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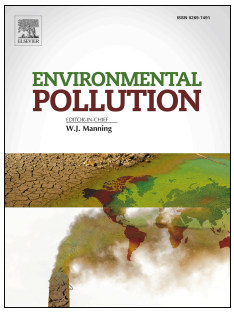

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The spatial distribution and abundance of microplastics in lake waters and ice during ice-free and ice-covered periods

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Abbreviations: ANOVA, analysis of variance; MP, microplastic; PE, polyethylene; PET, polyethylene terephthalate; PP, polypropylene; PS, polystyrene; PVC, polyvinyl chloride; QA/QC, quality assurance and quality control.

ABSTRACT

Understanding the spatial distribution and characteristics of microplastics (MPs) in lake waters is essential to assessing and addressing lacustrine MP pollution. This study investigated how lake ice affects the abundance, spatial distribution, and characteristics (size, shape) of MPs in Lake Ulansuhai by analyzing samples collected at ten sites uniformly distributed throughout the lake during ice-free and ice-covered periods. The abundance of MPs ranged between 204±28 and 1224±185 $n \cdot L^{-1}$ in lake waters during the ice-free period, and from 34±8 to 216±21 $n \cdot L^{-1}$ and 269±84 to 915 ±117 $n \cdot L^{-1}$ in water and ice during the ice-covered period, respectively. During the ice-covered period, MPs were 2.74–8.14 times higher in the ice than in water beneath the ice. Ice formation decreased MP abundance in lake waters, in part, by incorporating a relatively high percentage of MPs into the ice mass during freezing and by inhibiting atmospheric MPs from reaching the lake waters. The abundance of MPs in the water during the ice-free period was 4.50–11.30 times greater than during the ice-covered period. Seasonal variations in MP shape also occurred; the proportion of fibrous MPs in water decreased during the ice-covered period. Variations in MP abundance were partly due to differences in sedimentation rates; the settling of fibrous MPs is slower, making it easier for them to be captured during the formation of surface ice. Spatially, MPs were uniformly distributed during the ice-free period, but exhibited a spatially distinct pattern during ice-covered periods, when MPs in lake waters were higher in the northeast and lower in the southwest portions of the lake. During the ice-free period, small MPs (0.05–0.5 mm) were more likely to move with currents in the lake, whereas water velocities were reduced by ice formation, allowing small MPs to accumulate near the lake inlet.

Keywords: Microplastics; Spatial-temporal variations; Cold lacustrine ecosystems; Lake Ulansuhai

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Introduction

Microplastics (MPs) are plastic particles with a size of less than 5 mm (Shenoy et al., 2022). They are derived from two types of sources: primary and secondary. Primary MPs include mainly pellets used as feed stock in the industrial processing of plastic products (Perera et al., 2022). In contrast, secondary MPs are derived from the degradation of larger pieces of plastic and plastic debris (Zhao et al., 2022). Due to the widespread use of plastic products, and their derivation from multiple sources, MPs are nearly ubiquitous in aquatic ecosystems (Ajith et al., 2020; Schmidt et al., 2020; Forero-López et al., 2021; Kumar et al., 2021; Cai et al., 2022).

As an emerging pollutant, MPs are perceived to negatively affect freshwater ecosystems. For example, MPs can affect the feeding, growth and reproduction of aquatic organisms, in part because they have the potential to sorb and carry a wide range of chemical pollutants (e.g., toxic trace metals, organic cyclic aromatic hydrocarbons, and polybrominated diphenyl ethers) (Tan et al., 2019; Dong et al., 2020; Singla et al., 2020; Kutralam-Muniasamy et al., 2021; Kumar et al., 2022) that once ingested may affect biota (e.g., reduced mobility, ineffective foraging and subsequent mortality) (Anbumani and Kakkar, 2018; Kolandhasamy et al., 2018; Wu et al., 2019; Naidoo et al., 2020; Yin et al., 2020; Zhang et al., 2021; Ratnayake and Perera, 2022). Moreover, MPs often contain a wide range of toxic chemical additives (e.g., plasticizers, flame retardants, and antioxidants) (Liu et al., 2020; Luo et al., 2020a; Luo et al., 2020b). It follows, then, that the transport of MPs may represent an important migratory pathway of contaminants in lacustrine environments.

The sources, distribution, migration, toxicity, and biological effects of MPs in marine

and coastal environments are well recognized (Rezania et al., 2018; Campos da Rocha et al., 2021; Uzun et al., 2022). Microplastics in freshwater systems, such as lakes and rivers, has been less extensively studied (Li et al., 2020; Dusaucy et al., 2021; Silva-Cavalcanti et al., 2023). In fact, Rodrigues et al. (2018) found that only about 15% of the papers published on MPs pertain to freshwater ecosystems. Freshwater ecosystems of considerable importance are inland lakes, which represent an important storage site and regulator of MP transport to the oceans. As a result, both urban and alpine lakes have become the topic of recent MP studies. These investigations have shown that their pollution by MPs is becoming increasingly prominent (Galafassi et al., 2021; Li et al., 2022a).

In general, the distribution of MPs in lakes is predominantly controlled by human activities, hydrodynamic conditions, and the properties of the MPs themselves, such as their size and shape (Jian et al., 2020; Warrier et al., 2022; Yang et al., 2022). Climatic conditions or temporal changes in the lake environment may also alter the distribution and abundance of MPs in the lake water through time (Li et al., 2022b; Simantiris et al., 2022; Yao et al., 2022). For example, researchers have found in recent years that ice formation (freezing) may strongly influence the distribution of MPs in marine environments, where the abundance of MPs in the ice body is 2–5 orders of magnitude higher than that in the subglacial water (Kanhai et al., 2020; Kelly et al., 2020; von Friesen et al., 2020). Sea ice, then, appears to represent an important temporary storage site for MPs that aids in the transport and distribution of MPs in cold regions. It is currently unclear, however, if such processes also occur in freshwater lakes characterized by the seasonal formation of surface ice. Nonetheless, on the basis of Liu et al. (2017), it seems likely that MPs in cold-region

lakes will be partitioned among ice, water, and sediment, and the spatial distribution and abundance of MPs in these lakes will change seasonally as a result ice formation.

In this study, a shallow lake (Lake Ulansuhai) in a cold, arid region of China was investigated to assess the spatial distribution of MPs in water and ice during the ice-free and ice-covered periods to (1) document the spatial distribution and abundance of MPs in lake water during the two periods; and (2) determine the partitioning of MPs between water and ice during the ice-covered period, and the differences that occur in their spatial distribution between each media type.

