



## Monthly variability of floating plastic contamination in Lake Maggiore (Northern Italy)

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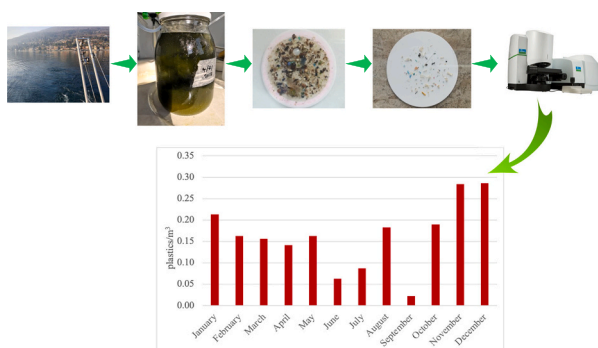
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### HIGHLIGHTS

- Plastic monitoring was conducted in Lake Maggiore water for one year (12 months).
- Lake Maggiore showed a low plastic pollution compared to other lakes worldwide.
- A level of plastic pollution from 0.02 to 0.29 plastics/m<sup>3</sup> was observed in the year.

### GRAPHICAL ABSTRACT



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### ABSTRACT

The monitoring of plastics in freshwater ecosystems has witnessed a significant increase in recent years, driven by the awareness that approximately 80 % of marine plastic litter originates from terrestrial sources transported to the seas through lakes and rivers. Consequently, it is imperative to develop monitoring plans that offer a comprehensive understanding of plastic contamination in these aquatic environments, given their seasonal variations in hydrochemical characteristics and anthropogenic sources. Historically, most global lake monitoring campaigns have been limited to one-time or, at most, seasonal sampling. In this context, the primary objective of the present study was to assess the quantitative and qualitative monthly variations of floating plastics in Lake Maggiore, a large European lake with high ecological and economic significance. Twelve transverse transects were conducted from January to December 2022 using a Manta-net with a 100 µm mesh. Characterization of each plastic particle was performed using a µ-Fourier Transform Infrared Spectroscopy (µFT-IR). The results revealed relatively low levels of contamination in Lake Maggiore when compared with other lakes worldwide exclusively from a secondary origin. However, a considerable heterogeneity was observed, both quantitatively and qualitatively. Notably, we identified a 13-fold difference between the minimum (0.02 plastics/m<sup>3</sup> in September) and maximum (0.29 plastics/m<sup>3</sup> in December) concentrations of plastics, accompanied by significant variations in polymer composition. Our monitoring underscored the necessity of also considering the temporal

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variation as a potential factor influencing plastic contamination in a lake. Moreover, frequent sampling emerged as a crucial requirement to accurately gauge the extent of plastic pollution, yielding robust and valuable data essential for effective environmental management.

## 1. Introduction

Since one of the most representative hallmarks of Anthropocene, the new geological time unit suggested by Crutzen and Stoermer (2021), is the production and accumulation of plastic litter, it has also been proposed to call Plasticene the period within the Anthropocene in which the negative effects of plastics on the Earth's biophysical systems became significantly evident at a global level (Rangel-Buitrago et al., 2022). On the other hand, plastic, composed of hundreds of classes of distinct polymers, possesses numerous characteristics, such as being lightweight, flexible, moldable, and inexpensive, that facilitate the manufacturing of a wide array of items. These products are now part of our daily lives and have often become nearly irreplaceable. This is reflected in the volumes of plastics produced globally, as the most recent data shows a production of 400.3 million tonnes in 2022 (Plastics Europe, 2023), although in Europe there has been a slight decrease in the last year (about 10 %; Plastics Europe, 2023) likely due to the bans on the use of many single-use plastic products (SUPs) now implemented for several years in various European Union countries (Directive EU 2019/904) and the greater awareness of the dangers inherent in plastic waste by EU citizens.

In recent years, plastic pollution has emerged as a primary environmental concern, with documented instances of plastic waste found in ecosystems globally. Over just a few decades, plastic products have accumulated in both terrestrial and aquatic environments, extending to remote areas such as the Poles and glaciers. This widespread issue prompted the United Nations to declare plastic pollution as one of the most pressing emerging environmental problems of our time (UNEP, 2016). While 85 % of marine litter is comprised of plastic items, it is essential to note that approximately 80 % originates from terrestrial ecosystems, primarily transported through freshwaters (Galgani et al., 2015). The pivotal role played by lakes and rivers as repositories and transport ways for plastics is underscored by the growing number of studies focusing on freshwater environments. Fig. S1 illustrates a trend of more than one order of magnitude increase in the number of papers over the last 10 years, highlighting the escalating attention to this issue.

Despite the growing interest in freshwater environments, one aspect of plastic pollution remains largely unaddressed: the intrinsic variability in plastic contamination in freshwaters. This variability may depend not only on variations in the plastics' quantity discharged from domestic and industrial activities but also on meteorological and hydrochemical changes, as well as tourism. However, there is still a lack of studies comprehensively evaluating the temporal evolution of plastic litter in freshwater ecosystems. Most studies focus solely on a single sampling in one or more sites within the studied freshwater environment. Upon entering the terms "plastics", "monthly variation," and "freshwater" as topics into the Web of Science database, we identified only one article exclusively monitoring both natural and synthetic fibers in the French Rivers Marne and Seine on a monthly basis (Dris et al., 2018). No results were found regarding plastic monitoring when utilizing the term "daily variations".

In this context, the goal of the present study was to assess the annual trend of contamination due to floating plastics in Lake Maggiore, one of the main European lakes and the second-largest Italian lake in terms of volume and surface (Giardino et al., 2014). This was achieved through a monthly monitoring campaign, aiming to provide a more accurate picture of plastic pollution compared to a single sampling. To fulfill this objective, we conducted monthly transverse transects in the central area of the lake using a Manta-net with a mesh size of 100  $\mu\text{m}$  from January to December 2022. Each sampled particle was characterized using a

$\mu$ -Fourier Transform Infrared Spectroscopy ( $\mu\text{FTIR}$ ) after appropriate sample treatment and separation of plastics. To our knowledge, this study represents the first widespread evaluation of the annual trend of plastic pollution in a lake system, obtained through monthly samplings.

