (PCA) (Fang et al., 2022; Y. Li et al., 2020; Lorenz et al., 2019). PCA is a universal method of static data analysis which projects high-dimensional data onto a low-dimensional space, commonly two dimensions. The basic idea of PCA is to convert a set of correlated variables into a new set of uncorrelated variables called principal components (PCs), which are linear combinations of the raw variables that account for a large proportion of the total variance of raw data (Jollife & Cadima, 2016). Wanting to simplify, PCA allows to manage large datasets by reducing their dimensionality, increasing the interpretability but at the same time minimizing the loss of information (Jollife & Cadima, 2016). Over the past few years, PCA has been increasingly used to identify and differentiate particles, materials or cells in various fields of research, such as chemistry and biology, but also environmental and forensic science (Fang et al., 2022). Therefore, PCA is considered suitable for the analysis of microplastics by de-

coding their spectrum matrix. It allows differentiation between the spectra of synthetic and natural origin, resulting in the enhanced visual accessibility by creating a two-dimensional image of the MP (Farooq et al., 2022). Data of similar spectra are grouped and labelled accordingly by comparing it to the reference spectra. A lot of studies state the effectiveness of using PCA for the identification of polymers, nevertheless further research is still needed to develop the reference library, in order to ensure the automatic decoding of the spectrum matrix for mapping and imaging. Furthermore, advances in analytical and data exploration techniques in recent years have helped researchers to evaluate larger patterns of contamination or "footprints" in the environment. Statistical methods, including PCA, provide additional analytical tools to obtain fingerprints, as they aim to correlate contamination with sources (I. Petrisor, 2005). Increasing or decreasing contamination trends are established over time and space by widely available statistical techniques. Therefore, more research is required to collect forensic information regarding the frequency and distribution patterns for different shapes, sizes, colours and types of polymers and the possible entering pathways (Browne et al., 2011). Tracing the MPs source is in fact extremely challenging, due to the different use of a single type of polymer, multiple manufacturing techniques, use of different additives in the same type of polymer by different industries, etc. (Kumar & Varghese, 2021). Only the application of robust environmental forensic approaches and protocols and a correct interpretation of the information obtained in the previous phases, starting from those collected during the sampling (i.e. GPS coordinates, along with local details such as geographical features, proximity to harbor or wastewater treatment plants, influence of river, type of beach activities, etc.) up to the analyzed data, can allow to address this challenge (C. Gwinnett et al., 2021; Kumar et al., 2021). Kumar and Varghese (S. K & Varghese, 2020), applying a framework developed for forensic investigation, were able to reach some useful conclusions regarding the source of each type of MP they observed; in some cases it was possible to identify the exact source, whereas in other cases only the pathway through which the MP reached the marine environment could be identified. Clearly, the further away the source of pollution is, the more difficult it is to trace it. This, for example, is the case of the Antarctic continent, the southernmost part of the planet, which despite its geographical isolation, is not protected from the negative impact of human activities (Marrone, La Russa, Brunelli, et al., 2021). Several contaminants, such as heavy metals and MPs, have been detected in this area, even in significant amounts, despite the pollution sources being very distant (Cincinelli et al., 2017; Marrone, La Russa, Brunelli, et al., 2021; Suaria, Perold, et al., 2020). Particularly interesting is a study recently published by Leistenschneider and collaborators, which using a forensic approach were able to discriminate between environmental and vessel-induced MPs, and thus revealing that 45.5% of all MPs they have sampled in the Weddell Sea (Antarctica), it was actually due to ship-induced contamination (Leistenschneider et al., 2021).

3. CONCLUSIONS AND FUTURE PERSPECTIVES

Plastic pollution has become one of the most pressing environmental issues. Existing data relating to MPs on surface waters suggest that they are globally widespread, however, there are still some bias that do not allow for a reliable and comparable quantitative data analysis between different studies. Several analytical steps in the study of MPs have become critical bottlenecks that prevent a global collaborative effort for large-scale data analysis. Therefore, the establishment of standardized and harmonized protocols for sampling, identification and expression of the results, and so of all operating procedures involved in the cycle of assessing environmental MPs from field sampling to laboratory analysis, it is essential to improve the current knowledge on MPs phenomenon. Furthermore, to try to answer still unresolved questions concerned with the real extent, source and fate of MPs, the forensic approach applied to environmental studies represents an added value for the development of more complete strategies and robust methods, thus allowing to provide a real and more complete picture of MP pollution in the marine environment.

