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Spatial distribution of microplastics in volcanic lake water and sediments: Relationships with depth and sediment grain size

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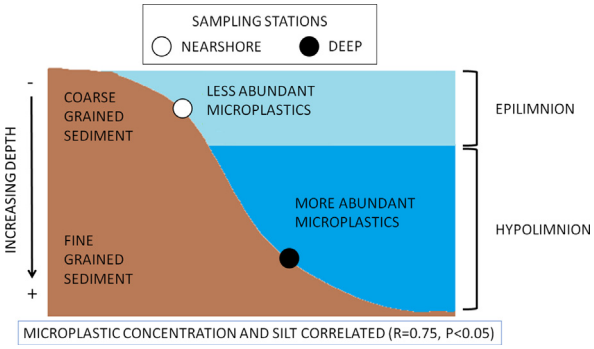
HIGHLIGHTS

- Microplastics survey in a volcanic warm monomictic lake during water stratification
- Microplastics were found in water and sediment with similar distribution pattern.
- Microplastic polymers in water differ from the ones in sediment.
- Microplastics abundance positively associates with fine-grained sediment.
- Microplastics are more abundant in lacustrine deep sediments than coastal ones.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastics (plastics <5 mm) are globally widespread pollutants of aquatic ecosystems. As microplastics contaminate both water and sediments, research on their spatial distribution in these different environmental matrices has increased. However, fresh waters are poorly studied and even less so are lentic ecosystems. To contribute filling this knowledge gap, this study analyses the distribution of microplastics in the water column and surface sediments of a volcanic lake, namely Lake Bracciano. Furthermore, it analyses in more detail the relationship between the concentration of microplastics in sediments, its grain size and the sampling depth (*i.e.* nearshore or deep). Water and sediment sampling was carried out in different sectors of the lake (northern, eastern, southern, western) using a plankton net and a van Veen grab sampler, respectively. Two sediment samples were collected at each station in order to analyse the abundance of microplastic and to perform grain size analysis. Results show a mean concentration of 2.4 items m^{-3} in water and 42 items kg^{-1} in sediments. The distribution of microplastics is uneven between the different sampling stations, with the northern sector being the most contaminated in both matrices. The chemical composition and shape of microplastics vary between water and sediment. In particular, polyethylene terephthalate and polyvinyl chloride are the most abundant polymers in water and sediments, respectively. Fibres are the main shape of microplastics in water while fragments are more abundant in sediments. In-depth analysis of sediment shows that sediments from deep stations are more contaminated than nearshore samples and have more fragment-shaped microplastics than fibre-shaped ones. Furthermore, there is a significant positive correlation between the concentration of microplastics and the abundance of silt, confirming data emerging from the scientific literature on marine and lotic ecosystems.

1. Introduction

Plastics are synthetic materials the production of which has dramatically increased over the last century due to their high versatility for domestic and industrial uses (Andrady and Neal, 2009). In fact, plastic products

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have several positive characteristics, such as extreme durability and reshaping through heat; furthermore, additives in plastics can modify their properties for more specific uses. However, the soaring production of plastics has also led to an increased dispersion of mismanaged plastic waste into the environment (Geyer et al., 2017). Plastics degrade slowly so they accumulate in the environment, contaminating both terrestrial and aquatic ecosystems (Horton et al., 2017; Law, 2017). The latter have gained great attention from the scientific community since the '70s, when reports of plastic pollution of seas and oceans began, and research has increased more and more since then (Ryan, 2015). Plastics in the marine environment affect ecosystem services, due to multiple negative impacts on ecosystems, society and economy (Kumar et al., 2021b). For instance, plastics impact provision by negatively affecting fishing and recreational activities. The estimated cost of one ton of marine ecosystems-polluting plastic is USD 3300–33,000 per year (Beaumont et al., 2019). Considering that land-based plastic input to global oceans ranges between 4.8 and 12.7 million metric tons and is predicted to increase in the forthcoming years under a business as usual scenario (Borrelle et al., 2020; Jambeck et al., 2015), the overall economic impact of plastic pollution reaches considerable levels.

Once plastics are dispersed in the environment, they are exposed to the action of biological organisms and to exogenous agents, such as solar radiation, wind and waves, which accelerate degradation through thermal, photo-oxidative and mechanical stimuli (Singh and Sharma, 2008). The breakdown of plastic items produces microplastics (plastics <5 mm; MPs), which are widespread in soil, water and air (Barnes et al., 2009; Rillig and Lehmann, 2020; Trainic et al., 2020). MPs cause negative physical and ecotoxicological impacts on organisms induced by their self or by chemicals conveyed by them. The latter include both the additives introduced during plastic production and the heavy metals and pollutants subsequently adsorbed by MPs dispersed in the environment (Lee et al., 2019). Among the negative impacts of MPs on organisms, sub-individual effects induced have been observed at molecular, cellular, and systemic level, showing alterations of physiological processes (Franzellitti et al., 2019).

The study of the distribution of MPs and the evaluation of the physical factors that influence their transport and accumulation in aquatic ecosystems are relevant topics of investigation (Akdogan and Guven, 2019). The distribution pattern of MPs is more widely studied in surface waters than in sediments, although sediments are considered to be main sinks of MPs in both marine and inland ecosystems, showing much higher concentrations than in surface waters (Woodall et al., 2014). Links between MPs distribution in waters, and in underlying sediments, as well as the relationships with sediment grain size are poorly studied aspects, especially in fresh waters, where research is lacking compared to marine systems (Blettler et al., 2018). This study aims to contribute to the understanding of MPs transport, settling, and exchanges between environmental matrices by examining the relationships between MPs concentration in water and sediments and the influence of sampling depth and grain size in a lentic system.