Methods and materials

Study area

Lake Ulansuhai is the largest freshwater lake in the Yellow River Basin. It is located in central Inner Mongolia (40°36′–41°03′N, 108°43′–108°57′E; Fig. 1), and covers an area of approximately 320 km^2. The depth of water in the lake ranges from 1.8 to 3 m, and averages 2.5 m. The water retention time is approximately 240–290 days. The dominant wind directions in the area are SSW, NNE, and NW. Lake Ulansuhai is frozen for approximately 4–5 months a year. The thickness of the ice sheet can reach one-third to one-half of the water depth.

Lake Ulansuhai is an important component of the water conservation project in the Hetao irrigation area (7,733 km^2), and receives more than 90% of the region's annual agricultural drainage. The lake also receives domestic sewage and industrial wastewater from several upstream cities and towns as well as sewage from livestock and poultry breeding facilities, most of which flows into the northwestern part of the lake. There are

also two cities within 100 km of the lake's east and west sides with a population of more than 1.5 million. Seven kilometers south of the lake, there is a city with a population of 250,000, and numerous small towns occur around the lake margins.

Sampling

According to the Technical Regulations for National Wetland Resources Survey (2008, China) and the characteristics of this body of water, the lake was subdivided into 2 km × 2 km grid cells. A total of 10 sampling sites were then located at the intersection of the grid lines. In some case, sampling conditions required minor alterations in the location of the sampling points, but they were as evenly distributed throughout the lake as possible (Fig. 1). Samples were collected in August 2020 (ice-free period) and again in January 2021 (ice-covered period). Three samples were collected from each sampling point and analyzed separately. During the ice-free period, the samples were collected by placing the mouth of a closed (capped), 2 L water bottle 50 cm below the water surface, after which the bottle was opened and raised to the water surface. Water samples were filtered through a 50-μm sieve. Materials captured on the sieve were subsequently washed using distilled water into a 250-ml flask contain a 5% formalin solution and stored at 4°C. During the ice-covered period, a block of ice was extract from the ice sheet and then placed in "cooler" for transport to the laboratory where the ice was allowed to melt at room temperature. The underlying lake water was also sampled through the hole in the ice (where the ice block was extracted) using the methods described for the ice-free period.

Sample preparation

In the laboratory, MPs were collected from the water samples using a modified density separation method (Xiong et al., 2018) that was initiated by filtering the sample through a 50-μm stainless-steel sieve. Particles on the sieve were then washed into an overflow flask contain a 1.54-g/L potassium formate solution. After standing for 24 h, additional potassium formate solution was added to flask until the surface liquid overflowed the flask's outlet and captured in a glass bottle. The overflow process was then repeated three times at 12-h intervals.

The collected lake waters contained organic and biological substances that could affect the analysis. To remove these substances, the overflow liquid obtained in the previous step was filtered again through a 50-μm stainless-steel sieve, and the collected material was washed into a 250-ml glass conical flask using 30% (V/V) hydrogen peroxide. An additional 100 ml of hydrogen peroxide was added to the flask, which was then capped and placed into an oscillating incubator at 45°C and 80 rpm for 24 h. If organic substances persisted, the digestion time was extended until complete. After digestion, deionized water was added to the sample before it was filtered through a 0.45-μm (GF/F, 47 mm ø, Whatman) membrane that was subsequently placed into a glass dish and dried at room temperature.

MP identification and characterization

MPs on the filter membrane were observed with a stereomicroscope (M165C, Leica, Germany) at a magnification of 160×. Suspected MPs were identified using previously developed MP identification criteria (e.g., Hidalgo-Ruz et al., 2012). The size and shape the MPs was documented using Leica Application Suite X software. Once determined, the

159 shape of MPs was subdivided into four types, including fibers, fragments, films, and grains
160 (Liu et al., 2022). The size of the MPs was categorized into three intervals: 0.05–0.5, 0.5–
161 2, and 2–5 mm.

162 The normality of the data was assessed using a Shapiro-Wilk normality test prior to
163 the application of statistical analyses. Based on these results, a one-way analysis of
164 variance (ANOVA) was used to analyze statistical differences in the abundance of MPs
165 between sampling points and media types during a given period. One-way ANOVA was
166 also used to analyze statistical differences in the abundance of MPs between ice-covered
167 and ice-free periods. Data that were not normally distributed were standardized using the
168 data standardization module in SPSS before one-way ANOVA was used.

169 A total of 130 MP particles were randomly selected from water samples collected
170 during the ice-free period, whereas 46 and 89 particles were selected from the water and ice
171 samples obtained during the ice-covered period, respectively. These samples were then
172 qualitatively and quantitatively analyzed using a microlaser confocal Raman spectrometer
173 (HR Evolution, HORIBA, France; laser wavelength, 532 nm; spectral scanning range,
174 100–3200 cm^{-1}) to: (1) determine the composition of the MPs, and (2) verify the accuracy
175 of visual identifying the MPs using microscopic methods. Determination of particle
176 composition was determined by comparing the sample spectrogram with spectra in a
177 Biorad (Knowitall) Raman database (which includes 18 categories of substances and over
178 28,000 spectra). Determination of a compound required a 70% match between the spectra
179 from the database and instrument (Di and Wang, 2018).

180

181 *Quality assurance and quality control (QA/QC)*

To ensure that the MP samples were not affected by external particles during sample collection, processing, and analysis, the sampling containers and experimental instruments were cleaned three times with ultrapure water before each experiment, and the experimental table and microscope wiped with alcohol. The surface after each step in the experiment was covered by aluminum foil. During the entire sampling and analysis process, the experimenter always wore a cotton lab coat, and none of the analyses used plastic containers. Both field and laboratory blanks were also analyzed to assess potential contamination. The abundance of MPs in the sample blank ranged from 0 to 2±1 $n \cdot L^{-1}$, indicating that sample contamination was negligible.

Results

Abundance and spatial distribution of MPs

As shown in Fig. 2 to Fig. 4, the abundance of MPs in lake water during the ice-free period was 4.50 to 11.30 times higher than that during the ice-covered period (concentrations in the collected samples were statistically different, $p < 0.05$). The abundance of MPs in water during the ice-free period also differs from the abundance MPs found in ice ($p < 0.05$). Similarly, the abundance of MPs in ice was 2.74–8.14 times higher than that in the water under the ice during the ice-covered period.