REFERENCES

Araujo, C. F., Nolasco, M. M., Ribeiro, A. M. P., & Ribeiro-Claro, P. J. A. (2018). Identification of microplastics using Raman spectroscopy: Latest developments and future prospects. Water Research, 142, 426–440. https://doi.org/10.1016/j.watres.2018.05.060

Browne, M. A., Crump, P., Niven, S. J., Teuten, E., Tonkin, A., Galloway, T., & Thompson, R. (2011). Accumulation of microplastic on shorelines woldwide: Sources and sinks. Environmental Science and Technology, 45(21), 9175–9179. https://doi.org/10.1021/es201811s

Cattle, J. A., Way, C. P., Fuller, S., Low, G., Vaughan, G. T., Cattle, J. A., Way, C. P., Fuller, S., Low, G., & Vaughan, G. T. (2010). Identification and Matching of Environmental Samples for Regulatory Purposes : A Systematic Approach Identification and Matching of Environmental Samples for Regulatory Purposes : A Systematic Approach. Environmental Forensics, July 2015, 185–194.

Cincinelli, A., Scopetani, C., Chelazzi, D., Lombardini, E., Martellini, T., Katsoyiannis, A., Fossi, M. C., & Corsolini, S. (2017). Microplastic in the surface waters of the Ross Sea (Antarctica): Occurrence, distribution and characterization by FTIR. Chemosphere, 175, 391–400. https://doi.org/10.1016/j.chemosphere.2017.02.024

Corcoran, P. L., Biesinger, M. C., & Grifi, M. (2009). Plastics and beaches: A degrading relationship. Marine Pollution Bulletin, 58(1), 80–84. https://doi.org/10.1016/j.marpolbul.2008.08.022

Cowger, W., Gray, A., Christiansen, S. H., DeFrond, H., Deshpande, A. D., Hemabessiere, L., Lee, E., Mill, L., Munno, K., Ossmann, B. E., Pittroff, M., Rochman, C., Sarau, G., Tarby, S., & Primpke, S. (2020). Critical Review of Processing and Classification Techniques for Images and Spectra in Microplastic Research. Applied Spectroscopy, 74(9), 989–1010. https://doi.org/10.1177/0003702820929064

Cózar, A., Sanz-Martín, M., Martí, E., González-Gordillo, J. I., Ubeda, B., Á.gálvez, J., Irigoien, X., & Duarte, C. M. (2015). Plastic accumulation in the mediterranean sea. PLoS ONE, 10(4). https://doi.org/10.1371/journal.pone.0121762

Crawford, C. B., & Quinn, B. (2017). Microplastic identification techniques. Microplastic Pollutants, 219–267. https://doi.org/10.1016/b978-0-12-809406-8.00010-4

de Haan, W. P., Sanchez-Vidal, A., & Canals, M. (2019). Floating microplastics and aggregate formation in the Western Mediterranean Sea. Marine Pollution Bulletin, 140, 523–535. https://doi.org/10.1016/j.marpolbul.2019.01.053

Dris, R., Gasperi, J., Rocher, V., & Tassin, B. (2018). Synthetic and non-synthetic anthropogenic fibers in a river under the impact of Paris Megacity: Sampling methodological aspects and flux estimations. Science of the Total Environment, 618, 157–164. https://doi.org/10.1016/j.scitotenv.2017.11.009

Elert, A. M., Becker, R., Duemichen, E., Eisentraut, P., Falkenhagen, J., Sturm, H., & Braun, U. (2017). Comparison of different methods for MP detection: What can we learn from them, and why asking the right question before measurements matters? Environmental Pollution, 231, 1256–1264. https://doi.org/10.1016/j.envpol.2017.08.074

Fang, C., Luo, Y., Zhang, X., Zhang, H., Nolan, A., & Naidu, R. (2022). Identification and visualisation of microplastics via PCA to decode Raman spectrum matrix towards imaging. Chemosphere, 286. https://doi.org/10.1016/j.chemosphere.2021.131736