2. Materials and methods

2.1. Study area

This study was conducted in Lake Bracciano (Fig. 1a), located in Central Italy, about 30 km northwest of Rome (Fig. 1b). It is a subcircular volcanic-tectonic depression developed during the Pleistocene and belonging to the Sabatini Volcanic District (De Rita et al., 1996). Lake Bracciano is located about 164 m above sea level, occupies an area of 57 km^2 with a volume of 5 km^3 and has a mean depth of 88 m, reaching a maximum depth of 160 m (Stella, 1984). It has a water retention time of 137 years (Ambrosetti et al., 2003). The drainage basin of the lake is 91 km^2 and the underground infiltration basin is approximately 110 km^2 (Dragoni et al., 2006). Lake Bracciano has minor tributaries (Baccetti et al., 2017) and one natural emissary river, the Arrone River (Fig. 1a), located in the south-eastern shore of the lake and currently controlled by an artificial dam. The Arrone River is mainly dry in its upper reach, surfacing about 4 km downstream of the lake outlet (Mazza et al., 2015). Lake Bracciano has been classified as an oligo-mesotrophic, meromictic, warm monomictic lake (Barbanti et al., 1996). The bathymetry of the lake in the northern, eastern and western sectors is characterized by steep slopes between the shoreline and the 50 m isobath, while more gentle slopes characterize the southern side (Fig. 1a).

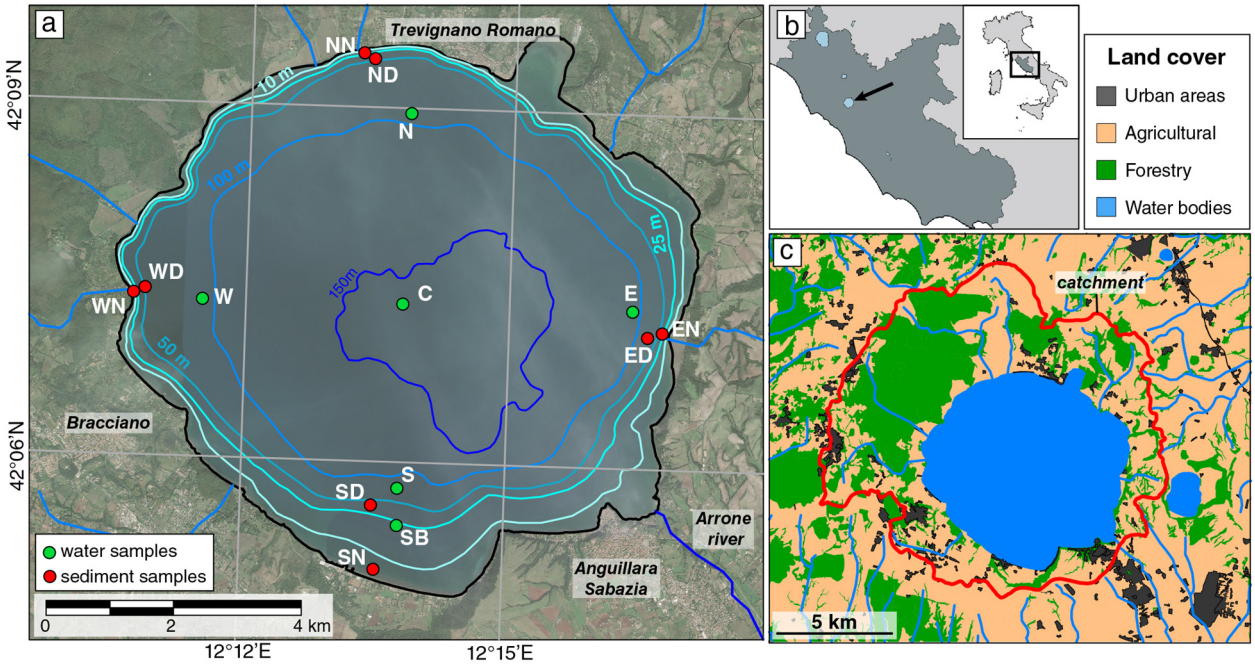


Fig. 1. a) Location of sampling stations in Lake Bracciano. Labels for water samples are: N-North; S-South; E-East; W-West. Labels for sediment samples are: NN-North nearshore; ND-North deep; SN-South nearshore; SD-South deep; EN-East nearshore; ED- East deep; WN-West nearshore; WD- West deep. The isobaths at 10, 25, 50, 150 m are shown. b) Geographical location of Lake Bracciano (Italy, Latium). c) Catchment of Lake Bracciano and land cover description. The map of land cover was downloaded from the national geo-database of the Italian Ministry of Environment (http://www.pcn.minambiente.it/mattm/). The blue lines represent the drainage network.

As regards the land use, the surroundings of Lake Bracciano are mostly characterized by agricultural lands and forestry areas (Fig. 1c), with small urbanized areas corresponding to the town of Trevignano Romano, Anguillara Sabazia and Bracciano (Fig. 1a). Trevignano and Anguillara are located on the north-eastern and south-eastern shores of the lake respectively, and host important touristic facilities along the shoreline. The town of Bracciano is located on a hill facing the lake in the southwestern sector and it hosts touristic facilities along the lake shoreline as well.