Spatially, the abundance of MPs in the lake water during the ice-free period were random distributed, and ranged between sampling points from 204±28 to 1224±185 $n \cdot L^{-1}$ ($p < 0.01$, between sampling points). During the ice-covered period, the abundance of MPs in water was highest near the inflow of water from the main drainage channels (i.e., the northeastern part of the lake). MP abundance, which ranged from 34±8 to ~216±21 $n \cdot L^{-1}$,

205 then decreased from northeast to southwest, away from these inputs. ($p < 0.01$, between
206 sampling points). Similarly, during the ice-covered period, the abundance of MPs in lake
207 ice at the 10 sampling sites ranged between 269±84 to ~915±117 $n \cdot L^{-1}$ ($p < 0.01$, between
208 sampling points), and was higher in the northeast and central parts of the lake.
209
210 *Variations in MP shapes and sizes in water and ice*
211 The proportions of fibrous, fragmented, filmy, and granular MPs in water and ice at
212 the different sampling points during the ice-free period and ice-covered period are shown
213 in Fig. 5. In general, the proportion of fibrous MPs in water decreased after the ice formed,
214 leading to a higher proportion of fibrous MPs in the ice body than in the water. The
215 proportion of fragments, films, and MP grains in water increased after ice-formation, and,
216 in the case of fragments and films, was similar to the proportions observed in ice. The
217 proportion of MP grains in the ice was lower than the in water beneath the ice.
218 In general, the proportions of MPs of different sizes in water did not significantly
219 change at the sampling points after the lake ice formed (Fig. 6). However, the proportions
220 of MPs with sizes ranging from 0.05 to 0.5 mm near the input canal increased significantly
221 after the lake surface froze. The relative size of MPs in the ice was similar to that in the
222 water under the ice. The statistical analysis results regarding the proportions of MPs of
223 different sizes and shapes in water between ice-free period and ice-covered period are
224 shown in Table 1.
225
226 *MP identification and characterization*
227 Most suspected plastic particles selected for analysis by Raman spectrometry (Fig. 7)

were, in fact, composed of plastics. More specifically, the percentage of particles collected from water during the ice-free period, and water and ice during the ice-covered period that were determined to be a plastic polymer was 98.5%, 84.8%, and 93.3%, respectively. These data indicate that the collection, separation, and manual identification methods adopted in this study were reliable. In this study, the MPs found in the water and ice were mainly composed of polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), and polyethylene terephthalate (PET).

Discussion

Partitioning of MP between ice and water

Studies conducted in marine environments have shown that MPs may be incorporated into ice during the freezing process such that it alters the relative proportions of MPs in the ice and underlying water (Peeken et al., 2018; Geilfus et al., 2019; Hoffmann et al., 2020). Kanhai et al. (2020), for example, investigated the abundance of MPs in Artic ice, and found that the abundance of MPs in the ice ranged from 2 to 17 $n \cdot L^{-1}$. In contrast, the abundance of MPs in the underlying water ranged from approximately 0 to 1.8×10^{-2} $n \cdot L^{-1}$, or a concentration that is about 1,000 times less than in the ice. Similarly, Kelly et al. (2020) found that sea ice in east Antarctica exhibited an abundance of MPs of approximately 6 to 33 $n \cdot L^{-1}$, which was nearly 1 million times greater than that observed by Isobe et al. (2017) in Antarctic waters (which was approximately 3.1×10^{-5} $n \cdot L^{-1}$).

In Lake Ulansuhai, the abundance of MPs in the ice was also found to be higher than that in the water beneath the ice (Figs. 3 and 4), indicating that a relatively high proportion of the total MPs entered the ice during the freezing process. The observed partitioning is

likely to be linked to both the mechanics through which ice is formed and the nature (size, shape) of the MPs present in the water (the latter is discussed in a subsequent section). When water bodies freeze, small, flat platelets of pure ice form first. As the ice mass grows, these platelets commingle and bond to form a compliant slush. Under sustained freezing, platelets fuse as their thickness increases, forming an ever-stiffening ice cover (Zhang et al., 2012). During the downward formation of the ice, pollutants are not incorporated into the ice crystals. However, cracks, bubbles, and other features commonly form in the ice as a result, in part, of wind and wave activity, creating open spaces in the ice (Dadic et al., 2010; Petrich and Eicken, 2010). MPs and other pollutants can enter these spaces as the ice grows downward from the surface, and occur as "impurities" in the ice mass (Ge et al., 2016; Liu et al., 2017).

Some MPs are less dense than water (such as PE and PP), and float on the surface when they enter a lake. Even high-density MPs, or MPs associated with aggregates can be re-suspended from lakebed sediments and remain in the water column for a long period of time (Su et al., 2016; Wang et al., 2017; Di and Wang, 2018). During the freezing process, these MPs are likely to be concentrated directly within open spaces (e.g., in fractures or ice bubbles) in the surface ice (Chubarenko, 2022). Incorporation of MPs may be especially high during the early stages of ice growth when the temperature difference between day and night is large, and the lake water repeatedly freezes and thaws, creating ice with a high porosity (Nürnberg et al., 1994; Petrich and Eicken, 2010).

The partitioning ratio of MPs between sea ice and sea water (observed in the studies cited above) is much higher than we found between lake ice and lake water. The difference may be due to the higher density and surface tension of sea water in comparison to lake

274 water. Sea water is likely to possess higher MP surface water concentrations, facilitating
275 the incorporation of MPs into the ice during initial stages of ice growth (Geilfus et al.,
276 2019). In addition, the ambient air temperature in high latitudes is lower (generally below
277 0°C) all year round, allowing marine ice in polar regions to remain throughout the summer.
278 The persistence of ice throughout the year prevents all of the MPs trapped in the sea ice
279 from being released (Obbard et al., 2014). Finally, due to its continuous movement, long
280 migration distance, and long time over which ice grows, sea ice may continuously capture
281 and accumulate MPs in the ocean over a broad area during vertical freezing (Kanhai et al.,
282 2020). In Lake Ulansuhai, however, the lake completely freezes for only a few months,
283 allowing the lake ice to only capture MPs during vertical (downward) ice growth. Owing to
284 the short period of ice-growth and the relatively thin nature of the ice layer in the lake the
285 accumulation of MPs is relatively small in comparison to sea ice.

