Microplastics in surface water of Laguna de Bay: First documented evidence on the largest lake in the Philippines

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Abstract

The pollution of aquatic systems by microplastics is a well-known environmental problem. However, limited studies have been conducted in freshwater systems, especially in the Philippines. Here we determined for the first time the amount of microplastics in the Philippines’ largest freshwater lake, the Laguna de Bay. Ten (10) sampling stations on the lake's surface water were sampled using a plankton net. Samples were extracted and analyzed using Fourier transformed infrared spectroscopy (FTIR). A total of 100 microplastics were identified from 10 sites with a mean density of 14.29 items/m$^3$. The majority of microplastics were fibers (57%), while blue-colored microplastics predominated in the sampling areas (53%). There were 11 microplastic polymers identified predominantly polypropylene (PP), ethylene vinyl acetate copolymer (EVA), and polyethylene terephthalate (PET), which together account for 65% of the total microplastics in the areas. The results show that there is a higher microplastic density in areas with high relative population density which necessitates the implementation of proper plastic waste management measures in the communities operating on the lake and in its vicinity to protect the lake's ecosystem services. Furthermore, future research should also focus on the environmental risks posed by these microplastics, especially on the fisheries and aquatic resources.

Introduction

The United Nations Environment Program considers plastic pollution to be a significant environmental problem. It has been identified as an emerging issue that may impact biological diversity and human health alongside climate change (Blettler et al., 2017). The number of data in the Philippines on plastic pollution in freshwaters is limited when compared to studies on marine ecosystems, even though pollution in both ecosystems is comparable (Peng et al., 2017; Superio and Abreo, 2020; Inocente and Bacosa, 2022; Sajorne et al., 2022a). Requiron and Bacosa (2022) studied macroplastics in Pulauan River, Dapitan City where a total of 1,636 macroplastics items were identified for 10 days of observation. Furthermore, in the same study, a total of 996 plastic litter were also observed on the riverbank. This evidence is concerning because pollution levels in rivers, streams, and lakes are almost identical to those found in the sea (Peng et al., 2017).

One of the emerging issues concerning plastics nowadays is microplastics. Microplastics are minute plastics usually less than 5 mm in size that could spread harmful substances, including toxins and polycyclic aromatic hydrocarbons (Abreo, 2018). These microplastics can also be classified further into primary and secondary microplastic. Primary microplastics are the ones that are intentionally reduced and synthesized into smaller pieces for commercial uses, while secondary microplastics are the ones that are environmentally degraded from plastics (Xu et al., 2020). Microplastic abundance has been discovered in various environments ranging from freshwater to the poles (Isobe at al., 2017). However, there is a scarcity of data on microplastic research in freshwater, particularly in lakes and reservoirs (Ramadan and Sembiring, 2020).
Evidence of microplastics was observed to be floating in the surface waters controlled by the currents (Ivar do Sul et al., 2014). In a study conducted in China's Yellow River, microplastic fibers < 200µm were dominant in its surface water, accounting for roughly three-fourths of the microplastics, and were mostly polyethylene (PET), polypropylene (PP), and polysterene (PS) in composition (Han, et al., 2020). Another study conducted in surface water from eastern coastal areas of Guangdong, South China showed mean abundance of microplastics of 8895 items/m³, with small white fragments dominating character (Zhang et al., 2020). In the Philippines, a study conducted in Molawin Creek in Makiling Forest Reserves indicated that it is also being polluted by these micropollutants, which are primarily coming from the area's residential, commercial, and university facilities, causing an intrusion into the ecological services provided by the watersheds (Limbago, et al., 2021). Furthermore, microplastic fragments measuring < 2.5mm in length were microplastic observed in the Pasig River (Deocaris et al., 2019). Pasig River flows through the urban areas from its upstream portion in Laguna de Bay (Deocaris et al., 2019). However, the amount of microplastics from Laguna de Bay to Pasig River remained to be ascertained. Laguna de Bay is the largest lake in the Philippines with an area of 911.7 km², and an economically important body of water that supports approximately 9,000 ha of fish pens and fish cages and provides a source of income for fishers in Laguna and Rizal provinces. It is split into four (4) bays: West Bay, Central Bay, East Bay, and South Bay. Talim Island separates the West and Central Bays. These divisions are caused by significant bathymetric differences between these areas (Delos Reyes, 1995).