Besides being a tourist destination, Lake Bracciano provides fish species of commercial interest that support amateur and professional fishing activities and it is a water reserve for Rome. A wastewater collector surrounds Lake Bracciano and conveys it to a treatment plant, which is located in Anguillara Sabazia and since 1986 has been flowing out the treated waters into the Arrone River (Bruno et al., 2006). This collector was built to maintain high water quality standards and to counteract the eutrophication of Lake Bracciano, also in order to provide drinking water to nearby Rome (Bruno et al., 2006), the population of which was approximately 2.9 million inhabitants in 2017 (https://www.comune.roma.it/web/it/roma-statistica-popolazione.page [consulted on February, 04th 2019]).

Lake Bracciano is characterized by the highest environmental quality among the volcanic lakes of central Italy as regards the community of zooplankton, littoral vegetation, and macrobenthos (Margaritora et al., 2003). Chemical analyses of water are scarce and show a good overall quality, although they also point out high levels of nitrogen in the epilimnion (Catalani et al., 2006). Lake Bracciano has been protected by the Regional Park "complesso lacuale di Bracciano Martignano" since 1987 (Regional Law number 36/99). Moreover, it belongs to the Mediterranean biogeographic region Special Protected Area named "Comprensorio Bracciano-Martignano" (code IT6030085). In recent years it has become a Site of Community Importance named "Lago di Bracciano" (code IT6030010) and a Special Area of Conservation (Ministerial Decree 06/12/2016) of the Natura 2000 ecological network (Decision n. 2019/22 of the European Commission – document of reference number C(2018) 8534). In fact, species of conservational interest inhabit the lake, such as *Rutilus rubilio* (Bonaparte, 1837) and *Padogobius nigricans* (Canestrini, 1867). Both *R. rubilio* and *P. nigricans* are included in Annex II of the Habitat Directive (92/43/CEE) and in Appendix III of the Bern Convention on the Conservation of European Wildlife and Natural Habitats. According to IUCN, *R. rubilio* is a near threatened species, while *P. nigricans* is a vulnerable species.

Recently, an ecological stress has been highlighted in Lake Bracciano due to the lowering of the water level (Rossi et al., 2019). The causes are attributable to a series of drought events in 2008, 2012, 2016, and 2017 and the simultaneous increase of water intake, especially by the joint-stock company "ACEA", which supplies drinking water to Rome and other areas in Central Italy and which uses the 54% of the water stock of Lake Bracciano (Baccetti et al., 2017).

As regards plastic contamination, the presence of polyethylene terephthalate, polystyrene, polysiloxanes and polyester items was detected on the beaches and on the exposed littoral zone (due to the recent drought) located in the northern and southern sectors of the lake (Corti et al., 2020). The concentration of MPs in sediment samples shows a variability related to the influence and seasonality of wind and waves, *e.g.* the beaches more exposed to winds have more MPs and more plastic waste is found in May than in September (Corti et al., 2020; Iannilli et al., 2020). Furthermore, presence of MPs (*i.e.* nylon and polyethylene) was detected in the amphipod *Cryptorchestia garbinii* (Ruffo, Tarocco & Latella, 2014), with MPs concentrations in organisms positively correlated to their concentration in the surrounding beach sediments (Iannilli et al., 2020).

2.2. Sampling activities

Sampling activities were performed during the summer stagnation phase in 2019 and included the collection of water and sediment samples. The water samples were collected at four stations located in the different sectors of the lake (*i.e.* North, East, South and West) along the −100 m isobath, plus one station located in the centre of the lake and one nearshore station along the −25 m isobath (Appendix A and Fig. 1). At each station, the water column was sampled from the surface down to 10 m depth (coinciding with the epilimnion) using a plankton net (80 μm mesh, diameter of 31 cm). The volume (V) of filtered water was obtained by the formula: $V = \pi(d/2)^2h$, where "d" is the diameter of the plankton net opening and "h" is the depth set (adapted from Viršek et al., 2016). Hence, 754 dm^3 of water was filtered in each station and all suspended material retained by the net was funnelled in the cod end bucket. 200 ml of water retained in the bucket were then collected. Water samples were stored in plastic containers, which were previously rinsed with distilled water to avoid contamination (Leslie et al., 2017). During the water sampling, procedural blanks were conducted by opening three containers filled with distilled water and leaving them exposed during field operations.

Sediment samples were collected in the different sectors of the lake (*i.e.* North, East, South and West). For each sector, we selected two stations at two different depth ranges (2–2.5 m and 17–42 m, hereafter referred to as nearshore and deep, respectively), resulting in 8 samples (Appendix A and Fig. 1). The sediment was collected by a Van Veen grab sampler (2 l), which allowed the collection of relatively undisturbed samples at most stations, due to sediment cohesion. The superficial layer of the sediment (about 1 cm thick) was tentatively taken using metal tools and placed in glass jars for the assessment of MPs content. Moreover, about 100 g of sediment were collected and stored separately for grain size analysis at all stations except the Western Deep, due to the abundant vegetal material occurring in this sample.

2.3. Microplastics extraction from water and sediment samples

The volume of water samples from each station was filtered on Whatman® GF/A glass microfiber filters (n. 1820–090, diameter of 90 mm, pore size of 1.6 μm) using a vacuum pump. Washing of the containers with distilled water on the filters was performed to maximise the recovery of particles that could remain attached to the walls.