## 2. Materials and methods

### 2.1. Study area and sampling

Lake Maggiore is a large, deep, subalpine lake shared between Italy and Switzerland (Fig. 1). While the lake is primarily situated in Italy, the watershed is almost equally distributed between the two countries and encompasses the outflow of another cross-border lake, Lake Lugano, along with Lake Orta and Varese. Spanning an area of 6599  $\text{km}^2$ , the watershed is predominantly mountainous but inhabited by over 500,000 people, a number that rises to an additional approximately 300,000 equivalent inhabitants during the tourist season (Piscia et al., 2023). Classified as holo-oligomictic, Lake Maggiore rarely undergoes complete vertical mixing (Fenocchi et al., 2018) and boasts a theoretical water renewal time of 4.2 years (Ambrosetti et al., 2012). The primary tributaries, namely the Rivers Ticino, Toce, Maggia, and Tresa, collectively drain around 82 % of the catchment area (Rogora et al., 2015). These tributaries exhibit different outflow patterns: Rivers Ticino and Toce experience a peak from May to October, attributed to the high altitude of their catchment basins, with a significant contribution from snowmelt and partial glacier melting. On the other hand, other tributaries showcase two peaks, one in spring and the other in autumn, primarily influenced by precipitation (Scapozza and Patocchi, 2023). Lake Maggiore is part of the Po River's catchment area, the principal river in Italy, connected to it through the Ticino outlet. Surface water sampling was conducted at the conclusion of each month from January to December 2022 using a manta trawl with a 60  $\times$  25 cm opening and a 100  $\mu\text{m}$  mesh size. The sampling was carried out along a transversal transect spanning from coast to coast in the central area, where the lake reaches its maximum depth (Fig. 1). The manta-net was towed astern of a vessel at a suitable distance to avoid turbulence from the screw, maintaining a constant speed of 3 knots. A flow meter, positioned at the mouth of the manta-net, allowed the measurement of the filtered water quantity. On average, each transversal transect covered a distance of 2.8 km, resulting in the sampling of an area of 1500  $\text{m}^2$  and the filtration of 314  $\text{m}^3$  of water. Table S1 provides details on the characteristics of the samplings. Plastics retrieved at the end of each transect were collected by washing the manta-net externally with lake water using an electric pump (Egessa et al., 2020; Akhbarizadeh et al., 2024). The concentrated materials within the collector at the manta-net's end were then transferred to glass bottles with metal caps, transported to the laboratory, and stored at 4  $^{\circ}\text{C}$  until the analyses.

### 2.2. Plastic quantification and characterization

To remove water from the collected samples, a filtration process through two overlapped sieves with mesh sizes of 5 mm and 63  $\mu\text{m}$  was employed. The filtered materials were then gathered in glass bottles by rinsing the sieves with 1 L of sodium chloride (NaCl) hypersaline solution (with a density of 1.2  $\text{g}/\text{cm}^3$ ). This solution, utilized for the density separation between plastics and particulate matter, had undergone prior filtration on glass fiber filters (1.2  $\mu\text{m}$  mesh; Whatman) to eliminate any potential plastic particles from the NaCl salt. Following overnight density separation, the obtained supernatants underwent filtration on nitrate cellulose membrane filters (8  $\mu\text{m}$  mesh; Sartorius™ 50 mm) using a

vacuum pump. Subsequently, the filtered material was digested with hydrogen peroxide ( $\text{H}_2\text{O}_2$ , 15 % v/v) to eliminate traces of organic matter from the filter surface. Suspected plastics on the filters were transferred to clean filters using tweezers and a stereomicroscope. The quantification and classification of each plastic particle in terms of shape, size, color, and polymer were carried out individually using  $\mu\text{FT-IR}$  (Spotlight 200i equipped with Spectrum Two, PerkinElmer).

The analysis was carried out using attenuated total reflectance (ATR) with 32 scans within the wavelength range of 600 to  $4000\text{ cm}^{-1}$ . Spectra were compared with standard libraries (PerkinElmer) using the Spectrum 10 software. Following a visual examination of spectrum peaks by the operator, a matching score  $\geq 0.70$  was considered. Plastics were categorized based on their shape, including lines, fibers, fragments, films, or pellets. Regarding plastic size, synthetic particles were classified according to the guidelines suggested by the International Organization for Standardization (ISO/TR 21960; 2020). This classification divided particles into macroplastics ( $> 5\text{ mm}$ ), large microplastics ( $1\text{ mm} < \text{large microplastics} \leq 5\text{ mm}$ ), and microplastics ( $1\text{ }\mu\text{m} < \text{microplastics} \leq 1\text{ mm}$ ). Plastic color was also recorded, taking into account its potential implications in the prey/predation relationship.

The quantity of synthetic particles was expressed both as the number of plastics/ $\text{m}^3$  and the number of plastics/ $\text{km}^2$ . This dual expression was employed since measurements were taken for both the volume of filtered water by the manta-net and the length of the transects (Magni et al., 2019, 2021, 2022; Sbarberi et al., 2024).

### 2.3. Quality control and assurance

To prevent plastic contamination, particularly from fibers, filters underwent processing under a laminar flow hood, remained covered in

Petri dishes, and laboratory cotton coats were worn throughout all analytical procedures. The  $\mu\text{FT-IR}$  filter holder plate was shielded from potential accidental contamination by an external protective cap.

To assess the likelihood of laboratory contamination by plastics during filtration, extraction, and analysis steps, two nitrate cellulose membrane filters placed in open Petri dishes were concurrently processed with each sample as blanks. Corrections to the results were applied when particles identified on the blank filters exhibited similar characteristics (shape, polymer, color) as those found in the samples (Magni et al., 2019, 2021, 2022; Sbarberi et al., 2024). Throughout the entire monitoring campaign, only two polyester (PEST) fibers, one black and the other transparent, detected in the June and November samplings, respectively, were eliminated.

### 2.4. Statistical analyses

The relationships between total plastics' concentrations and fibers or fragments measured in each season were checked by the Pearson correlation ( $p < 0.05$ ). The same analysis was applied to examine potential covariations between plastic concentrations and rainfall (data sourced from the Regional Agency for the Protection of the Environment, Piemonte, Italy), as well as phytoplankton density. Differences among seasonal densities were evaluated through a one-way ANOVA followed by Fisher's Least Significant Difference (LSD) *post-hoc* test ( $p < 0.05$ ).

## 3. Results

We selected 845 distinct particles through visual sorting, which were subsequently individually analyzed using  $\mu\text{FT-IR}$ . Out of these, 594 were identified as synthetic polymers (70 %; Table S2), 191 were cellulose or