The occurrence of microplastics in the environment is recognized as an environmental challenge. Microplastics in the studies conducted in Molawin Creek and Pasig River pose an environmental microplastic pollution threat to Laguna de Bay as the Pasig River flows through its upstream portion (Deocaris et al., 2019). Furthermore, the Laguna Lake watershed area has been subjected to environmental pressures and unjustified exploitation. Despite the significant economic contribution of Laguna de Bay, and the potential occurrence of microplastics in water that could threaten the resources it supports, the amount of this emerging pollutants in this economically important body of water is yet to be determined and quantified. Thus, this study aimed to assess the microplastic pollution in the surface water of Laguna de Bay. We determined the density, color, shape, type of polymer and distribution of microplastics in the lake. To our knowledge this is the first study to document the presence of microplastics in a lake in the Philippines.

**Materials And Methods**

The study was conducted in Laguna Lake, popularly referred to as Laguna de Bay (Fig 1), the largest lake in the Philippines. The lake supports approximately 9,000 ha of fish pens and fish cages and provides a source of income for fishers in Laguna and Rizal provinces.

Sampling was carried out in ten sites (Fig 1). Sampling stations were grouped according to the lake's bathymetric differences. The four groups were South Bay, West Bay, Central Bay, and East Bay. South Bay was composed of Station 1 and Station 7; West Bay was comprised of Station 2, Station 3, and Station 4. Central Bay was composed of Station 5 and Station 6, and East Bay was composed of Station 8, Station
9, and Station 10 (Fig 1). Within each sampling site, water sample was collected yielding 10 samples for microplastic analysis.

**Water sampling**

Microplastics were collected using a plankton net from the surface water of Laguna de Bay, following the methods by Viršek et al. (2016) with some modifications. A plankton net with a mesh size of 20µm was used at each sampling site. The boat traveled 10 m while the net was set at a depth of 20cm to keep the entire net submerged in water. GPS coordinates were also recorded at the start and end to aid in calculating the distance trawled. This formula was used to calculate the volume of water passing through the net:

\[
\text{water volume} = \pi r^2 h
\]

where:

- \( r \) = radius of the plankton net
- \( h \) = distance traveled by the net

**Sampling processing**

In the laboratory, all glassware were washed thoroughly with distilled water for decontamination. Solids collected in the plankton nets were soaked in KOH solution and heated in a 60°C oven for 24 hours to digest organic material found during the water sampling. The sample is then floated to extract the plastic debris using density separation in NaCl at a 30% concentration. Vacuum filtration using Millipore set was performed after floatation. The probable microplastic particles were collected into a clean Whatman glass filter. Each glass filters were washed with distilled water and was oven-dried until dry. A clean Petri dish kept the filter paper for optical microscopy analysis using 40x magnification. The particles’ shape, size, and colors were also measured and identified (Hidalgo-Ruz et. al, 2012).

**Microplastic Identification**

Microscopy

All suspected microplastic particles were mounted on glass slides using a clean needle. A microscope with a magnification of 40x was used to determine the morphological characteristics of the sampled microplastics from various sampling sites through visual inspection. Additionally, a film was characterized as a tiny, very thin coating or a substantial chunk of plastic trash, while a fiber was described as a microplastic with a long, slender appearance. Pellets were described as spherical, rounded microplastic particles. The classification of fragment was used when a microplastic could not be classified as a fiber, pellet, or film. This technique was based on the classification method used by Su et al. (2016). All laboratory activities were done at the Chemistry Department of Caraga State University, Butuan City, Philippines.
Abundance

The density of microplastics was calculated by the total number of microplastic items divided by the total water volume. This method was modified based on the study of Egessa et al. (2020).