286 Spatially, however, sea ice only grows and traps MPs in cold regions, and the
287 thickness of sea ice is usually much smaller than the depth of the water. Thus, the effect of
288 sea ice on the distribution and fate of MPs in the ocean is limited. In contrast, shallow lakes
289 are often completely covered with ice during the winter months, and the thickness of the
290 lake ice is close to or even greater than the depth of the water. Moreover, lake ice continues
291 to capture MPs from the lake water during the ice growth period. After the ice melts, all of
292 the MPs in the ice enter the lake water in a relatively short time (especially when the
293 surface ice melts). Thus, ice is an important source and sink of MPs throughout in the entire
294 lake, and the effect of ice formation on the migration and distribution of MPs in shallow
295 lakes is greater than that in the oceans.
296

Potential impacts of surface ice on MP abundance and character in lake waters

The abundance of MPs in lake water decreased greatly as ice formed on the surface of the lake. The abundance of MPs in lake water during the ice-free period was 4.50–11.30 times higher than that during the ice-covered period. Part of the observed depletion was undoubtedly due to the enhanced partitioning of MPs into the ice by the processes described in the previous section. It is likely, however, that the observed lowering of MP concentrations in lake water during the ice-covered period was enhanced by several other factors. First, data from previous studies suggest that the predominate source of MPs in the lake is likely to change seasonally. For example, Hao et al. (2022) demonstrated that MPs in Lake Ulansuhai during the warm, ice-free period are mainly derived from fishing activities and drainage (surface runoff), including that from farmlands. Plastic products such as plastic films and fishing nets are widely used in agricultural production in the surrounding irrigated areas and in fishery activities, leading to the production of a large amount/number of MPs. In fact, 42.36 t of MPs are annually transported to Lake Ulansuhai by drainage from the Hetao irrigated area (Wang et al., 2020). However, as lake ice forms, surface drainage greatly decreases, and fishing activities in the lake stop, both of which greatly reduce the quantity of MPs received by Lake Ulansuahi during the ice-covered period.

The second control on MP abundance is that a significant, but unquantified amount of the MPs in the lake are presumably derived from the atmospheric deposition of particulate matter. Varying concentrations of MPs have been, for example, observed in samples of airborne dust (Brahney et al., 2020), snowfall (Bergmann et al., 2019), and aerosols (Abbasi et al., 2019) in terrestrial regions. Such MPs in the atmosphere can be transported

over long distances as a result of atmospheric circulation processes, after which they are redeposited in urban, agricultural, and aquatic ecosystems through both wet and dry deposition (Dris et al., 2016; Ding et al., 2021). As noted in the description of the study area, several large cities (with a population >1.5 million) that are likely to produce significant quantities of airborne MPs (Hao et al., 2022) exist within 100 km of Lake Ulansuhai. Wind, and the dust that it transports, occurs frequently in the basin, increasing the likelihood that atmospheric deposition is an important source of MPs in the lake. Surface ice may affect the initial input as well as the abundance and partitioning of atmospheric MPs in the lake in two important ways: (1) during the ice-covered period, MPs cannot directly enter the lake water, thereby reducing MP concentrations in the water during the ice-covered period, and (2) the atmospherically deposited MPs will accumulated on the ice surface where they may be subsequently incorporated into the ice.

The third way in which surface ice may influence the abundance of MPs in lake water is through its impact on waves and currents. High-density MPs, or low-density MPs associated with aggregates, will settle to the lakebed and become incorporated in the sediment (Reisser et al., 2013; Leiser et al., 2021). Sediments, then, serve as both a source and sink of MPs in the lake. Lake Ulansuhai is a relatively shallow lake, and frequent winds often create hydrologic flows (Liu et al., 2019) capable of remobilizing lakebed sediments during ice-free periods. However, when the lake surface is frozen, such winds are unable to generate flows that are capable of resuspended MPs within the lakebed sediments, thereby reducing MP concentrations in the lake water.

In general, after the lake enters the ice-covered period, a large amount of MPs are captured by the ice, and the input of MPs to the water is greatly reduced. When coupled

with the weakening of hydrodynamic force, the abundance of MPs in lake water is greatly reduced. Although no long-term continuous observations have been made, we infer that the abundance of MPs in Lake Ulansuhai waters alternated between high and low concentrations during the ice-free and ice-covered periods, respectively, a trend that is likely to occur in the water of other shallow, seasonally ice-covered lakes.

Seasonal controls on the spatial distribution of MP abundance in lake waters

The spatial distribution of MPs in the water of Lake Ulansuhai differed seasonally (Figs. 2 and 3). Such spatial differences in MP concentrations within cold region lakes may be controlled by the location of MP inputs and outputs, variations in the amount of MPs input into the lake from specific sources, and hydrodynamic conditions associated with drainage and seasonal ice cover. In the case of Lake Ulansuhai, the main input drainages (dominated by the main and eighth drainage channels; Fig. 1) are found in the northwestern section of the lake and account for 95%–100% of the total inflow. Upon entering the lake, the water primarily flows from northwest to the lake's southern outlet under the influence of gravity. Additional surficial flow patterns generated by gales that occur between April to November are superimposed on this primary current. These gales (winds) are primarily oriented to the SSW, ENE, and NNE (Wang et al., 2020), and generally increase the velocity of the flow toward the lake's southern outlet. During the ice-free period, then, discharge into the lake is large and the flow rate in the lake is relatively fast. The velocity at different locations in the ice-free lake varied during the study period from 0.02 to 0.10 m/s; the average flow velocity was 0.04 m/s (Liu et al., 2019). Liu et al. (2022) found that during the ice-free period, hydrodynamic forces driven in large part by wind were able to

resuspend MPs in the lake sediments, after which they were transported by the general northwest to south currents to the lake's outlet. These conditions presumable resulted in the relatively high, but uniform, MP concentrations throughout the majority of the lake during the ice-free period (Fig. 2).

With the formation of surface ice, the remobilization of lakebed sediments by wind-driven waves would have become negligible. In addition, ice formation was associated with a concurrent decrease in drainage to the lake from the main drainage channels, resulting in a significant decline in gravity-driven flow from the northwest to south. For example, the velocity at different locations in the lake varied from 0 to 0.01 m/s during the ice-covered period, and order of magnitude lower than the velocities measured during the ice-free period. In combination, the distribution of MPs in lake water decreases significant from northeast to south during the ice-covered period.

Seasonal controls on MP characteristics

Factors influencing particle shape

It is important to recognize that not all MP particles were incorporated at equal rates into the lake ice. After entering the ice-covered period, the proportion of fibrous MPs in the lake water decreased. Fibrous MPs are light, small, and easily suspended, making them easy to capture during freezing. Khatmullina and Isachenko (2017), for instance, used settling experiments to show that the rotation of fibrous MPs in water (including rolling, bending, and twisting) increased their resistance to settling and lowered their settling velocity. Because such fibrous MPs have a lower settling rate, they are more likely to be captured by the ice during freezing (Chubarenko, 2022).