Farooq, S., Qadir, A., & Jameel, N. (2022). Evaluation of Different Metrics to Study Microplastics as an Environmental Forensic Tool. 89–107. https://doi.org/10.1007/978-3-030-89220-3_5

Fries, E., Dekiff, J. H., Willmeyer, J., Nuelle, M. T., Ebert, M., & Remy, D. (2013). Identification of polymer types and additives in marine microplastic particles using pyrolysis-GC/MS and scanning electron microscopy. Environmental Sciences: Processes and Impacts, 15(10), 1949–1956. https://doi.org/10.1039/c3em00214d

Gago, J., Filgueiras, A., Pedrotti, M. L., Caetano, M., & Firas, J. (2019). Standardised protocol for monitoring microplastics in seawater. JPI-Oceans BASEMAN project. January, 96. https://doi.org/10.13140/RG.2.2.14181.45282

Gray, A. D., & Weinstein, J. E. (2017). Size- and shape-dependent effects of microplastic particles on adult daggerblade grass shrimp (Palaemonetes pugio). Environmental Toxicology and Chemistry, 36(11), 3074–3080. https://doi.org/10.1002/etc.3881

Gwinnett, C., Harrison, E., Osborne, A., Pivato, A., & Varghese, G. (2021). Sampling microplastics for environmental forensic applications. Detritus, 14, I–III. https://doi.org/10.31025/2611-4135/2021.14080

Gwinnett, C. M. B., Osborne, A. O., & Jackson, A. R. W. (2021). The application of tape lifting for microplastic pollution monitoring. Environmental Advances, 5. https://doi.org/10.1016/j.envadv.2021.100066

Haddad, R. I. (2004). Invited editorial: What is environmental forensics? Environmental Forensics, 5(1), 3. https://doi.org/10.1080/15275920490424006

Harrison, J. P., Hoellein, T. J., Sapp, M., Tagg, A. S., Ju-Nam, Y., & Ojeda, J. J. (2018). Microplastic-associated biofilms: A comparison of freshwater and marine environments. Handbook of Environmental Chemistry, 58, 181–201. https://doi.org/10.1007/978-3-319-61615-5_9

Hidalgo-Ruz, V., Gutow, L., Thompson, R. C., & Thiel, M. (2012). Microplastics in the marine environment: A review of the methods used for identification and quantification. Environmental Science and Technology, 46(6), 3060–3075. https://doi.org/10.1021/es2031505

Huppertsberg, S., & Knepper, T. P. (2018). Instrumental analysis of microplastics—benefits and challenges. Analytical and Bioanalytical Chemistry, 410(25), 6343–6352. https://doi.org/10.1007/s00216-018-1210-8

Jahnke, A., Arp, H. P. H., Escher, B. I., Gewert, B., Gorokhova, E., Kühnel, D., Ogonowski, M., Potthoff, A., Rummel, C., Schmitt-Jansen, M., Toorman, E., & MacLeod, M. (2017). Reducing Uncertainty and Confronting Ignorance about the Possible Impacts of Weathering Plastic in the Marine Environment. Environmental Science and Technology Letters, 4(3), 85–90. https://doi.org/10.1021/acs.estlett.7b00008

Jambeck, J. R., Geyer, R., Wilcox, C., Siegler, T. R., Perryman, M., Andrady, A., Narayan, R., & Law, K. L. (2015). Plastic waste inputs from land into the ocean. Science, 347(6223), 768–771. https://doi.org/10.1126/science.1260352

Jollife, I. T., & Cadima, J. (2016). Principal component analysis: A review and recent developments. Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 374(2065). https://doi.org/10.1098/rsta.2015.0202

Käppler, A., Fischer, D., Oberbeckmann, S., Schernewski, G., Labrenz, M., Eichhorn, K. J., & Voit, B. (2016). Analysis of environmental microplastics by vibrational microspectroscopy: FTIR, Raman or both? Analytical and Bioanalytical Chemistry, 408(29), 8377–8391. https://doi.org/10.1007/s00216-016-9956-3