\[
\text{Density} = \frac{\text{number of microplastics}}{\text{watervolume (m}^3\text{)}}
\]

Attenuated total reflectance–fourier transform infrared (ATR-FTIR) spectroscopy analysis

FTIR produces an infrared absorption spectrum to identify chemical bonds in a molecule. The spectra generate a sample profile, a unique molecular fingerprint that can be used to screen and scan samples for various components. Furthermore, ATR-FTIR is the most used spectroscopy in identifying microplastic polymers. This was used to determine the plastic-type present in each study sample. Samples were mounted in the Perkin-Elmer Spectrum Two FT-IR Spectrometer, which features a software program called Spectrum Search Plus. This program has five different algorithms, a spectral interpretation expert system, and library search and match functionality. All laboratory activities were done at the Chemistry Department of the Caraga State University, Butuan City, Philippines.

Quality Control

Quality control of this study was done following the methods by Egessa et al, (2020) with some modifications. During microscopy, all the dried samples to be analyzed were kept covered in glass Petri dishes. Background contamination from laboratory sources via the air and laboratory tools and equipment were tested using procedure blanks made from glass filter contained in the Petri dishes and distilled water. At each stage of sample collection and analysis, the Petri dishes were left open to the air. The contents of control Petri dishes were processed and screened for microplastic contamination. The procedural blanks contained no microplastic.

Results And Discussion

Abundance and distribution

Microplastics were recorded in all ten (10) sampling stations, in varying abundance (Table 1). Across all sites, the mean density microplastics in surface water of the lake was 14.29 items/m\(^3\). The density was highest in West Bay comprising of Station 2, Station 3, and Station 4, associated with more intensive anthropogenic activities and lowest in Central Bay comprising of Station 5 and Station 6 areas of the lake with less anthropogenic activities. In West Bay, microplastic abundance ranged from 17.14-24.17 items/m\(^3\), being highest at Station 2 and lowest at Station 3. Central Bay comprising Station 5 and 6 which were associated with fish landing beaches (Bagarinao, 1998) in rural communities, was intermediate, with the density in the range of 7.14 -11.43 items/m\(^3\) being higher at Station 5 and lower at Station 6 (Table 1). These findings are consistent with previous research that has found a high
abundance of microplastics in areas of high anthropogenic activity, such as densely populated cities (Browne et al., 2011), tourist beaches and aquaculture (Laglbauer et al., 2014), as well as fishing activities (Dowarah and Devipriya, 2019. In this study, the contribution of sites to the microplastic abundance was significant for sites in West Bay and South Bay associated with intensive anthropogenic activities. West Bay of the Laguna Lake includes cities in the Philippines’ National Capital Region (NCR). NCR is the most densely populated region in the country (Schachter and Karasik, 2022). For instance, NCR has a population of approximately 14 million which accounts for roughly 13% of the total national population. The National Capital Region (NCR) remains the most densely populated region in the country in 2020. It is more than 60 times denser than the national average, with 21,765 people per square kilometer (Philippine Statistics Authority, 2021). Population and domestic waste generation have a positive linear relationship (Han et al., 2020), thus, domestic wastes on the lake could be sources of a large amount of plastic (Wang et al., 2018) which are the likely sources of microplastic bers in Laguna Lake.

The observed densities of microplastics in the surface water of Laguna Lake is within the range of the baseline assessment of microplastic concentrations in freshwater environments in Southeast Asian countries and other regions (Strady et al., 2021) except for Dongting Lake and Hong Lake in China which recorded high abundance of microplastics. Table 2 shows different freshwater environments that were studied in Asia and other regions, representing a total of six sampling sites, chosen for their environmental characteristics and their accessibility. Microplastics were measured in six surface waters. This variation in abundance of microplastics observed among different lakes from different parts of the world (Table 2) is an indication of differences in the lake's conditions and activities on the lake and that these conditions affect microplastic concentrations.

**Morphological Characteristics**

Microplastics were classified into fiber, fragments, film, granule, and filament (Fig 2) and were products of degradation of large plastic materials. On average, fiber was the most numerically abundant microplastic in surface water of the lake contributing 57% of total abundance, followed by fragment (21%), film (17%), filament (3%), and granule (2%) (Fig 3A).