Pre-treatment of sediment samples was carried out following the protocol by Frias et al. (2018). Samples were dried at 40 °C in a laboratory stove to obtain the dry weight. The organic matter was removed by adding 300 ml of H_2O_2 (10% solution). After stirring for 60 s the samples were covered with aluminium foil and put in a fume hood for 18 h to complete the digestion process. The H_2O_2 was then removed by rinsing the samples with distilled water on a 63 μm stainless steel sieve.

MPs and light particles were separated from the sediment according to the density separation approach developed by Thompson et al. (2004). This approach aims to isolate the lighter plastic items from the heavier sediment grains by mixing the sample with a hypersaline solution in which the plastic items can float or remain in suspension, while the sediment settles rapidly. A saturated NaCl solution (1.2 g cm^{-3}) was added to the sediment samples, with a volumetric ratio of approximately 3:1 (NaCl solution: sediment). The choice of NaCl was made since this salt is cheap, easily available, environment-friendly, has been widely used to date and allows the recovery of the most common polymers (European Commission, 2013). The samples were stirred for 30 min and the supernatant was collected in bakers after 5 min of decantation. Current scientific literature does not provide clear indication of tested and validated durations for stirring and decantation (Lusher et al., 2020). For the samples in this study, the settling time of 5 min was considered effective, since the fine fraction of sediment (*i.e.* the fraction that requires more time to settle) had been removed during pre-treatment, as well as the organic matter. The supernatant was then filtered on Whatman® GF/A glass microfiber filters by a vacuum pump. To maximise recovery efficiency, the bakers were rinsed with distilled water, which was also filtered, and the density separation process was repeated 3 times for each sample (Woodall et al., 2014). All filters were dried at 20 °C in sealed glass Petri dishes. Putative plastic items from water and sediment samples were identified by stereomicroscope observations and their colour and shape described (Lusher et al., 2020).

A quality control was conducted on laboratory preparation, air condition and by negative controls, according to Koelmans et al. (2019). Cotton lab coats and gloves were used during all laboratory activities. In addition, the NaCl solution was previously filtered on a 63 μm sieve to eliminate contaminating particles that may occur in the salt. Negative controls were performed using three procedural blanks obtained from distilled water (samples exposed during field sampling) and three from the NaCl solution, to correct for background contamination for water and sediment samples, respectively. Atmospheric contamination in the laboratory was assessed by placing a clean filter in an open glass Petri dish on the workstation during the MPs extraction process. Procedural blanks for different contaminations (distilled water/field sampling, NaCl solution, laboratory air) were then observed using a stereomicroscope to detect potential MPs.

2.4. Polymer identification

All suspected plastic items from field and control samples were analysed using an InVia Renishaw Micro-Raman spectrometer equipped with a Leica DM2700 M confocal microscope. Two objectives, an Olympus 20× LWD, and a Leica 50× LWD have been used to focus the beam down to few microns (≈ 5 μm). Two solid-state diode laser sources, one at 532 nm (nominal output 100 mW) and the other at 785 nm (nominal output 200 mW), have been used. The high-contrast rejection for elastically scattered light is provided by holographic edge filters. Anelastically scattered light is dispersed by a 1800 line/mm diffraction grating suitable to achieve a spectral resolution of 1 cm^{-1} when using the 532 nm excitation wavelength; a 1200 line/mm grating has been used with primary wavelength of 785 nm instead. A Peltier cooled 1024 × 256 pixel CCD detector collects the dispersed light. Raman spectra have been collected in the 100–3200 cm^{-1} range. To achieve suitable statistics, an integration time of about 10 s and one accumulation has been found to be adequate for most of the investigated samples. Wire software has been used to set the experimental conditions.

The interpretation of spectra used freely available software *i.e.* SpectraGryph (Menges, 2020), and reference spectra from literature. Raman reference spectra were obtained from the "SLOPP" and "SLOPP-E" databases (Munno et al., 2020) and from literature (*e.g.* Bokobza et al., 2015). Similarities values less than 60% obtained from an automatic search were not accepted. If the similarity value was between 60% and 70%, the sample and reference spectra were visually confirmed. If the value was greater than 70%, the identification was accepted (European Commission, 2013). Sample spectra that did not have an automatic accepted identification were visually compared with spectra available in the scientific literature. All suspected plastic particles that were identified as non-plastic by Raman analysis were excluded from our results. The results were expressed in items of MPs m^{-3} for water and items of MPs kg^{-1} d.w. (dry weight) for sediments according to the prevalent units of measurement used by scientific literature (Cera et al., 2020).

2.5. Grain size analysis

Seven sediment samples were analysed to assess the grain size distribution (Appendix A). Pre-treatment of samples was performed using H_2O_2 (20% solution) and distilled water to remove organic matter and salts. Samples were then dried at 40 °C in a laboratory stove to obtain the dry weight. Gravel and sand (particles >63 μm) were separated from silt and clay (particles <63 μm) by wet sieving. Grain size for the coarse fraction (>63 μm) was determined by dry sieving at half phi intervals with vibro screens, while the fine fraction was analysed by a laser diffraction grain size analyser (Sympatech). The total grain size distribution was obtained by proportional recombination of the individual data sets.

The descriptive statistics parameters of grain-size distribution (mean grain size, sorting, skewness, kurtosis) were calculated using the Folk and Ward (1957) formulas and samples were classified based on Folk classification scheme (Folk, 1954, 1974) using SEDPLOT software (Poppe and Eliason, 2008).