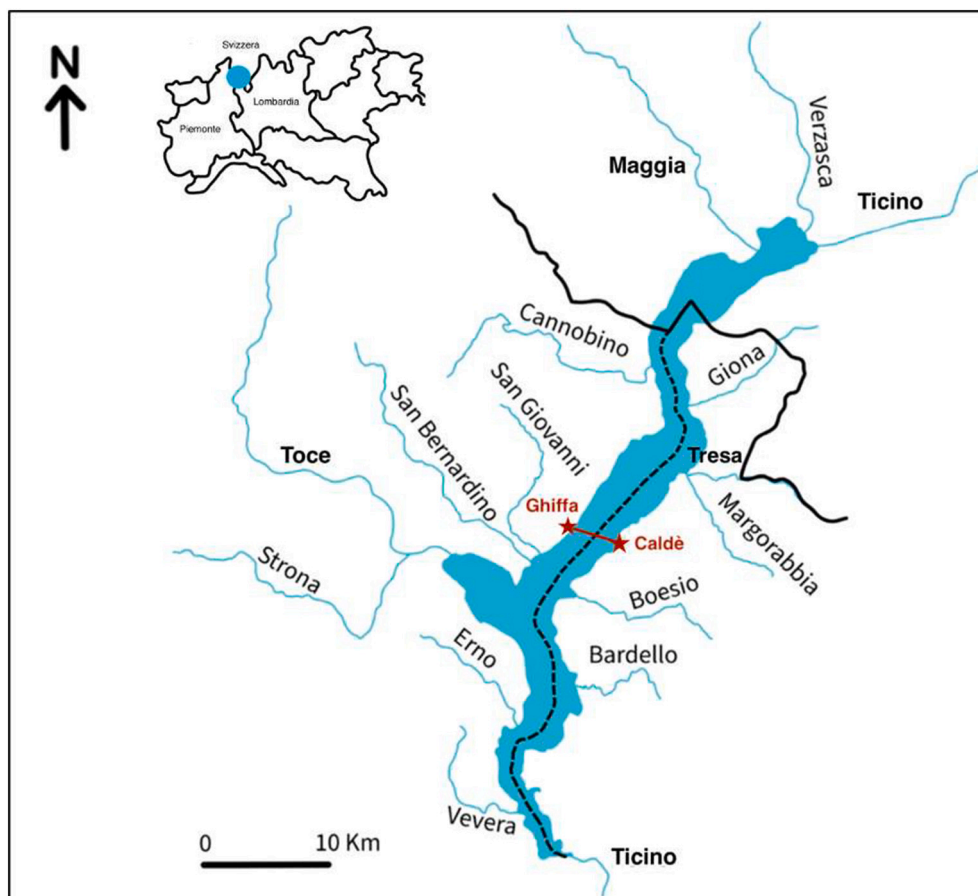


Fig. 1. Lake Maggiore with the position of the sampling transect (redline).

regenerated cellulose-based fibers and fragments (23 %; Table S3), while 60 remained unknown (7 %). The inability to define their composition was attributed to plastic weathering and the presence of interfering material on their surface, which could not be eliminated even through digestion with  $H_2O_2$ .

### 3.1. Quantitative evaluation

Fig. 2 illustrates the concentration of plastics detected in each month of 2022, presented in both plastics/ $m^3$  (Fig. 2A) and plastics/ $km^2$  (Fig. 2B), with emphasis on the two mean annual values. The minimum value of 0.02 plastics/ $m^3$  was recorded in September, corresponding to approximately 4375 plastics/ $km^2$ , considering the water flow sampled by the Manta-net along the transect. In contrast, the maximum value of 0.29 plastics/ $m^3$  was observed in December, equivalent to 57,692 plastics/ $km^2$ , representing approximately 13 times higher than the value measured in September. The notably heterogeneous trend observed throughout 2022 is summarized by the mean annual value of  $0.16 \pm 0.08$  plastics/ $m^3$  ( $33,621 \pm 16,008$  plastics/ $km^2$ ) and the corresponding high standard deviation.

### 3.2. Qualitative characterization

From a qualitative point of view, we observed a heterogeneous situation, detecting the majority of microplastics ( $\leq 1$  mm) and large microplastics (between 1 mm and  $\leq 5$  mm), which together represented approximately 86 % of the total debris in September, up to 100 % in April and June (Fig. 3). Delving into further detail, large microplastics were the most representative size in 8 out of the 12 months, with values ranging from 47 % (January) to as high as 80 % (August). On the other hand, microplastics prevailed in only 3 months, fluctuating between 56 % (February) and 78 % (March). In September, the two dimensions were equivalent, each accounting for 43 %. Lastly, macroplastics ( $>5$  mm) were always detected, except in April and June, with percentages ranging from 2 % (March) to 14 % (September). Shifting focus to plastic shape, fibers and fragments constituted the majority of the plastic litter

detected in 2022. Specifically, the percentage of fibers ranged from 19 % in the May transect to 96 % in the November transect, resulting in an annual average of approximately 58 % of the plastic debris' shape. Conversely, fragments ranged from a minimum of 4 % (November) to a maximum of 75 % (May), yielding a mean annual value of 41 %. Pellets and lines were detected at negligible percentages, not surpassing 8 % (Fig. 4). Polymer characterization once again highlighted diverse contamination levels among the monthly samplings. As depicted in Fig. 5, three polymers predominated in the plastic debris sampled in Lake Maggiore in 2022: PEST, polypropylene (PP), and polyethylene (PE), accounting for mean annual percentages of 41.5 %, 31.7 %, and 20.5 %, respectively. Other polymers did not exceed 7 %. In more detail, the percentage of PEST varied between complete absence (May) and 91 % (November), PP ranged from 1 % (November) to 60 % (May), and PE exhibited percentages ranging from 4 % (October and November) to 47 %, observed in January (Fig. 5). The most frequently detected colors in the plastic debris were black, red, and transparent (Fig. 6). Black varied from 6 % (August) to 29 % (February, May, June, September, December), red particles were detected at percentages ranging from 5 % (March) to 23 % (January), and transparent debris were observed in percentages varying between 12 % (July) and 54 % (August).

## 4. Discussion

Before delving into the obtained results and their comparison with previous data from other lakes worldwide, some general considerations are essential. Firstly, the decision to characterize all the particles selected through visual sorting for subsequent instrumental analysis was crucial since up to 43 % of the debris was identified as *non-plastics* (in September; Fig. 7). These materials were mainly cellulose fibers or regenerated cellulose-based fibers and fragments (Table S3), which we do not classify as plastics. Additionally, it is not possible to distinguish between them using infrared techniques. This once again emphasizes that visual recognition alone or reliance on *non-instrumental* methods is not sufficiently robust to allow an accurate assessment of plastic contamination. Such approaches tend to overestimate, sometimes significantly, the actual pollution caused by synthetic polymeric debris. Consequently, since our results provide an authentic depiction of plastics' contamination, analyzing all 845 selected particles individually, the findings underscore the highly heterogeneous nature of plastic contamination in Lake Maggiore with monthly densities that exhibited variations exceeding an order of magnitude (Fig. 2A, B). The logical consequence of this heterogeneity, despite the intrinsic challenges associated with sampling and the time-consuming nature of a complete instrumental analysis, is the need for more than a single sampling to ensure accurate Environmental Risk Assessment (ERA). This is particularly crucial in aquatic ecosystems with complex basin catchments or those undergoing seasonal changes in anthropic impact that can influence the release of plastics. In our specific case, we could have recorded negligible plastic contamination if we had conducted sampling solely in September (0.02 plastics/ $m^3$ ), or conversely, a significantly higher level of plastic pollution would have been observed if we had sampled the lake only in December (0.29 plastics/ $m^3$ ). This poses a significant challenge that will need to be addressed in future guidelines or legislation mandating the monitoring of synthetic polymeric debris in surface water bodies. A potential solution could involve pre-evaluating the optimal periods for conducting samplings (e.g. worst-case scenarios, average conditions, seasonal variations). Alternatively, a more comprehensive approach would be to conduct studies involving more frequent sampling to ascertain whether a certain category of lakes or rivers exhibits similarity in contamination patterns, thereby determining the most suitable frequency and seasonality for routine monitoring. For example, it would be interesting to replicate this study in other large Italian subalpine lakes (Lake Como, Lake Iseo, Lake Garda) to assess whether plastic contamination varies similarly in aquatic environments with comparable hydrological characteristics and anthropic