389 Generally, MPs transported in the atmosphere and deposited over the landscape are
390 dominated by fibers. Abbasi and Turner (2021), for example, found that in Iran, fibers
391 comprised >99% of the total plastic particles in atmospheric dust and precipitation.
392 Similarly, the preliminary analysis by Zhou et al. (2017) showed that fibers accounted for
393 more than 95% of the total atmospheric plastics in Yantai. In marked contrast, Wang et al.
394 (2020) investigated the proportions of MPs of various shapes in the drainage inflowing to
395 Lake Ulansuhai, and showed that the proportions of fibrous MPs in the drainage water
396 ranged from 34.98% to 70.39%, which was much lower than the proportions of fibrous
397 MPs in the water during the ice-free period during this study (66.23%–95.16%). Moreover,
398 Wang et al. (2020) found that the proportions of MP grains, films, and fragments in the
399 drainage water (runoff) ranged between 29.61–65.02%, which was higher than the
400 proportions of these MPs in lake water during ice-free period (4.84%–33.77%). The
401 difference in MP shape observed between runoff and the atmosphere suggests that MP
402 shape in lake waters will be affected by seasonal variations in source inputs, with increases
403 in fibers occurring during the ice-free period when the atmospheric deposition of fibrous
404 MPs is enhanced. In contrast, during the ice-covered period, the fibrous MPs in the
405 atmosphere cannot enter the lake water, presumably decreasing the proportion of fibrous
406 MPs in the water. However, the proportion of MP grains, films, and fragments increased in
407 the water, along with their overall abundance. In general, then, seasonal changes in runoff
408 input and ice cover had a significant effect on the proportion of MPs of different shapes in
409 the water body of the lake.
410
411 *Season controls on the proportion and spatial distribution of MPs of differing size in lake*

412 *waters*

413 The proportion and spatial distribution of small-sized MPs in lake water changed with
414 the occurrence of lake ice. More specifically, the proportion of MPs with a size ranging
415 from 0.05 to 0.5 mm in the northern part of the lake during the ice-free period was much
416 smaller than during the ice-covered period (Fig. 6). We suggest that the seasonal, spatial
417 changes in MP size is related to the location of lake's primary runoff inputs and the size
418 characteristics of the runoff's MPs. In general, the smaller the size of the MPs, the easier it
419 is for them to be transported by flowing water. Besseling et al. (2017), for example, used
420 the DUFLOW model to simulate the migration of MPs in the Domer River, and showed
421 that larger MPs settled faster and were transported over a shorter distance. Smaller MPs
422 settled more slowly and migrate farther. In our previous study (Liu et al., 2022), MP traps
423 were used to explore the deposition of high-density MP in Lake Ulansuhai. We also found
424 that larger MPs settled faster and moved shorter distances, while small-sized MPs settled at
425 a lower rate and moved easily with the water currents during the ice-free period. In this
426 study, the relatively high concentration of MPs within the size range of 0.05 to 0.5 mm in
427 the southern part of the lake (near the outlet) during the ice-free period also appears to be
428 due to the relatively high flow rates that can readily transport small-sized particles.
429 However, during the ice-covered period, lower flow velocities decreased the transport of
430 these relatively small MPs from the northern sites of runoff input. Rather, particles of all
431 sizes, accumulated near the entrance to runoff channels in the northern part of the lake
432 during the ice-covered period. Put differently, seasonal ice cover in Lake Ulansuhai
433 influenced the spatial distribution of MPs of relatively small size but had no obvious
434 influence on the distribution of larger MPs.

435

436 **Conclusions**

437 The seasonal formation of lake ice was found to have a significant effect on the
438 abundance, characteristics (size, shape), and spatial distribution of MPs in lake waters.
439 During the ice-covered period, for example, the resuspension of MPs in lakebed sediments
440 was presumably reduced by wind and waves, while atmospheric inputs of MP fibers were
441 severely reduced as they were captured by the ice surface. The amount of MPs entering the
442 lake with runoff and from fishing activities were also greatly reduced.

443 Similar to cold, marine environments, lake ice captured MPs during ice formation; the
444 incorporation of fibers was particularly significant, presumably because of their propensity
445 to float near the water surface. While fibers were preferentially captured by the ice, the
446 atmospheric inputs of fibers to the water were reduced by the surface ice, which in
447 combination significantly change the form of the MPs on a seasonal basis. The ice, then,
448 served as a temporary sink of MPs, particularly fibers, in the lake during the ice-covered
449 period as well as a source of MPs to the water as the MPs were released from the ice as it
450 melted. The freeze-thaw process is likely to have prolonged the residence time of MPs in
451 the lake, and may have enhanced the abundance of MPs that could come into contact with
452 aquatic organism during the spring when the ice melted. While additional research is
453 needed, the collected data show that it is necessary to investigate the effects of lake icing on
454 the abundance, spatial distribution, and character of MPs in cold region lakes, and that ice
455 formation may influence the potential ecological impacts of MPs on biota.

456

457 **Declaration of competing interest**

458 The authors declare that they have no known competing financial interests or personal
459 relationships that could have appeared to influence the work reported in this paper.
460
461 **Data availability**
462 Data will be made available on request.
463
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736 **Table 1**

737 The statistical differences in the proportions of microplastics (MPs) of different sizes and

738 shapes in water between ice-free period and ice-covered period.

Types of MPs	p value
Fibrous	<0.01
Fragmented	<0.01
Filmy	<0.01
Granular	<0.01
0.05–0.5 mm	0.367
0.5–2 mm	0.140
2–5 mm	0.695

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740 **Figure captions**

741 **Fig. 1.** Map showing the location of the study are in China (a) and Inner Mongolia (b); Map
742 showing the location of sampling sites in Lake Ulansuhai, and the major drainage channels
743 to the lake (c).

744 **Fig. 2.** Spatial distribution and abundance of MPs in water during the ice-free period.

745 **Fig. 3.** Spatial distribution and abundance of MPs in water during the ice-covered period.

746 **Fig. 4.** Spatial distribution and abundance of MPs in ice during ice-covered period.

747 **Fig. 5.** The proportion of MPs of differing shape in water and ice at different sampling
748 points during the ice-free and ice-covered periods.

749 **Fig. 6.** The proportion of MPs of differing size in water and ice at different sampling points
750 during the ice-free and ice-covered periods.

751 **Fig. 7.** Typical Raman spectra of selected MPs.