2.6. Statistical analysis

The relationships between the concentration of MPs in sediments and the environmental parameters (*i.e.* depth range, grain size characteristics) were explored using statistical analysis. In particular, two hypotheses were tested:

1) to test whether there was a difference between the concentrations of MPs in nearshore and deep stations, a Mann-Whitney test was carried out. This non-parametric analysis for two independent samples was chosen since the Shapiro normality test showed a distribution of data not normal.
2) To assess the potential relationships between MPs concentration in sediments and grain size characteristics, Pearson's correlation test was used, since data distribution was normal according to Shapiro test. Spearman's correlation test was performed only to analyse the relation between the concentration of MPs and the abundance of gravel in the sediments, since the latter did not have a normal distribution. All statistical analyses were performed using PAST software ver.3 (Hammer et al., 2001).

3. Results

3.1. Microplastics in water

The check of controls showed neglectable contamination of the samples as only one spectrum, *i.e.* a blue coloured polypropylene (PP), collected on a western sample, matched a spectrum of the controls. That item was excluded by the MPs counted in the western station. Cumulatively, 11 MPs were found in stations N, W, SB, and C while no MPs were detected in the remaining stations (*i.e.* S and E). The mean contamination was 2.4 MPs m^{-3}. The most contaminated stations were N (5.3 MPs m^{-3}) and W (5.3 MPs m^{-3}), followed by C (2.7 MPs m^{-3}) and SB (1.3 MPs m^{-3}) (Fig. 2a).

The MPs in water were composed of fibres (10 MPs), fragments (1 MPs) and no beads. Black and clear MPs were the most abundant ones with 7 MPs and 3 MPs of occurrence, respectively. One blue MPs was also observed, which corresponds to the only fragment detected (Appendix B).

Polymers found included polyethylene terephthalate (PET) (4 MPs), polycarbonate with a black colourant (PC) (4 MPs), polyester (PL) (1 MPs), aromatic polyamide (aPA) (1 MPs) and PP (1 MPs) (Fig. 2a-3 and Appendix C).

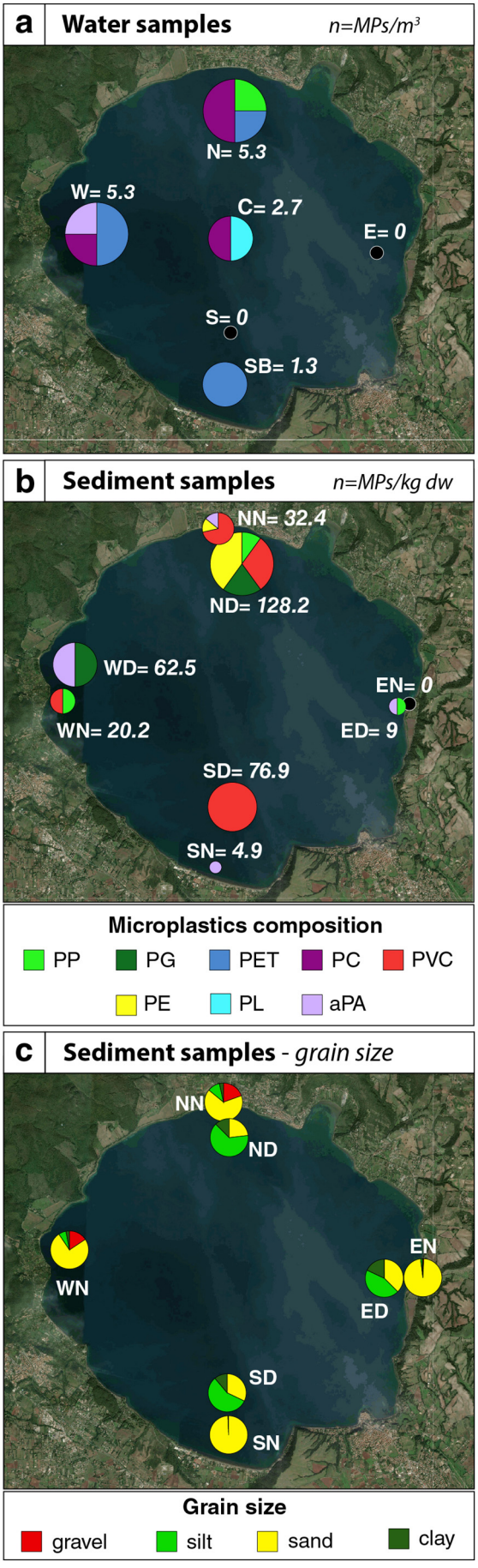
3.2. Microplastics in sediment

Samples were not contaminated with exogenous plastics according to our quality control. A total of 27 MPs were found in sediment samples corresponding to 42 MPs kg^{-1} d.w. (Fig. 2b), with the highest concentration observed in ND station (128 MPs kg^{-1} d.w.) and the lowest in SN station (5 MPs kg^{-1} d.w.) (Fig. 2b, Appendix D). No MPs occurred in EN station. The northern sector (mean = 80 MPs kg^{-1} d.w.) was more contaminated than the western (mean = 41 MPs kg^{-1} d.w.), southern (mean = 41 MPs kg^{-1} d.w.), and eastern (mean = 5 MPs kg^{-1} d.w.) sectors.

Regarding the difference in MPs concentration in relation to depth, nearshore stations (*i.e.* NN, WN, SN, EN; mean = 17 MPs kg^{-1} d.w.) were less contaminated than deep stations (*i.e.* ND, WD, SD, ED; mean = 67 MPs kg^{-1} d.w.) (Mann-Whitney, $p > 0.05$).