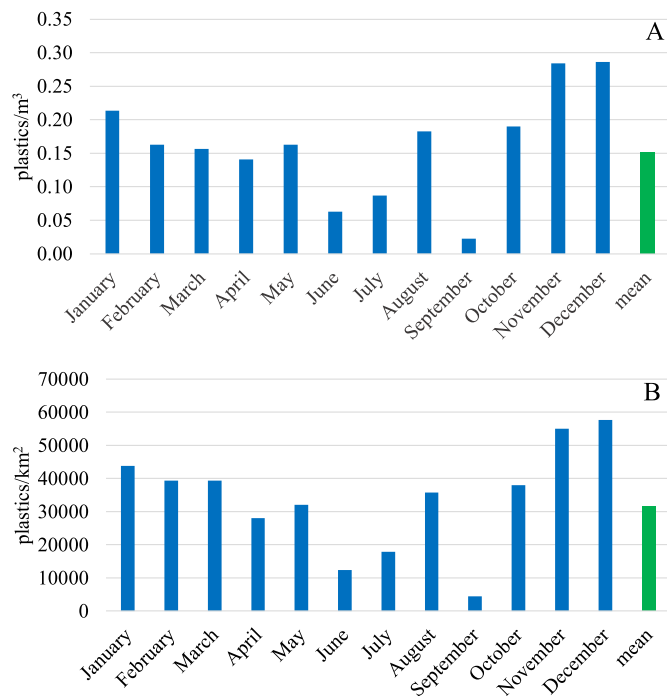


Fig. 2. Monthly concentration of plastics measured along a transverse transect in the L. Maggiore expressed as plastics/ $m^3$  (A) and plastics/ $km^2$  (B). The corresponding annual mean values are also reported.

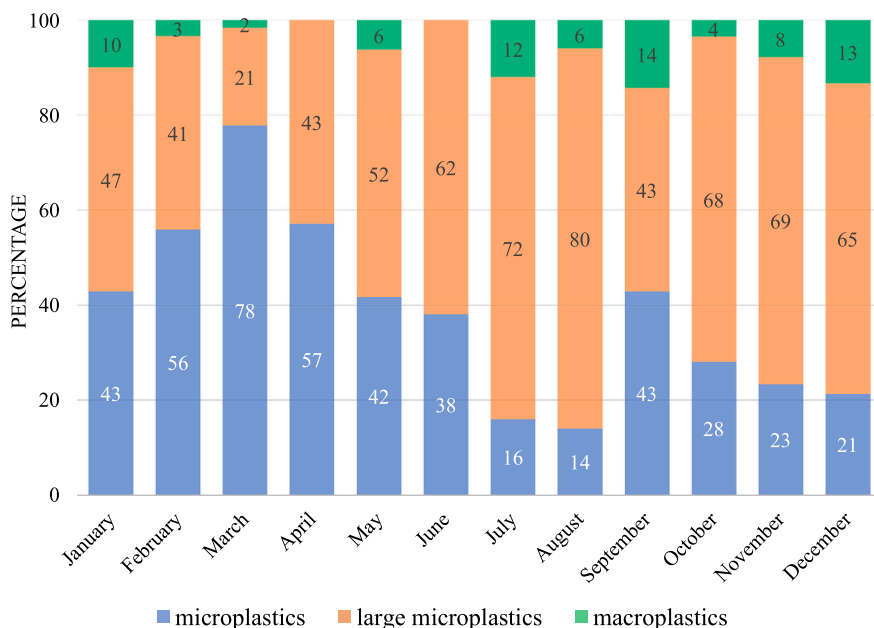


Fig. 3. Percentage of plastic size monitored during the monthly samplings in L. Maggiore. We used the size definition suggested by ISO (ISO/TR21960; 2020): macroplastics >5 mm; 1 mm < large microplastics ≤5 mm; 1 μm < microplastics ≤1 mm.

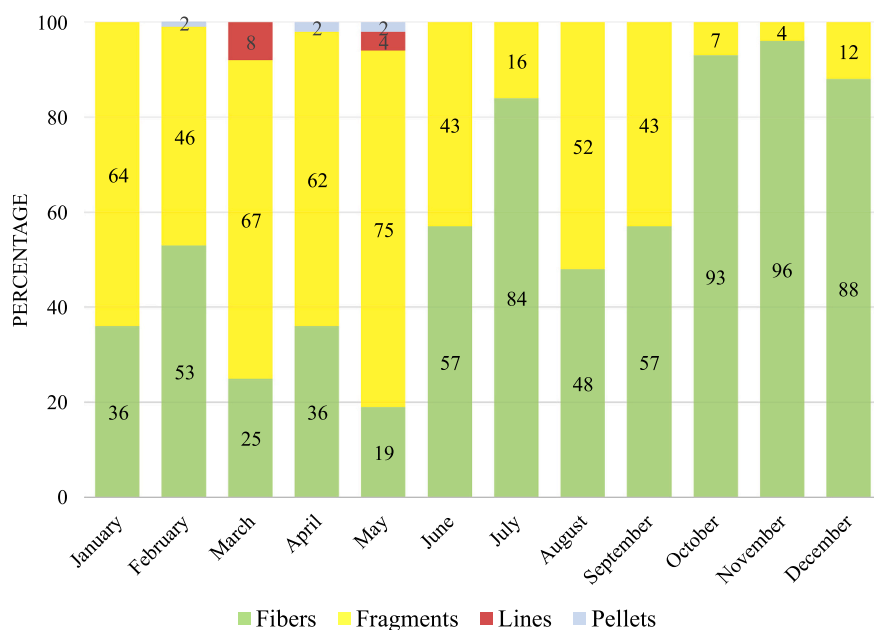


Fig. 4. Percentage of plastic shapes observed in the monthly samplings of L. Maggiore.

impacts. This approach would facilitate the establishment of a unified sampling program for this category of lakes, streamlining comparisons and enhancing environmental management. Hence, we can attempt to apply these concepts to the results obtained in this study, proposing an appropriate sampling plan for future routine monitoring campaigns. An effective approach for managing the extensive dataset acquired through monthly sampling is to aggregate the data seasonally (Fig. 8). The two seasons during which the highest quantity of floating plastics was detected were autumn (mean of  $50,230.8 \pm 10,677.4$  plastics/km<sup>2</sup>) and winter (mean of  $40,819.4 \pm 2538.0$  plastics/km<sup>2</sup>), while values approximately half as low were recorded in spring (mean of  $24,117.6 \pm 10,383.0$  plastics/km<sup>2</sup>), and especially in summer, where a mean of  $19,315.5 \pm 15,720.5$  plastics/km<sup>2</sup> was observed (Fig. 8A). These

similarities and differences are corroborated by the fact that summer did not exhibit significant differences ( $p > 0.05$ ) when compared to spring data, as also noted between autumn and winter, which, conversely, are both significantly different ( $p < 0.05$ ) from the concentration of plastics measured in summer (Fig. 8A). The declining contamination trend observed during the spring and summer months (Fig. 2) might be attributed to potential interference caused by the formation of biofilm on plastic particles, leading to an increased sedimentation rate and consequently a reduction in the density of sampled floating plastics (Jalón-Rojas et al., 2022). To explore this potential interfering factor, we examined correlations between monthly plastic concentration and data related to phytoplankton analyzed during the same months at Ghiffa (data kindly provided by Dr. Martina Austoni, IRSA-CNR), representing