752

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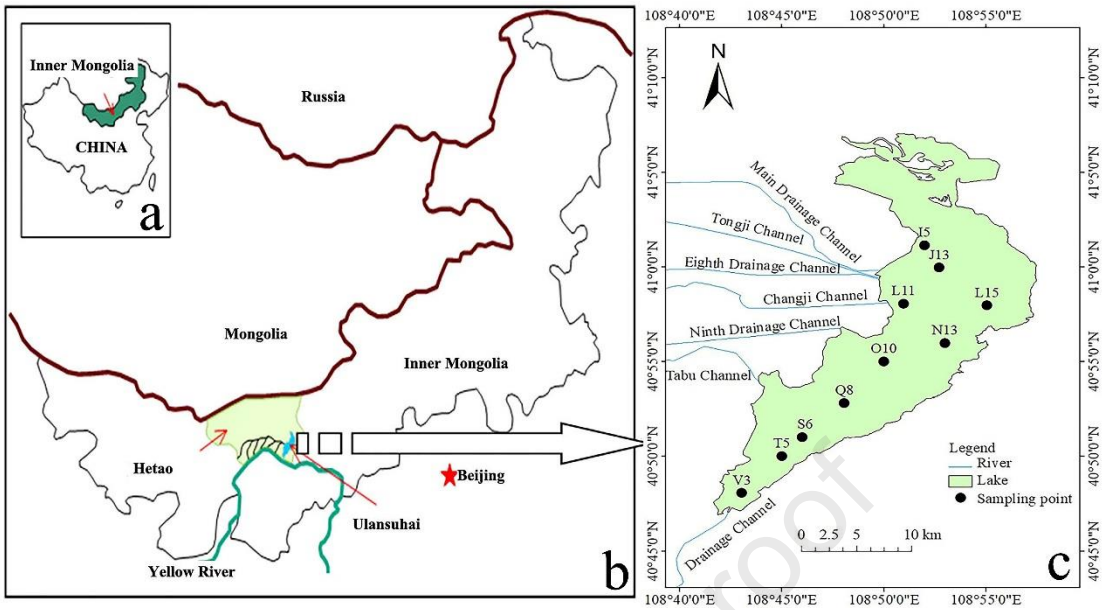

753

754 **Fig. 1**

755

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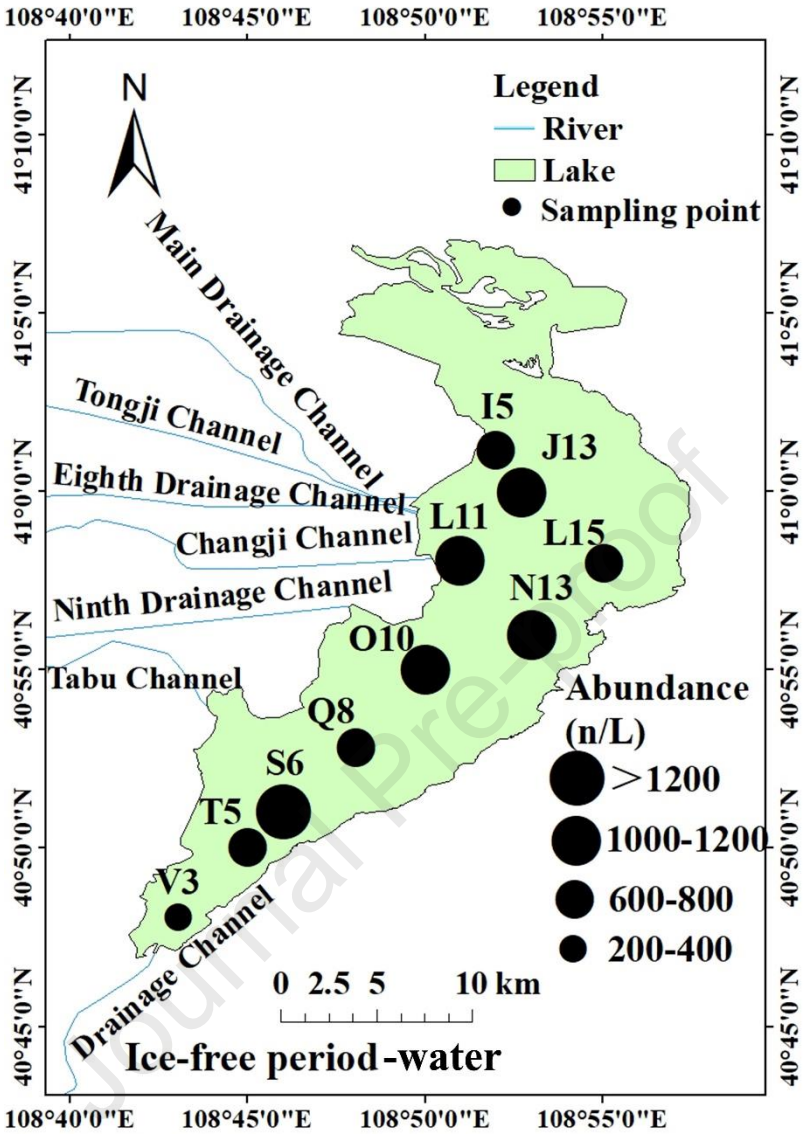
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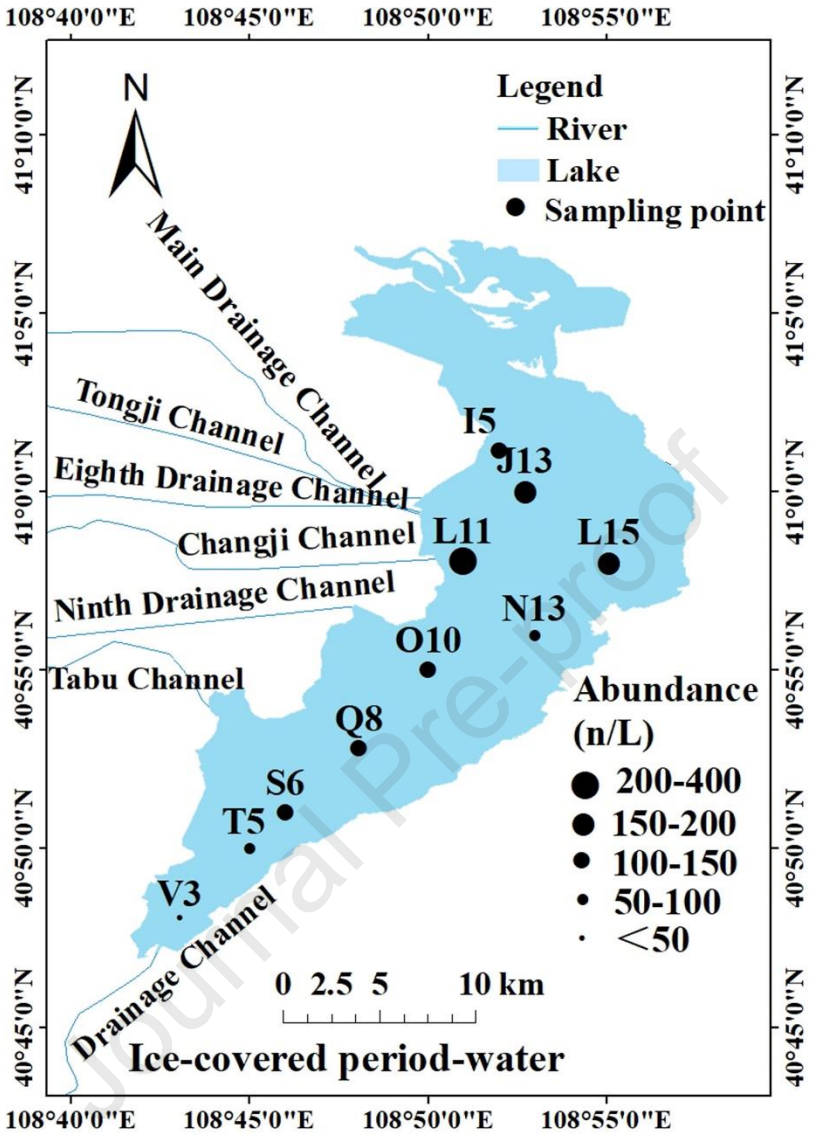
40°55'0"N
40°50'0"N
40°45'0"N
N
Legend
River
Lake
Sampling point
Main Drainage Channel
Tongji Channel
Eighth Drainage Channel
Changji Channel
Ninth Drainage Channel
Tabu Channel
Drainage Channel
I5
J13
L11
L15
N13
O10
Q8
S6
T5
V3
Abundance (n/L)
>1200
1000-1200
600-800
200-400
0 2.5 5 10 km
Ice-free period-water