The majority of MPs in sediments were fragments (21 MPs), followed by fibres (6 MPs). No beads were observed (Appendix D). Fragments were more abundant in deep stations than in nearshore stations, while the opposite was observed for fibres. The main colour of the observed MPs was blue (15 MPs), followed by green and black (4 MPs each), and clear and red (2 MPs each) (Appendix D). With the exception of one blue and one red fibre, all remaining 4 fibres were black. Differently, fragments were mostly blue, then in decreasing order of abundance: green, clear, and red.

The polymers found included polyvinyl chloride (PVC) (12 MPs), polyethylene (PE) (5 MPs), aPA (4 MPS) and PP (3 MPs) (Fig. 4). In addition, a



colourant used for plastics called phthalo green (PG) was also detected in fragment-shaped items (3 MPs) (Fig. 2b). We interpreted the presence of PG as a hint of the plastic origin of the analysed items, thus we considered those items as MPs (Appendix C).

3.3. Grain size analysis

The analysed samples could be roughly divided into two groups, with a prevalence of coarse (gravel and sand) or fine (silt and clay) fraction, which corresponded to samples from nearshore and deep stations respectively (Table 1, Fig. 2c). Specifically, sand was prevalent in nearshore samples (NN, WN, SN and EN) ranging between 66% and 99%. Samples from the northern and western sectors (NN and WD) were also characterized by a gravelly fraction of 20% and 16% respectively and by a higher proportion of silt and clay (6–10% and 3–3.5% respectively), resulting in poorly sorted sediment. Conversely, nearshore samples from southern and eastern station were well sorted (Table 1). Silt was prevalent in deep samples (ND, SD and ED), ranging from 45% to 64%. Sand was present at 23 to 37%, while clay content ranged from 11 to 17%, resulting in a poor sorting for deep samples. Sediment distribution from nearshore samples was symmetrical and generally leptokurtic while the deep ones had a strong positive asymmetry and generally platykurtic distribution (Table 1, Appendix E). There was a significant positive correlation between the percentage of silt and the concentration of MPs in sediments ($R = 0.75$, $p < 0.05$).

4. Discussion

Results of the analysis on water and sediment samples collected in Lake Bracciano showed a higher concentration of MPs in the northern sector of the lake. Despite the different sampling procedures and methods for MPs extraction, these observations are in agreement with the distribution pattern described in the report analysing the water contamination by MPs in Lake Bracciano and which confirms the northern sector as the most contaminated (Legambiente, 2018). Conversely, other studies carried out along the beaches of this lake found an opposite trend, with greater plastic contamination in the southern sector, more exposed to the action of wind, compared to the northern shore of the lake (Corti et al., 2020; Iannilli et al., 2020). However, it should be taken into consideration that Iannilli et al. (2020) analysed plastics over a wider dimensional range (from 1 mm to 32 mm), so these data are not entirely comparable. Furthermore, although Corti et al. (2020) reported more stable and higher MPs pollution in the southern beach than in the northern one, they found a much wider range of MPs variability (particularly PET) within samples from the northern beach, which can locally reach up to 9 times the weight of MPs found on the opposite side.

Overall, the studies cited above suggest that the distribution of MPs on beaches is strongly influenced by the action of wind and waves, and highlight that wind-exposed beaches are more contaminated, as indicated by observations in three different volcanic lakes of this region, namely Bracciano, Albano and Vico (Corti et al., 2020; Iannilli et al., 2020). Similarly, there are reports of variations in MPs concentration in lacustrine waters linked to the wind direction and strength (*e.g.* Fischer et al., 2016). Wind is widely recognized as an important factor influencing distribution of MPs in lakes even in remote areas (Free et al., 2014) and transitional ecosystems (Lorenzi et al., 2021). Therefore, the observed distribution of MPs in the sediments and waters of Lake Bracciano could be the result of the interplay between the action of wind, waves and currents. However, the impacts of local sources of MPs cannot be excluded as the northern stations, which are more contaminated than the other stations, are the only ones located near both an urban centre (*i.e.* Trevignano Romano) and the mouth of a small tributary (Fig. 1). Hence, it is suggested that these two potential sources of MPs could act cumulatively resulting in a higher abundance of MPs in northern sector. Nevertheless, considering the uncertainties on the relative influence of physical

Fig. 2. Microplastics abundance and polymeric composition present in a) water and b) sediment sampling stations. c) Results of the grain size analysis showing the percentages of gravel, sand, silt and clay in each sampled station.

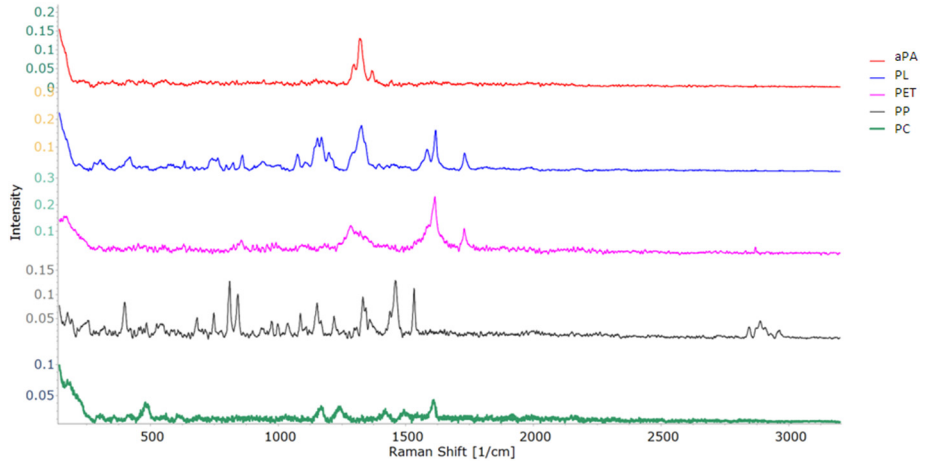


Fig. 3. Interpretation of Raman spectra collected from water samples: black aromatic polyamide (aPA), black polyester (PL), black polyethylene terephthalate (PET), blue polypropylene (PP), and black polycarbonate (PC). Spectra obtained by 785 nm source, magnification 50×, 0.1%–5% range of laser intensity, 10 s acquisition time, 1 accumulation. Spectra processed by smoothing and background, spikes removed.