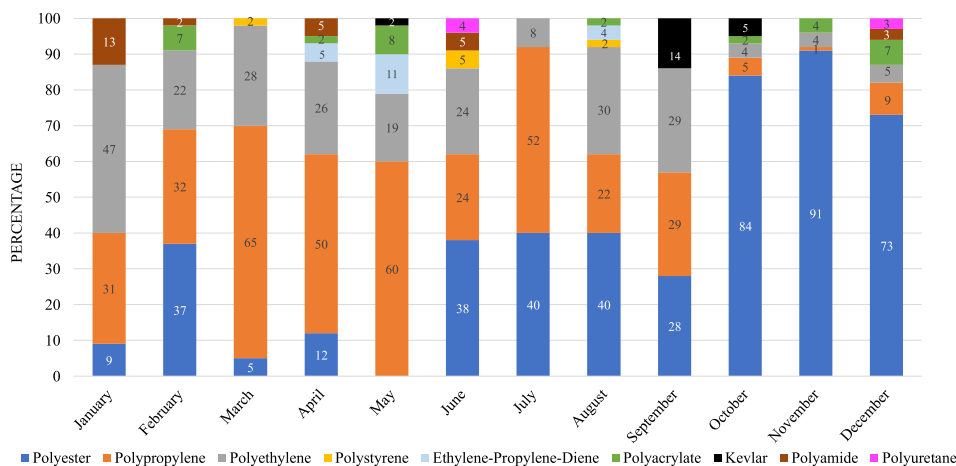


Fig. 5. Percentage of polymeric composition of plastics monthly samplings in L. Maggiore.

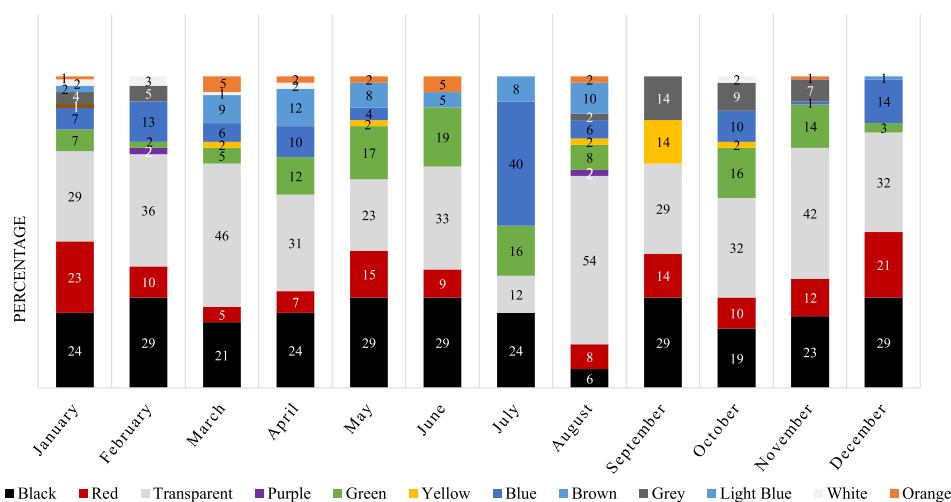


Fig. 6. Percentage of plastic colours monthly samplings in L. Maggiore.

one of the two extreme points of our transverse transects (Fig. 1). No correlation was identified between plastic concentration and phytoplankton density ( $r = -0.5168$ ;  $p = 0.085$ ), phytoplankton biomass ( $r = -0.0309$ ;  $p = 0.928$ ), and cyanobacteria density ( $r = -0.4629$ ;  $p = 0.152$ ). Thus, these rules out the possibility of biofilm interference in the sampling conducted during the spring-summer months. To investigate the main source of contamination (whether primary or secondary), it is necessary to provide a preliminary remark. Our transverse transect was positioned in the central area of the lake (Fig. 1) to enable a comparison of results with findings from two previous monitoring campaigns (Sighicelli et al., 2018; Binelli et al., 2020). Given that the primary flow in Lake Maggiore is in the north-south direction, disregarding any minor coastal currents and internal seiches, we predominantly sampled plastics originating from rivers and wastewater treatment plants (WWTPs) situated in the central-northern part of the lake. In detail, 25 WWTPs discharge their wastewater into the tributaries of Lake Maggiore, serving approximately 463,982 equivalent inhabitants. Among these, seven are located north of the sampled transect, catering to almost 36 % of the population (approximately 167,000 equivalent inhabitants). Therefore, our data can provide insight into the origin of plastic contamination only for the central-northern area of the lake. First of all, the almost complete absence of pellets (Fig. 4), which are characteristic indicators of a primary origin of plastics, is particularly informative. On the contrary, fibers and fragments, typical indicators of secondary contamination, were the main shape detected, with seasonal variations favoring the former in

summer and autumn, and the latter in winter and spring (Fig. 8B).

In more detail, we established a highly significant ( $r = 0.8309$ ;  $p = 0.001$ ) and positive correlation between plastic concentrations and the quantity of fibers detected seasonally. Conversely, no correlation was found between the seasonal concentrations of plastics and the fragments identified in the samples ( $r = 0.1320$ ;  $p = 0.683$ ). This implies that fibers serve as the primary indicators for plastic contamination in Lake Maggiore, likely originating from atmospheric transport (Dris et al., 2017; Zhang et al., 2020), discharge from textile industries present in the watershed (Guzzella et al., 2008; Poma et al., 2014), and the washing of synthetic clothes (Napper and Thompson, 2016; De Falco et al., 2018; Wang et al., 2023), which is more prevalent in cold seasons when the use of such garments is higher (Browne et al., 2011). The likely source of plastics from the washing of synthetic clothes leads to another consequence related to the impact of tourism, which results in an approximate 60 % increase in the population, especially during spring and summer (refer to paragraph 2.1). This impact is only noticeable in August, where we observed a more than double increase in the concentration of plastics compared to July and approximately eight times higher than in September (Fig. 2). The mixed secondary origin of plastic pollution is further confirmed by the absence of any significant correlation with environmental parameters that could impact the release of plastics into the lake. Specifically, not only did we fail to identify a correlation between the concentration of plastics and monthly rainfall in the basin ( $r = 0.355$ ;  $p = 0.258$ ), but also when considering rainfall measured 72 h

before samplings ( $r = -3723$ ;  $p = 0.233$ ) and in the previous 7 days ( $r = 0.0324$ ;  $p = 0.920$ ). From an ecotoxicological perspective, it is crucial to assess the size of the detected plastic particles, as microplastics, along with nanoplastics, are the fractions that can more easily penetrate the tissues of organisms, resulting in several adverse effects. The heterogeneity of the annual contamination is also confirmed by Fig. 8C, illustrating that microplastics were the predominant category only in winter, accounting for 58 %, compared to 36 % for large microplastics and just over 5 % for macroplastics. In the other three seasons, large microplastics were predominantly found, ranging from 50 % in spring to 74 % in summer. The seasonal results obtained for the different polymers constituting fibers and fragments in the transverse transects are particularly surprising. As depicted in Fig. 8D, the polymeric percentage composition remains similar in winter, spring, and, to a lesser extent, even in summer. However, a significant change occurs in autumn. Specifically, while the first three seasons predominantly feature PE and PP debris, constituting over 55 % of the total polymeric composition (Fig. 8D), autumn presents almost exclusively PEST fibers (Fig. 8D). While the latter serves as a typical marker for plastic sources derived from clothes washing, textile industries and atmospheric transport, the origin of PE and PP particles is more diverse. PE is widely utilized in numerous objects for both common and technical purposes, such as old shopping bags, protective packaging for transporting fragile items (Pluriball), food films, water and gas pipes, toys. On the other hand, PP is used for packaging in the food industry, sportswear, carpets, plates, toys, and suitcases. The reason for the dramatic change in polymer composition, particularly with regard to the fibers (Table S2), observed in all three autumn months is currently unknown. It would certainly be interesting to investigate this phenomenon in future studies.