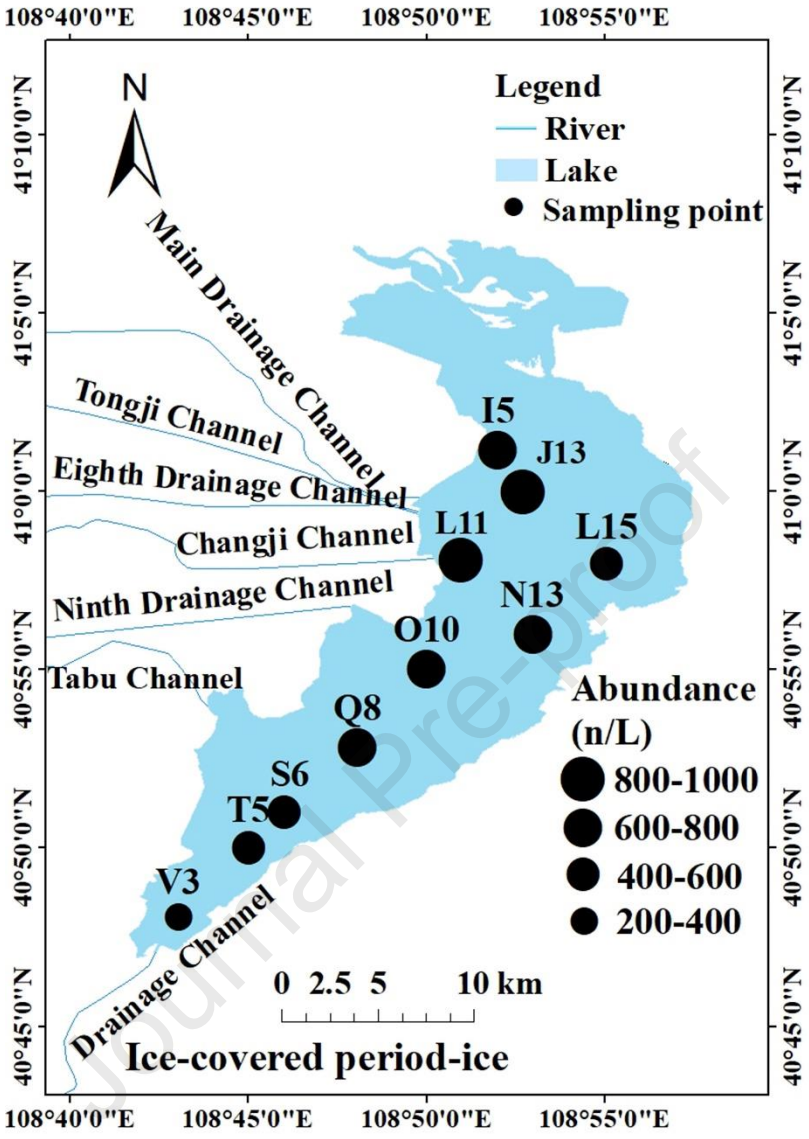
Fig. 2



759

760 **Fig. 3**

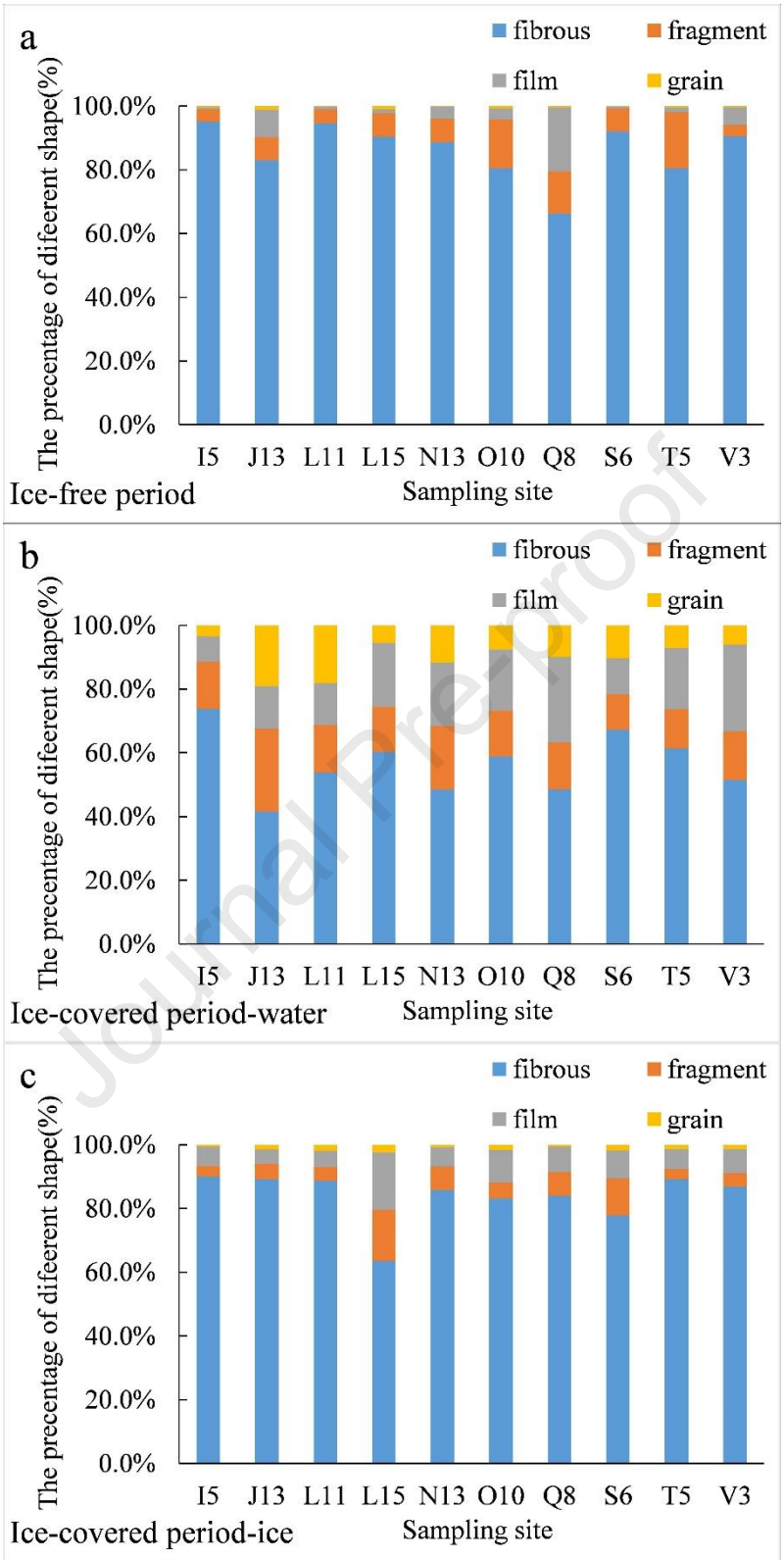