At the site-specific level, fibers were present in all stations (Fig 3B). In East Bay (S8, S9 and S10) and West Bay (S2, S3 and S4) the abundance of microplastics varied in the order: Fiber>Film>Fragment>Filament>Granule where granule is only present in West Bay (S2) while In Central Bay (S5 and S6) and South Bay (S1 and S7) the order was Fiber>Fragment>Film>Granule where film and granules were only present in South Bay (S7). This is a similar observation in Dongting Lake and Hong Lake where 41.9% - 91.9% of bers dominate in surface water samples of the lakes (Wang et al., 2018). Moreover, studies conducted in freshwater lakes in China revealed that filament microplastics were the most abundant in both surface waters of Lakes Poyang (Yuan et al., 2019), and Taihu (Su et al., 2016).

Domination of microplastic fibers may represent land-based origin (Browne et al., 2011) and some can be due to abrasion and fiber release from synthetic fabrics especially in freshwater ecosystems. More than 1900 microplastic fibers were shed during the washing of a single polyester garment, resulting in more
than 100 per liter of effluent water (Browne et al., 2011). Additionally, sources of these type of microplastics includes laundering of synthetic textiles, tire erosion, total city dust (Boucher and Friot, 2017), household and office dust (Mishra et al., 2019) and materials from construction sites (Waldschlager et al., 2020). Polyester, the fiber form of polyethylene terephthalate, is also widely used in fabrics for apparel and other finished textile goods, accounting for nearly half of the global fiber market (Carr, 2017). The presence of microplastic fibers is concerning because recent studies have revealed several negative effects of microplastic fibers on aquatic organisms, including tissue damage, reduced growth, and body condition, and even mortality (Rebelein et al., 2021).

Microplastics on the glass filter were identified primarily using morphological characteristics (such as color, surface structure, and shape) and detailed criteria described in previous research (Hidalgo-Ruz et al., 2012; Su et al, 2016). Microplastics were found in a wide range of colors, including black, white, brown, blue, transparent, and red. The color distribution across size classes in each microplastic type was consistent, indicating that small-sized particles were byproducts of larger particle breakdown (Fig 4). The most prevalent color was blue, accounting for 53% of total microplastic count, with transparent, black, brown, white, and red accounting for 19%, 10%, 9%, 5% and 4% respectively (Fig 4).

In today's world, consumers are bombarded with plastic products in a variety of colors to increase their market potential (Thetford et al., 2003). All the microplastic debris were breakdown products of large plastic products, indicating that the various microplastic colors represented original product colors, though bleaching as the plastic debris wears out may change the original color of the plastic product (Stolte et al., 2015). Among the 132 microplastics research, the dominant microplastic color is blue which accounts for 32.9% among published research (Ugwu et al., 2021). The same research also revealed that the dominant microplastic shape is bers which has been consistent with our findings. Moreover, it can be also implied that sources of this microplastic are from disposable face masks (DFM) (Sajorne et al., 2022). The dominant color blue in this study was also one of the main colors of the fabrics used to make DFMs. Blue fabrics were mostly seen on the face masks' outer layers. This increased their exposure to radiation and abrasion, both of which favored the production of microplastics (Song et al., 2017).