Despite the significant variation in plastic contamination observed over the 12 months, it does not seem sufficient to provide a comprehensive picture of lake pollution. However, our data can suggest, at least, the season that may be most representative and informative about the characteristics and origins of plastics. Based on the findings of this study, we recommend winter as the optimal season for a potential single sampling in Lake Maggiore for routine plastic pollution campaigns. Primarily, winter was the only season during which we observed greater quantitative homogeneity over the three sampled months, as indicated by the very low standard deviation (Fig. 8A). In contrast, in the other three seasons, notable differences were observed for certain months (June, September, October) compared to the other two within the same season (Fig. 2A, B). This suggests that the choice of the winter month for a single sampling may not be critical, while in other seasons, there is a risk of significant underestimation or overestimation of plastic pollution. Furthermore, even though it may not represent the worst

contamination scenario, we contend that sampling during the winter months provides a more representative view of the annual contamination trend. Despite a non-statistical difference in quantitative terms of approximately 20 % compared to the highest contamination observed in autumn (Fig. 8A), we believe that the qualitative data offers a more comprehensive perspective for Ecological Risk Assessment (ERA) and subsequent risk management. Specifically, winter is the season in which we observed greater homogeneity in plastic shapes, with percentages of synthetic fragments and fibers being more similar compared to the other seasons (Fig. 8B). If the characteristic related to plastic size does not serve as a discriminator, as all three categories of plastics were detected in each of the four seasons (Fig. 8C), we believe that the observed variation in the polymer component is crucial for determining the optimal season for sampling. In autumn, the dominant polymer was PEST, constituting 83 % of the total, while the other six polymers were detected with percentages  $\leq 5$  %. In contrast, during winter, the polymer percentages were more uniform, and, most importantly, the contamination footprint was much more similar to that observed in spring and summer (Fig. 8D). In conclusion, while it would be ideal to conduct at least four samplings, one per season, to obtain the most comprehensive understanding of plastic contamination in L. Maggiore, if environmental management and control agencies will opt for a single annual sampling for routine monitoring, we recommend conducting it during the winter months for the reasons outlined above.

#### 4.1. Comparisons with other lakes worldwide

Comparing plastic contamination levels among lakes worldwide is challenging due to the absence of guidelines standardizing the various variables that can alter plastic densities. Consequently, such comparisons can be easily distorted. The primary parameters influencing the monitoring of these physical contaminants include the sampling method (net, sieves, or pump) and type (one or few single points, or trawl), mesh of nets, plastic separation methodology, size classification, and the analysis system used to discriminate natural particles from those made of synthetic polymers. The results obtained from our study have introduced an additional variable linked to the sampling period, which, in our case, influenced plastics' density by more than one order of magnitude (Fig. 2). Therefore, it is crucial to specify all these parameters, some of which are not reported or clearly described in certain papers, rendering any comparison with other aquatic environments impossible. Neglecting to account for all these variables, Table 1 displays the most recent monitoring data detected in numerous lakes worldwide, categorized by continents. It places particular emphasis on data available for the large Italian subalpine lakes, which, owing to their

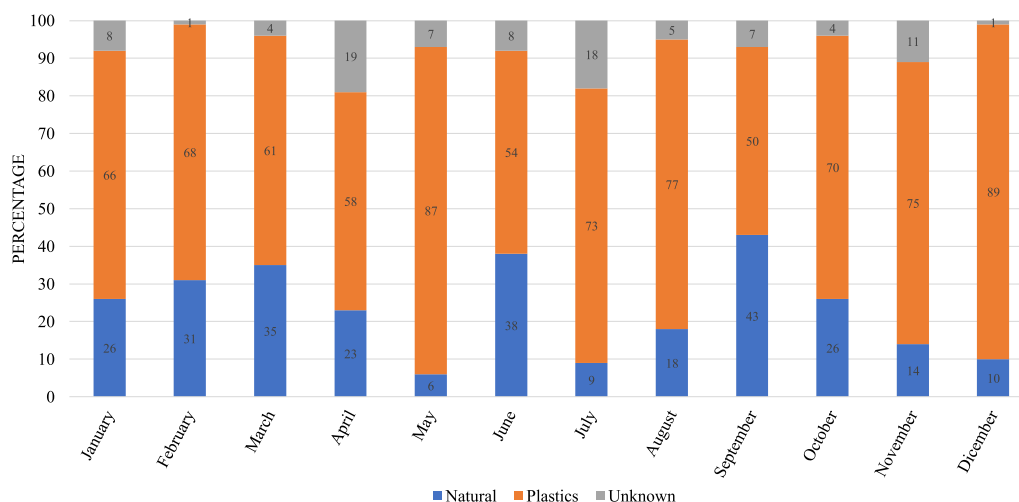
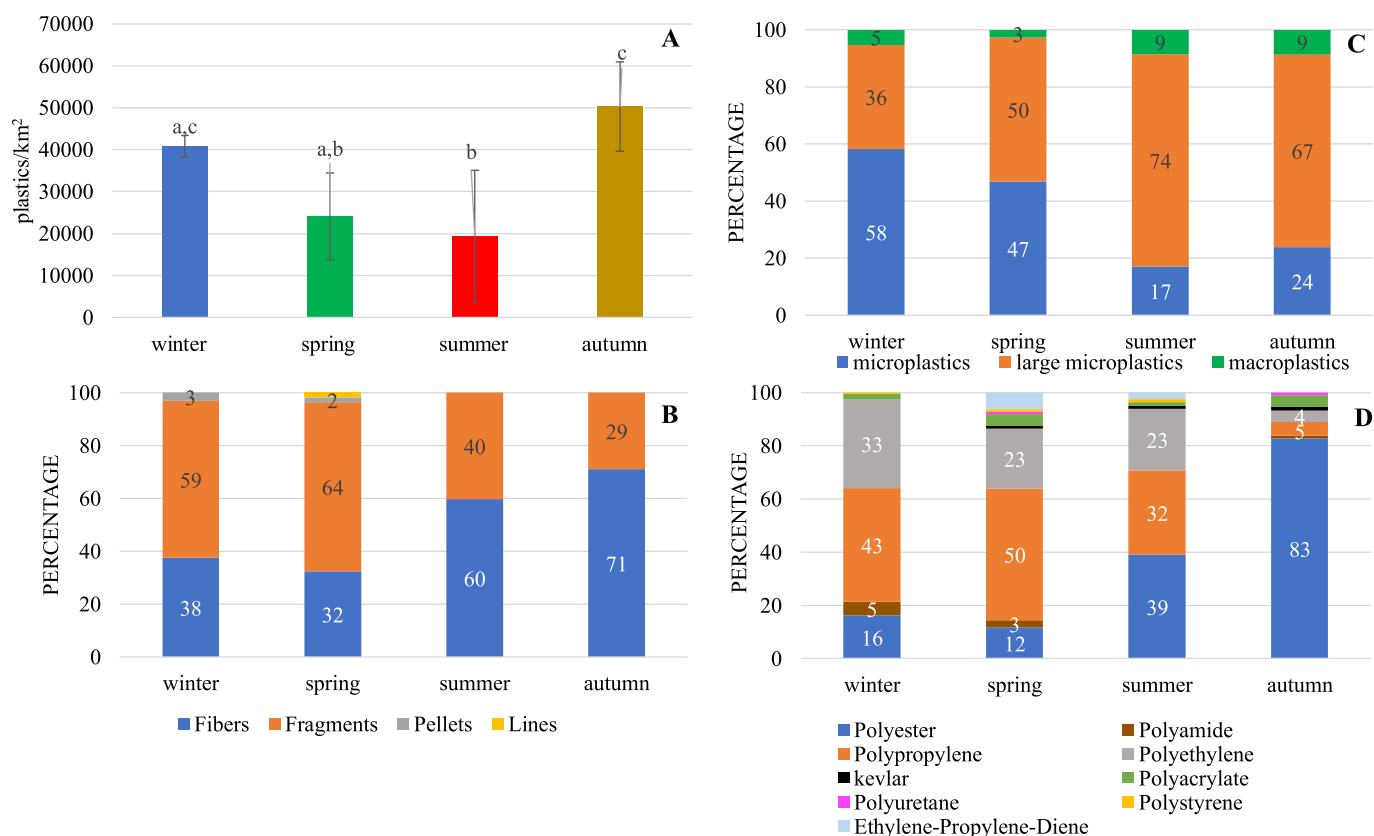