and anthropic factors, further research is mandatory for a better understanding of the distribution of MPs in this lentic system.

Regarding the distribution of MPs in surficial sediments and their relationship with depth and grain size, the greater abundance of MPs in the deep stations is consistent with the positive association of MPs with silt. In fact, deep sediments are characterized by a higher percentage of mud. Conversely, nearshore sediments are less contaminated by MPs and have a prevalence of coarse sediments. The MPs deposition pattern could be influenced by the grain size of the sediment, as MPs have been shown to infiltrate deeper into coarse sandy and gravelly sediments than fine sediments (Waldschläger and Schüttrumpf, 2020), thus suggesting that fine sediments may act as a "MPs trap" which concentrates these particles on the surficial layer. However, the strength of the water currents may also play a major role in the deposition of MPs (Tibbetts et al., 2018). In this sense, the strong energy of wave motion in the epilimnion of Lake Bracciano could limit the deposition of both fine sediments and MPs in nearshore stations, while the less intense hydrodynamics in hypolimnion could favour the settling of MPs in the deeper stations.

Our interpretation is supported by the scientific literature examining the mechanisms of MPs accumulation in sediments based on grain size (Ling et al., 2017). However, few studies are available on this topic, especially in fresh waters. In their study on the occurrence of MPs in riverine sediments, Corcoran et al. (2020) found a clear relationship between MPs and grain size; in particular, MPs were significantly more abundant in very fine sand than in fine and medium sand samples (Corcoran et al., 2020). Similar results were obtained from studies on coastal marine environments, where MPs concentration was significantly higher in mud than in sand (Marques Mendes et al., 2021). A significant negative correlation was observed between MPs concentration and increasing grain size in semi-enclosed costal zones (Sun et al., 2021), as well as a significant positive correlation between MPs concentration and silt and a negative correlation with sand (Falahudin et al., 2020). A comprehensive survey including estuarine, coastal and offshore settings found a relationship between MPs size and grain size, proposing the latter as a proxy of MPs abundance (Enders et al., 2019). However, the influence of anthropogenic factors is also reported in the literature and, for instance, it is considered that the distance of the sampled site from the source of pollution significantly contributes to the relationship between MPs and grain size: the further the sampling site is from the source, the less significant is the relationship (Marques Mendes et al., 2021). In addition, open coastal areas subject to intense hydrodynamics do not show a significant relationship between MPs abundance and sediment grain size (Sun et al., 2021).

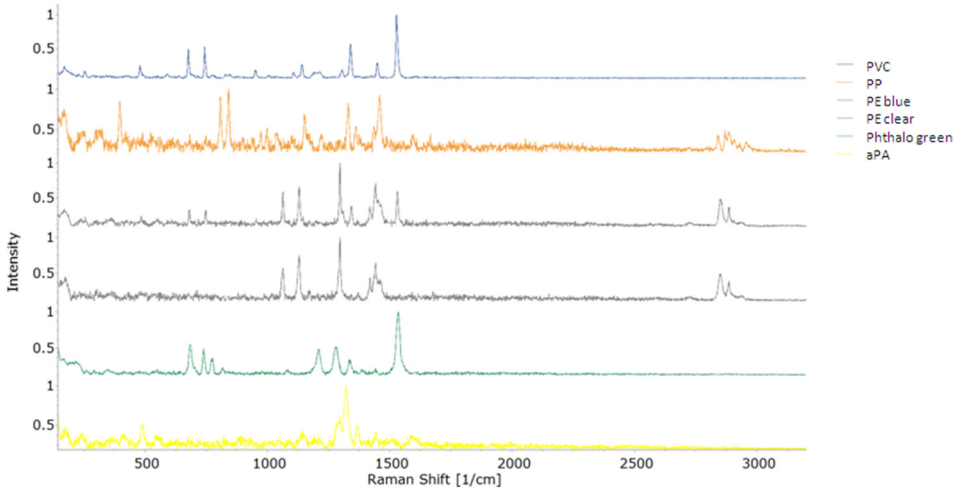


Fig. 4. Interpretation of Raman spectra collected from sediment samples: blue polyvinylchloride (PVC), blue and clear polyethylene (PE), black aromatic polyamide (aPA), purple polypropylene (PP), and green phthalo green. Spectra obtained by 785 nm source, magnification 50×, 0.1%–10% range of laser intensity, 10 s acquisition time, 1 accumulation. Spectra processed by smoothing and background, spikes removed.

Table 1
Granulometric characteristics of sediment samples, including percentages of sediment fractions for each station and corresponding Folk class, mean grain size, sorting (σ), skewness (Sk) and kurtosis (Ku).