**Polymer composition and its potential sources**

Out of 123 items extracted, there were 100 (81%) items confirmed as plastic polymer upon the alignment of the generated spectra with reference database from PerkinElmer FTIR analysis with spectral matches based on its library (Table 1). The reduced number can be associated with some articles of microplastics which showed limitations of the FTIR from a technical perspective (Xu et al., 2019). One limitation of an ATR-FTIR measurement is that it will detect materials on the sample's surface (Scientific T.F., 2018). Environmental exposure leads to polymer ageing and oxidative weathering of microplastic (Xu et al., 2019). Thus, if a sample has been weathered (has an irregular surface), this may make identification difficult (Scientific T.F., 2018). Additionally, the measurable particle size of an ATR-FTIR is roughly 500um to 5mm (Scientific T.F., 2018). Apart from technical perspective, collection of this minute particles can also be a consideration.
There were 11 types of polymers identified in the surface water of Laguna de Bay. Polymers identified were low-density polyethylene (LDPE), polypropylene (PP), polyethylene terephthalate (PET), general purpose polystyrene (GPPS), polyamide, high-density polyethylene (HDPE), polymethyl methacrylate (PMMA), polyvinyl chloride (PVC), acrylonitrile butadiene styrene (ABS), ethylene-vinyl acetate (EVA), and polybutylene terephthalate (PBT) (Fig 5A). Majority of the polymers were fibers which were all present in all types of polymers confirmed by FTIR except for PMMA (Fig 5B). The most abundant polymer assessed in Laguna de Bay's surface water was polypropylene (30%) (Fig 5A). Some of the major sources of PPs are plastic bags, storage containers, and microbeads in personal care products. Apart from personal cares, polypropylene was also used in making protective mask with 20% of those commercially available masks were made of polypropylene (Ellison et al., 2007). The abundance of polypropylene in this study can also be linked to the use of protective masks as an infection control measure, which was common in East and South-East Asia at the start of the COVID-19 and eventually in the world during 2020 and 2021 (Worby and Chang, 2020; Sajorne et al., 2022). In fact, the majority of the microplastics released from the face masks were medium-sized polypropylene fibers derived from nonwoven fabrics. Additionally, the abrasion and aging caused by wearing face masks increased the release of microplastics, particularly medium and blue microplastics (Chen et al., 2021). In the Philippines, 377 items of face masks were collected along eastern coast of Palawan (Sajorne, et al., 2022). Additionally, disposable masks made with density of 0.014 items/m$^2$ was observed in Davao Gulf in Mindanao (Abreo and Kobayashi, 2021). PMMA, ABS and PBT were among the least common microplastic polymer out of eleven (11) polymers comprising only 3%, 2% and 1% respectively.

The density of plastic debris and its behavior in aquatic systems are determined by the composition of microplastic particles. Microplastic debris, for example, may be suspended in the water column or sink to the sediment when discharged in an aquatic environment, depending on its density (Cole et al., 2011). Low density plastics, such as polyethylene and polypropylene, are less dense than fresh water and thus float on the water’s surface. Moreover, detection of these microplastics despite these limitations proved the occurrence of microplastics in the lake’s surface and should be given attention. GPPS which is denser than fresh water, were also found in the water surface samples studied (Fig 5A). The floating GPPS particles were most likely blown into foam, making them buoyant and thus able to float (Brignac et al., 2019). The sources of microplastics were linked to anthropogenic activities on the water, recreation, and nearby trading centers. Microplastic fibers were common among the eleven (11) polymer types except for PMMA which happens to be microplastic films.

Conclusions

Our results demonstrated that the surface water of Laguna Lake is contaminated with microplastics. Microplastics were ubiquitously detected in all sites with the concentration highest in areas of the lake characterized by intensive human activities. This study provides the first documented evidence of microplastics in the surface water of Laguna de Bay and the first among the lakes in the Philippines. All the microplastics were pieces of plastic used by most of the community with the major polymers being
polypropylene, ethylene-vinyl acetate copolymer and polyethylene terephthalate. A majority of the microplastics were small colored particles specifically blue colored microplastics which pose a threat to water quality and fisheries of the lake as these can easily enter the food chain. Furthermore, use of plastic bags and other plastic materials by fishermen to hold stones used as sinkers for the fish gillnets and floaters for aquaculture in the lake calls for urgent interventions aimed at reducing microplastic pollution of the lake. PPEs such as disposable face masks were also linked to be contributors of microplastic pollution in the lake. The risks that microplastics pose to fish and its natural foods especially invertebrates, and the possible link to human health need to be understood. Strategies such as proper waste management, plastic recycling, and penalties for illegal dumping in areas close to water resources should be promoted and implemented in the communities. The accumulation and effects of these microplastics to aquaculture species and sediment in Laguna de Bay deserve further investigation.