Fig. 7. Monthly percentages of natural, plastic and unknown particles evaluated by  $\mu$ FT-IR in L. Maggiore.



**Fig. 8.** Seasonal trends in plastic levels (mean  $\pm$  standard deviation); (A), percentage of shape (B), size (C) and polymeric composition (D) of plastic particles measured in L. Maggiore in 2022. In panel A the same letter shows the absence of a significant difference ( $p > 0.05$ ).

hydro-morphological characteristics and anthropic presence, bear resemblance to Lake Maggiore. The contamination identified in Lake Maggiore through monthly sampling in 2022 consistently exhibits lower or comparable plastic concentrations compared to most other sampled lakes (Table 1). Among the considered data, the Asian continent appears to be the most affected by plastic pollution, with 5000–34,000 plastics/m<sup>3</sup> reported in Lake Poyang (China; Yuan et al., 2019), and an average density of  $291 \pm 252$  plastics/m<sup>3</sup> in 7 sampling points in Lake Baikal (Siberia, Russia; Moore et al., 2022). These values are approximately more than 6 and 4 orders of magnitude higher than the highest data recorded in Lake Maggiore ( $0.29$  plastics/m<sup>3</sup> in December 2022). Several lakes in South America also exhibit higher pollution levels than those observed in Lake Maggiore: Alfonso et al. (2020a) measured a plastic concentration of  $143.3 \pm 40.4$  plastics/m<sup>3</sup> in Lake La Salada (Argentina), approximately 500 times higher than our worst case, while Lake Vintter (Argentina; Alfonso et al., 2020b) demonstrated a contamination more than 6 times higher. Contamination levels found in North America seem to be closer to those detected in Lake Maggiore; for instance, Lake Erie reported a value ( $45,000$  plastics/km<sup>2</sup>) precisely comparable to the highest levels measured in some months of our sampling, while Lake Ontario (Canada; Mason et al., 2020) and Lake Flathead (USA; Xiong et al., 2022) exhibited contamination 4 and 3 times greater than the highest level recorded in Lake Maggiore, respectively. In the European context, the highest contamination levels recorded in Lake Maggiore are approximately 40 times lower than the concentration measured in Lake Lugano ( $11.5$  plastics/m<sup>3</sup>; Nava et al., 2023) and even over  $6 \times 10^3$  times lower than the alarming contamination level ( $1812$  plastics/m<sup>3</sup>) identified in Lake Tollense (Germany). It is essential to note that Lake Tollense is the only one where the Nile Red dye was utilized to characterize polymeric particles, with only 2 % of them verified by  $\mu$ -Raman (Tamminga et al., 2022). Conversely, the contamination identified in Lake Maggiore aligns well with the levels

( $0.27 \pm 0.18$  plastics/m<sup>3</sup>) recently reported in Lake Kallavesi (Finland; Uurasjärvi et al., 2020). The low plastic contamination observed in Lake Maggiore is corroborated by data collected both within the same lake basin and in the other large Italian subalpine lakes. Notably, two previous monitoring campaigns conducted in Lake Maggiore reported a density of polymer particles that is directly comparable to the maximum value recorded in 2022 by our study (Table 1). These values ranged between  $29,000 \pm 17,000$  plastics/km<sup>2</sup> and  $45,000 \pm 13,000$  plastics/km<sup>2</sup> (Sighicelli et al., 2018) and an average of  $100,000 \pm 35,000$  plastics/km<sup>2</sup> (Binelli et al., 2020; Galafassi et al., 2021). Our findings align with those obtained for other large subalpine lakes within the Po River catchment basin (Table 1). The only data contradicting this trend is presented by Nava et al. (2023), where a value of  $8.24$  plastics/m<sup>3</sup> was recorded in Lake Maggiore in 2021. This concentration is almost 30 times higher than our maximum density detected in 2022, but over 400 times greater than our minimum value (Table 1). Considering this data anomaly, especially given that the sampled area is the same as that of Sighicelli et al. (2018), it is plausible to hypothesize an overestimation in the average plastic concentration in Lake Maggiore, likely attributed to the chosen sampling period. This elevated contamination level might depict the aftermath of the extraordinary flood event in three major tributaries of Lake Maggiore (Rivers Ticino, Maggia, and Toce) on 2 and 3 October 2020. This event led to an exceptional increase in the lake hydrometric level by 2.18 m in just 37 h and could have resulted in the transport of a substantial quantity of plastic materials from the basin. This further underscores the importance of considering the temporal parameter as a potential variable in lake basin contamination. It emphasizes the necessity for increasing the frequency of samplings to enable better comparisons across different aquatic ecosystems and to provide more accurate data for environmental management agencies.