Kazour, M., Jemaa, S., Issa, C., Khalaf, G., & Amara, R. (2019). Microplastics pollution along the Lebanese coast (Eastern Mediterranean Basin): Occurrence in surface water, sediments and biota samples. Science of the Total Environment, 696. https://doi.org/10.1016/j.scitotenv.2019.133933

Kögel, T., Bjorøy, Ø., Toto, B., Bienfait, A. M., & Sanden, M. (2020). Micro- and nanoplastic toxicity on aquatic life: Determining factors. Science of the Total Environment, 709. https://doi.org/10.1016/j.scitotenv.2019.136050

Kukulka, T., Proskurowski, G., Morét-Ferguson, S., Meyer, D. W., & Law, K. L. (2012). The effect of wind mixing on the vertical distribution of buoyant plastic debris. Geophysical Research Letters, 39(7). https://doi.org/10.1029/2012GL051116

Kumar, A. S., Beggio, G., Pivato, A., Gwinnett, C., & Varghese, G. (2021). Analysis of microplastics for environmental forensic applications. Detritus, 16, I–IV. https://doi.org/10.31025/2611-4135/2021.15125

Kumar, A. S., & Varghese, G. K. (2021). Microplastic pollution of Calicut beach - Contributing factors and possible impacts. Marine Pollution Bulletin, 169. https://doi.org/10.1016/j.marpolbul.2021.112492

Leistenschneider, C., Burkhardt-Holm, P., Mani, T., Primpke, S., Taubner, H., & Gerdts, G. (2021). Microplastics in the Weddell Sea (Antarctica): A Forensic Approach for Discrimination between Environmental and Vessel-Induced Microplastics. Environmental Science and Technology, 55(23), 15900–15911. https://doi.org/10.1021/acs.est.1c05207

Lenz, R., Enders, K., Stedmon, C. A., MacKenzie, D. M. A., & Nielsen, T. G. (2015). A critical assessment of visual identification of marine microplastic using Raman spectroscopy for analysis improvement. Marine Pollution Bulletin, 100(1), 82–91. https://doi.org/10.1016/j.marpolbul.2015.09.026

Li, C., Wang, X., Liu, K., Zhu, L., Wei, N., Zong, C., & Li, D. (2021). Pelagic microplastics in surface water of the Eastern Indian Ocean during monsoon transition period: Abundance, distribution, and characteristics. Science of the Total Environment, 755. https://doi.org/10.1016/j.scitotenv.2020.142629

Li, Y., Lu, Z., Zheng, H., Wang, J., & Chen, C. (2020). Microplastics in surface water and sediments of Chongming Island in the Yangtze Estuary, China. Environmental Sciences Europe, 32(1). https://doi.org/10.1186/s12302-020-0297-7

Llorca, M., Álvarez-Muñoz, D., Ábalos, M., Rodríguez-Mozaz, S., Santos, L. H. M. L. M., León, V. M., Campillo, J. A., Martínez-Gómez, C., Abad, E., & Farré, M. (2020). Microplastics in Mediterranean coastal area: toxicity and impact for the environment and human health. Trends in Environmental Analytical Chemistry, 27. https://doi.org/10.1016/j.teac.2020.e00090

Lorenz, C., Roscher, L., Meyer, M. S., Hildebrandt, L., Prume, J., Löder, M. G. J., Primpke, S., & Gerdts, G. (2019). Spatial distribution of microplastics in sediments and surface waters of the southern North Sea. Environmental Pollution, 252, 1719–1729. https://doi.org/10.1016/j.envpol.2019.06.093

MacArthur Dame Ellen, Waughray Dominic, M. R. S. (2016). The New Plastics Economy: Rethinking the future of plastics. Ellen MacArthur Foundation, January, 120.