Station	Gravel (%)	Sand (%)	Silt (%)	Clay (%)	Folk class	Mean size (φ)	σ	Sk	Ku
NN	19.49	66.48	10.55	3.48	Gravelly muddy sand	1.24	2.83	0.04	1.30
ND	0.00	23.21	64.31	12.48	Sandy silt	5.47	1.83	0.34	0.98
WN	15.65	74.96	6.40	2.99	Gravely muddy sand	0.90	2.28	0.01	1.92
SN	0.00	99.37	0.51	0.12	Sand	2.54	0.44	0.09	1.05
SD	0.06	32.05	56.75	11.14	Slightly gravelly sandy mud	5.33	1.79	0.42	0.88
EN	0.78	97.26	1.32	0.64	Slightly gravelly sand	2.17	0.76	−0.03	1.20
ED	0.00	37.47	44.56	17.97	Sandy silt	5.45	2.20	0.37	0.71

Despite the sources of MPs in Lake Bracciano are likely the same for sediment and water matrices, the results showed a striking difference in MPs shape and polymeric composition. As regard to the difference in shape observed between the MPs in water and sediment samples (*i.e.* fibres are more abundant in water and fragments in sediments), this phenomenon has been observed in another lentic system, specifically Poyang Lake (China) (Jian et al., 2020). The potential causes of the observed pattern were identified in the increased suspension of fibre-shaped MPs or the presence of different sources of MPs (Jian et al., 2020).

As regards the polymeric composition of the MPs found in Lake Bracciano, Raman analysis revealed several polymeric types of MPs (PE, PP, PVC, PET, PC, aPA). PE and PP have a lower density than water (thus they should tend to float) while PVC, PET, PC, and aPA have a higher density than water (thus they should tend to sink). Some exceptions to this rule were found both in the sediments and in the water of Lake Bracciano. Regarding sediments, PP fragments were found mainly in nearshore samples (EN, WN), where sinking could be entailed by the biofilm growth on the surface of the fragments (Di Pippo et al., 2020). As an additional possible explanation, the deposition of PP fragments in coastal stations could be favoured by a greater exchange between MPs in water and sediment, due to intense hydrodynamics in shallow water setting (Kumar et al., 2021a). The PP and PE fragments found in the deep stations were all in ND, thus suggesting the influence of local factors that can increase aggregation or flocculation with higher density particles (Yan et al., 2021). Regarding water samples, the occurrence of PC and aPA fibres in epilimnion could be due to the temperature-dependent variations in the density of water associated with the thermocline and the concomitant action of water currents, which can entail a slowing of the sedimentation of PC and aPA fibres. In fact, it has been observed in marine water bodies that MPs tend to concentrate near rapid changes in water density, such as the thermocline (Uurasjärvi et al., 2021). It was also observed that the density of MPs polymers was not related to the depth at which they were sampled (Uurasjärvi et al., 2021).

Although there are methodological differences between the studies assessing MPs distribution, we compared our results and literature data, to frame the level of contamination in Lake Bracciano in a global context. Based on information obtained from recent reviews (Cera et al., 2020; Yang et al., 2022), the MPs contamination of lacustrine sediments from 47 sites located in Africa, Asia, America and Europe, ranges between 0.27 and 13,925 MPs kg^{-1}, with a mean concentration of 525 MPs kg^{-1}. The mean concentration of MPs in Lake Bracciano is much lower than global data, being 42 MPs kg^{-1}. Similarly, the level of water contamination (2.4 MPs m^{-3}) is lower than that of 62 lakes, whose contamination is between 0 and 8925 MPs m^{-3}, with a mean of 2561 MPs m^{-3}. Hence, Lake Bracciano is generally less polluted than other lakes studied. Particularly, it is less contaminated than most of the Asian, South American, and some African lakes but has MPs concentrations comparable to other European, North American, and some Asian lakes, especially in the Jiangsu and Jianxi regions (China). The presence of a circular wastewater collector that discharges water into the emissary river and not into Lake Bracciano could have contrasted the plastic pollution in this lake. Finally, the comparison with the literature data has highlighted that Lake Bracciano is among the smallest lakes in the world sampled for the assessment of MPs pollution and one of the few of volcanic origin.

5. Conclusions

This work analyses the distribution of microplastics in the water and sediment of Lake Bracciano, addressing the scientific debate on the relationship between physical factors, such as depth or characteristics of the sediment, and the occurrence of MPs, by providing novel information from a lentic system. The results obtained showed that the concentration of MPs was higher in the deep fine-grained samples than in the nearshore coarse-grained samples. Fragments were more abundant than the fibres in the deep samples. A positive and significant correlation was found between MPs concentration and silt content.

Research on the transport and deposition of MPs in fresh waters is an under-studied topic, particularly on lentic systems. Further investigations are mandatory to better assess spatial and temporal distribution of MPs in water and sediment. A better understanding of the transport, settling, and exchanges between environmental matrices of MPs could provide the basis for the assessment of the biogeological cycle of MPs in lentic ecosystems. The outputs would lead to a more accurate description of environmental contamination and risk assessment for biota to support management policies.

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CRediT authorship contribution statement

Alessandra Cera: conceptualization, data curation, formal analysis, investigation, methodology, validation, visualization, writing – original draft; **Martina Pierdomenico:** conceptualization, investigation, methodology, resources, supervision, validation, visualization, writing – original draft; **Armida Sodo:** methodology, resources, validation, writing – review & editing; **Massimiliano Scalici:** conceptualization, funding acquisition, project administration, resources, supervision, writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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