**Table 1**  
Most recent monitoring data of plastic pollution in lakes worldwide.

Lake	Country	Sampling type	Sampling method	Mesh ( $\mu\text{m}$ )	Plastic concentration	Characterization method	References
<i>Americas</i>							
La Salada	Argentina	One point	Net	38	$143.3 \pm 40.4$ plastics/ $\text{m}^3$	Visual	Alfonso et al., 2020a
Vintter	Argentina	Trawl	Net	38	$1.9$ plastics/ $\text{m}^3$	Raman (15 % of particles)	Alfonso et al., 2020b
Erie	Canada	Trawl	Manta-net	355	$45,000$ plastics/ $\text{km}^2$ (mean)	FTIR	Mason et al., 2020
Ontario	Canada	Trawl	Manta-net	355	$230,000$ plastics/ $\text{km}^2$ (mean)	FTIR	Mason et al., 2020
Flathead	USA	Trawl ( $n = 12$ )	Manta-net	330	$189,000$ plastics/ $\text{km}^2$ (mean)	Raman (12.5 % of particles)	Xiong et al., 2022
<i>Asia</i>							
Hong	China	One point	Net	74	$1800$ plastics/ $\text{m}^3$	Raman (total particles)	Li et al., 2019
Poyang	China	Some points	Sieve	50	$5000\text{--}34,000$ plastics/ $\text{m}^3$	Raman (100 particles)	Yuan et al., 2019
Hovsgol	Mongolia	Trawl ( $n = 9$ )	Manta-net	333	$20,264$ plastics/ $\text{km}^2$ (mean)	optical microscope	Free et al., 2014
Rawal	Pakistan	Trawl ( $n = 3$ )	Net	100	from $6.4 \pm 0.5$ to $8.8 \pm 0.5$ plastics/ $\text{m}^3$	FTIR (total particles)	Bashir and Hashmi, 2022
Baikal	Russia	7 points	Pump+net	20	$291 \pm 252$ plastics/ $\text{m}^3$ (mean)	$\mu\text{FTIR}$ ( $\sim 30$ % filter area)	Moore et al., 2022
<i>Africa</i>							
Victoria	Tanzania	Trawl ( $n = 8$ )	Manta-net	300	$0.73$ plastics/ $\text{m}^3$ (mean) $120,588$ plastics/ $\text{km}^2$ (mean)	FTIR ( $\sim 20$ % of particles)	Egessa et al., 2020
<i>Europe</i>							
Kallavesi	Finland	Trawl	Manta-net	333	$0.27 \pm 0.18$ plastics/ $\text{m}^3$	FTIR (total particles)	Uurasjärvi et al., 2020
Lugano	Switzerland	Trawl (3 replicates)	Manta-net	300	$11.50$ plastics/ $\text{m}^3$	Raman ( $\sim 25$ % of particles)	Nava et al., 2023
Tisza	Hungary	One point	Pump	100	$23.12$ plastics/ $\text{m}^3$	FTIR (total particles)	Bordós et al., 2019
Tollense	Germany	One point	Pump/ filtration	20	$1812$ plastics/ $\text{m}^3$	Nile Red/ $\mu\text{Raman}$ ( $\sim 2$ % of particles)	Tammenga et al., 2022
<i>Italy</i>							
Garda	Italy	Trawl ( $n = 3$ )	Manta-net	300	From $4000 \pm 2700$ plastics/ $\text{km}^2$ to $55,000 \pm 29,000$ plastics/ $\text{km}^2$	FTIR (total particles)	Sighicelli et al., 2018
Iseo	Italy	Trawl ( $n = 3$ )	Manta-net	300	From $15,000 \pm 11,000$ plastics/ $\text{km}^2$ to $57,000 \pm 36,000$ plastics/ $\text{km}^2$	FTIR (total particles)	Sighicelli et al., 2018
Maggiore	Italy	Trawl ( $n = 3$ )	Manta-net	300	From $29,000 \pm 17,000$ plastics/ $\text{km}^2$ to $45,000 \pm 13,000$ plastics/ $\text{km}^2$	FTIR (total particles)	Sighicelli et al., 2018
Garda	Italy	Trawl ( $n = 7$ )	Manta-net	300	$36,000 \pm 28,000$ plastics/ $\text{km}^2$ (mean)	FTIR (total particles)	Galafassi et al., 2021
Orta	Italy	Trawl ( $n = \text{n.a.}$ )	Manta-net	300	$63,000 \pm 25,000$ plastics/ $\text{km}^2$ (mean)	FTIR (total particles)	Galafassi et al., 2021
Como	Italy	Trawl ( $n = 5$ )	Manta-net	300	$29,000 \pm 14,000$ plastics/ $\text{km}^2$ (mean)	FTIR (total particles)	Galafassi et al., 2021
Maggiore	Italy	Trawl ( $n = 6$ )	Manta-net	300	$100,000 \pm 35,000$ plastics/ $\text{km}^2$ (mean)	FTIR (total particles)	Galafassi et al., 2021
Maggiore	Italy	Trawl (3 replicates)	Net	200	$8.24$ plastics/ $\text{m}^3$	Raman ( $\sim 25$ % of particles)	Nava et al., 2023
Maggiore	Italy	Trawl	Manta-net	100	From $0.02$ to $0.29$ plastics/ $\text{m}^3$ from $4375$ to $57,692$ plastics/ $\text{km}^2$	$\mu\text{FTIR}$ (total particles)	Present study

“Total particles” indicates the instrumental analysis of the particles selected through visual sorting.

## 5. Conclusions

To the best of our knowledge, this study represents the first investigation into plastic monitoring in a lake through monthly samplings spanning an entire year. Our data illustrates the marked heterogeneity of plastic contamination in Lake Maggiore, although the overall levels remain comparatively low when compared with recent studies. The origin of plastics released at least in the central-northern part of the catchment basin appears to be predominantly secondary. Conversely, we can rule out a significant primary source, as evidenced by the negligible percentage of pellets detected, which typically serve as markers for such contamination.

Our observations are largely centered on the identification of PEST fibers, released from the washing of synthetic clothing, textile industries, and atmospheric transport, or the presence of PE and PP particles, indicative of plastic object fragmentation released directly into the lake or originating from the catchment basin. Consequently, interventions to

mitigate the presence of plastics in Lake Maggiore should primarily target these sources. From a broader perspective, our monthly sampling has unveiled an additional variable in the assessment of plastic pollution in aquatic environments. Not only did we observe a disparity of over an order of magnitude between the maximum and minimum plastic density recorded throughout the year, but we also discerned significant qualitative discrepancies from one month to another. This underscores the inadequacy of a single sampling, especially in expansive lakes with intricate catchment basins like Lake Maggiore, for obtaining a comprehensive understanding of the pollution caused by these emerging contaminants. Such nuanced insights are crucial not only for effective environmental management but also for furnishing precise information to an increasingly vigilant and conscientious civil society.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2024.170740>.

## CRedit authorship contribution statement

**Andrea Binelli:** Writing – original draft, Visualization, Supervision, Resources, Project administration, Funding acquisition, Conceptualization. **Stefano Magni:** Writing – original draft, Visualization, Supervision, Methodology, Investigation, Data curation, Conceptualization. **Camilla Della Torre:** Writing – review & editing, Methodology, Investigation. **Riccardo Sbarberi:** Validation, Formal analysis. **Cristina Cremonesi:** Formal analysis, Data curation. **Silvia Galafassi:** Writing – review & editing, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Andrea Binelli and Silvia Galafassi reports financial support was provided by CIP AIS (International Commission for the Protection of Italian-Swiss Waters). If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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