Marrone, A., La Russa, D., Brunelli, E., Santovito, G., La Russa, M. F., Barca, D., & Pellegrino, D. (2021). Antarctic Fish as a Global Pollution Sensor: Metals Biomonitoring in a Twelve-Year Period. Frontiers in Molecular Biosciences, 8. https://doi.org/10.3389/fmolb.2021.794946

Marrone, A., La Russa, M. F., Randazzo, L., La Russa, D., Cellini, E., & Pellegrino, D. (2021). Microplastics in the center of mediterranean: Comparison of the two calabrian coasts and distribution from coastal areas to the open sea. International Journal of Environmental Research and Public Health, 18(20). https://doi.org/10.3390/ijerph182010712

Martellini, T., Guerranti, C., Scopetani, C., Ugolini, A., Chelazzi, D., & Cincinelli, A. (2018). A snapshot of microplastics in the coastal areas of the Mediterranean Sea. TrAC - Trends in Analytical Chemistry, 109, 173–179. https://doi.org/10.1016/j.trac.2018.09.028

Miller, E., Sedlak, M., Lin, D., Box, C., Holleman, C., Rochman, C. M., & Sutton, R. (2021). Recommended best practices for collecting, analyzing, and reporting microplastics in environmental media: Lessons learned from comprehensive monitoring of San Francisco Bay. Journal of Hazardous Materials, 409. https://doi.org/10.1016/j.jhazmat.2020.124770

Nuelle, M. T., Dekiff, J. H., Remy, D., & Fries, E. (2014). A new analytical approach for monitoring microplastics in marine sediments. Environmental Pollution, 184, 161–169. https://doi.org/10.1016/j.envpol.2013.07.027

Pan, Z., Guo, H., Chen, H., Wang, S., Sun, X., Zou, Q., Zhang, Y., Lin, H., Cai, S., & Huang, J. (2019). Microplastics in the Northwestern Pacific: Abundance, distribution, and characteristics. Science of the Total Environment, 650, 1913–1922. https://doi.org/10.1016/j.scitotenv.2018.09.244

Petrisor, I. (2005). Fingerprinting in environmental forensics. Environmental Forensics, 6(2), 101–102. https://doi.org/10.1080/15275920590952702

Petrisor, I. G. (2014). Environmental Forensics Fundamentals. Environmental Forensics Fundamentals. https://doi.org/10.1201/b17158

Philp, R. P. (2014). An overview of environmental forensics. Geologica Acta, 12(4), 363–374. https://doi.org/10.1344/GeologicaActa2014.12.4.7

Plastics Europe. (2019). Plastics - the Facts 2019. An Analysis of European Plastics Production, Demand and Waste Data.

Prata, J. C., Reis, V., da Costa, J. P., Mouneyrac, C., Duarte, A. C., & Rocha-Santos, T. (2021). Contamination issues as a challenge in quality control and quality assurance in microplastics analytics. Journal of Hazardous Materials, 403. https://doi.org/10.1016/j.jhazmat.2020.123660

Primpke, S., Christiansen, S. H., Cowger, W., De Frond, H., Deshpande, A., Fischer, M., Holland, E. B., Meyns, M., O'Donnell, B. A., Ossmann, B. E., Pittroff, M., Sarau, G., Scholz-Böttcher, B. M., & Wiggin, K. J. (2020). Critical Assessment of Analytical Methods for the Harmonized and Cost-Efficient Analysis of Microplastics. Applied Spectroscopy, 74(9), 1012–1047. https://doi.org/10.1177/0003702820921465

Rivers, M. L., Gwinnett, C., & Woodall, L. C. (2019). Quantification is more than counting: Actions required to accurately quantify and report isolated marine microplastics. Marine Pollution Bulletin, 139, 100–104. https://doi.org/10.1016/j.marpolbul.2018.12.024

Robertson James, Roux Claude and Wiggins, K. G. (2017). Forensic Examination of Fibers (Third Edit). Taylor & Francis Group. ISBN 9781439828649.