Declarations

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Author contribution

Cris Gel Loui Arcadio conceived the present study, planned out the sampling, and analyzed the data. Sheila Mae B. Ancla helped in the actual sampling. Sherley Ann Inocente, Carl Kenneth P. Navarro, Kaye M. Similatan and Marybeth Hope T. Banda assisted in the sorting and analysis of microplastics in the laboratory. Rey Y. Capangpangan, Armi G. Torres, and Hernando P. Bacosa actively contributed to the conceptualization of the study and review of the manuscript.

Declarations  Ethical approval  Not applicable

Consent to participate  All authors have agreed to participate to this study.

Consent for publication  All authors have agreed to be co-authors of this manuscript.

Competing interest  The authors declare no competing interest.

Availability of data and materials  The datasets used and/or analyzed in this study are available upon reasonable request from the corresponding author.
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Tables

**Table 1** Geographical coordinates for each sampling station with the number and density of microplastics assessed

| Station | Latitude       | Longitude      | No. of Microplastics | Density (particles/m³) |
|---------|----------------|----------------|----------------------|------------------------|
| 1       | 14°11'18.26"N  | 121°11'25.13"E| 12                   | 17.14                  |
| 2       | 14°19'21.82"N  | 121° 7'48.33"E| 19                   | 24.17                  |
| 3       | 14°25'22.94"N  | 121° 7'53.29"E| 12                   | 17.14                  |
| 4       | 14°30'51.88"N  | 121° 7'4.79"E | 13                   | 18.57                  |
| 5       | 14°29'25.77"N  | 121°16'15.97"E| 8                    | 11.43                  |
| 6       | 14°25'9.21"N   | 121°16'35.28"E| 5                    | 7.14                   |
| 7       | 14°13'53.01"N  | 121°15'39.83"E| 7                    | 10.00                  |
| 8       | 14°15'41.62"N  | 121°20'52.60"E| 10                   | 14.29                  |
| 9       | 14°19'25.82"N  | 121°24'29.50"E| 6                    | 8.57                   |
| 10      | 14°22'59.58"N  | 121°27'9.51"E | 8                    | 11.43                  |

**Table 2** List of surface water microplastics studies in freshwater lakes in Asia and other regions and corresponding density.
| Study Site                        | Density (particles/m³) | Method of analysis                | Reference        |
|----------------------------------|------------------------|-----------------------------------|------------------|
| Laguna de Bay, Philippines       | 14.29                  | ATR-FTIR Analysis                 | This study       |
| Chiusi Lake, Italy               | 3.02                   | Visual inspection                 | Fischer et al. (2016) |
| Bolsena Lake, Italy              | 2.51                   | Visual inspection                 | Fischer et al. (2016) |
| Dongting Lake, China             | 1191.7                 | SEM Analysis                      | Wang et al. (2017) |
| West Dongting Lake, China        | 616.67                 | Raman Spectroscopy                | Jiang et al. (2018) |
| South Dongting Lake, China       | 716.67                 | Raman Spectroscopy                | Jiang et al. (2018) |
| Hong Lake, China                 | 2282.5                 | SEM Analysis                      | Wang et al. (2017) |
| Taihu Lake, China                | 0.123                  | m-FT-IR and SEM/EDS               | Su et al. (2016)  |
| Poyang Lake, China               | 0.005                  | Raman Spectrometer                | Liu et al. (2019) |

**Figures**

Figure 1

Sampling stations for microplastic assessment in surface water of Laguna de Bay, Philippines
Figure 2

Types of microplastic assessed in surface water of Laguna de Bay based on shapes A) filament B) fragments C) fibers D) granules E) film

Figure 3

Relative abundance of microplastics based on shape (A) and its composition in different sampling stations (B)
Figure 4

Relative abundance of microplastics based on color (A) and its composition in different sampling stations (B).

Figure 5

Percent composition of different microplastic polymer type (A) and the different microplastic shapes within its polymer type (B).