Ross, P. S., Chastain, S., Vassilenko, E., Etemadifar, A., Zimmermann, S., Quesnel, S. A., Eert, J., Solomon, E., Patankar, S., Posacka, A. M., & Williams, B. (2021). Pervasive distribution of polyester fibres in the Arctic Ocean is driven by Atlantic inputs. Nature Communications, 12(1). https://doi.org/10.1038/s41467-020-20347-1

Ruiz-Orejón, L. F., Sardá, R., & Ramis-Pujol, J. (2016). Floating plastic debris in the Central and Western Mediterranean Sea. Marine Environmental Research, 120, 136–144. https://doi.org/10.1016/j.marenvres.2016.08.001

S. K, A., & Varghese, G. K. (2020). Environmental forensic analysis of the microplastic pollution at "Nattika" Beach, Kerala Coast, India. Environmental Forensics, 21(1), 21–36. https://doi.org/10.1080/15275922.2019.1693442

Shahul Hamid, F., Bhatti, M. S., Anuar, N., Anuar, N., Mohan, P., & Periathamby, A. (2018). Worldwide distribution and abundance of microplastic: How dire is the situation? Waste Management and Research, 36(10), 873–897. https://doi.org/10.1177/0734242X18785730

Shim, W. J., Hong, S. H., & Eo, S. E. (2017). Identification methods in microplastic analysis: A review. Analytical Methods, 9(9), 1384–1391. https://doi.org/10.1039/c6ay02558g

Silva, A. B., Bastos, A. S., Justino, C. I. L., da Costa, J. P., Duarte, A. C., & Rocha-Santos, T. A. P. (2018). Microplastics in the environment: Challenges in analytical chemistry - A review. Analytica Chimica Acta, 1017, 1–19. https://doi.org/10.1016/j.aca.2018.02.043

Suaria, G., Achtypi, A., Perold, V., Lee, J. R., Pierucci, A., Bornman, T. G., Aliani, S., & Ryan, P. G. (2020). Microfibers in oceanic surface waters: A global characterization. Science Advances, 6(23). https://doi.org/10.1126/sciadv.aay8493

Suaria, G., Avio, C. G., Mineo, A., Lattin, G. L., Magaldi, M. G., Belmonte, G., Moore, C. J., Regoli, F., & Aliani, S. (2016). The Mediterranean Plastic Soup: Synthetic polymers in Mediterranean surface waters. Scientific Reports, 6. https://doi.org/10.1038/srep37551

Suaria, G., Perold, V., Lee, J. R., Lebouard, F., Aliani, S., & Ryan, P. G. (2020). Floating macro- and microplastics around the Southern Ocean: Results from the Antarctic Circumnavigation Expedition. Environment International, 136. https://doi.org/10.1016/j.envint.2020.105494

van der Hal, N., Ariel, A., & Angel, D. L. (2017). Exceptionally high abundances of microplastics in the oligotrophic Israeli Mediterranean coastal waters. Marine Pollution Bulletin, 116(1–2), 151–155. https://doi.org/10.1016/j.marpolbul.2016.12.052

Vermaire, J. C., Pomeroy, C., Herczegh, S. M., Haggart, O., & Murphy, M. (2017). Microplastic abundance and distribution in the open water and sediment of the Ottawa River, Canada, and its tributaries. Facets, 2(1), 301–314. https://doi.org/10.1139/facets-2016-0070

Waller, C. L., Griffiths, H. J., Waluda, C. M., Thorpe, S. E., Loaiza, I., Moreno, B., Pacherres, C. O., & Hughes, K. A. (2017). Microplastics in the Antarctic marine system: An emerging area of research. Science of the Total Environment, 598, 220–227. https://doi.org/10.1016/j.scitotenv.2017.03.283

Wang, S., Chen, H., Zhou, X., Tian, Y., Lin, C., Wang, W., Zhou, K., Zhang, Y., & Lin, H. (2020). Microplastic abundance, distribution and composition in the mid-west Pacific Ocean. Environmental Pollution, 264. https://doi.org/10.1016/j.envpol.2020.114125

Wang, W., & Wang, J. (2018). Investigation of microplastics in aquatic environments: An overview of the methods used, from field sampling to laboratory analysis. TrAC - Trends in Analytical Chemistry, 108, 195–202. https://doi.org/10.1016/j.trac.2018.08.026

Woodall, L. C., Gwinnett, C., Packer, M., Thompson, R. C., Robinson, L. F., & Paterson, G. L. J. (2015). Using a forensic science approach to minimize environmental contamination and to identify microfibres in marine sediments. Marine Pollution Bulletin, 95(1), 40–46. https://doi.org/10.1016/j.marpolbul.2015.04.044