761



762

763 **Fig. 4**

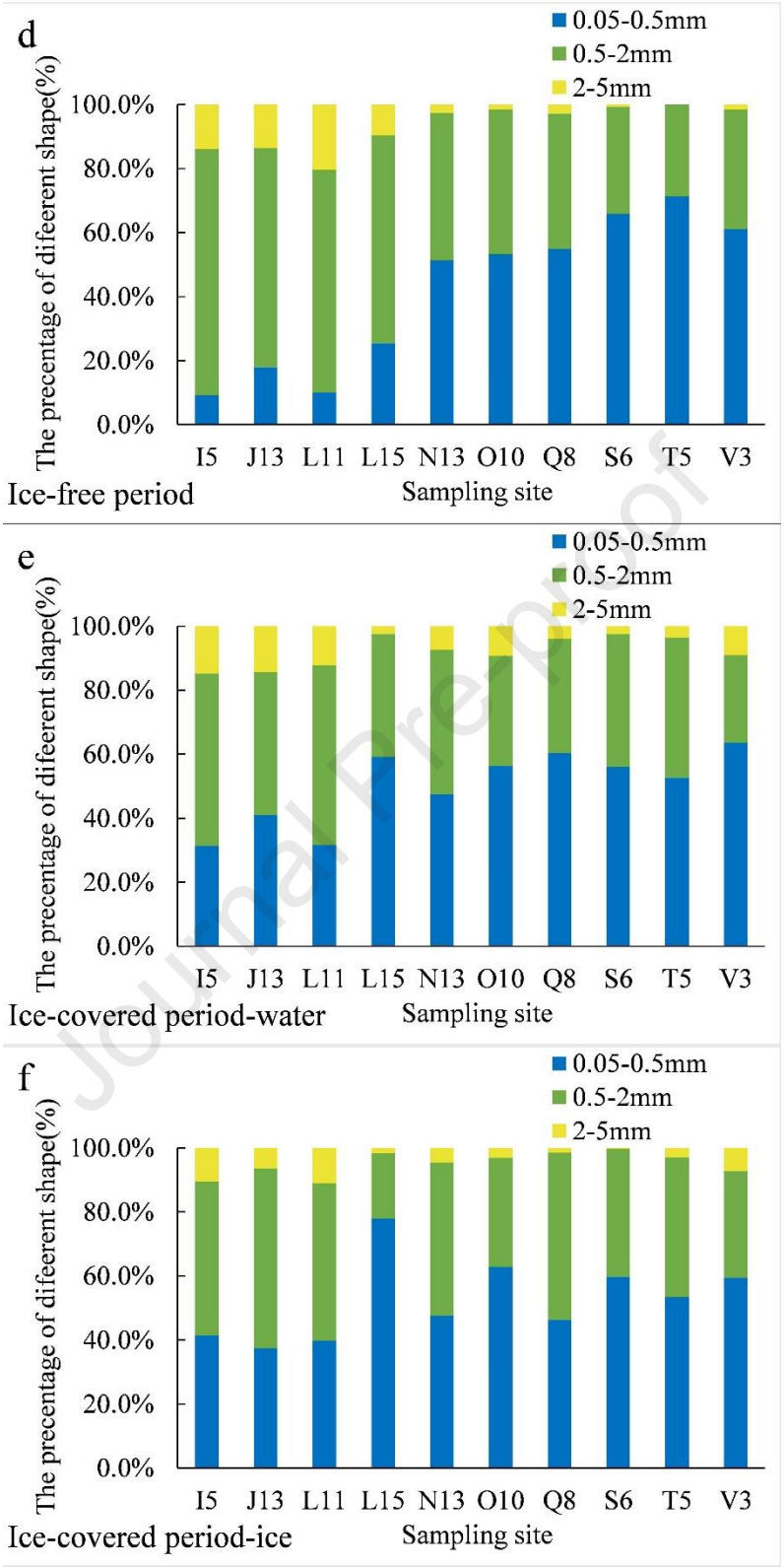
764



765

766 **Fig. 5**

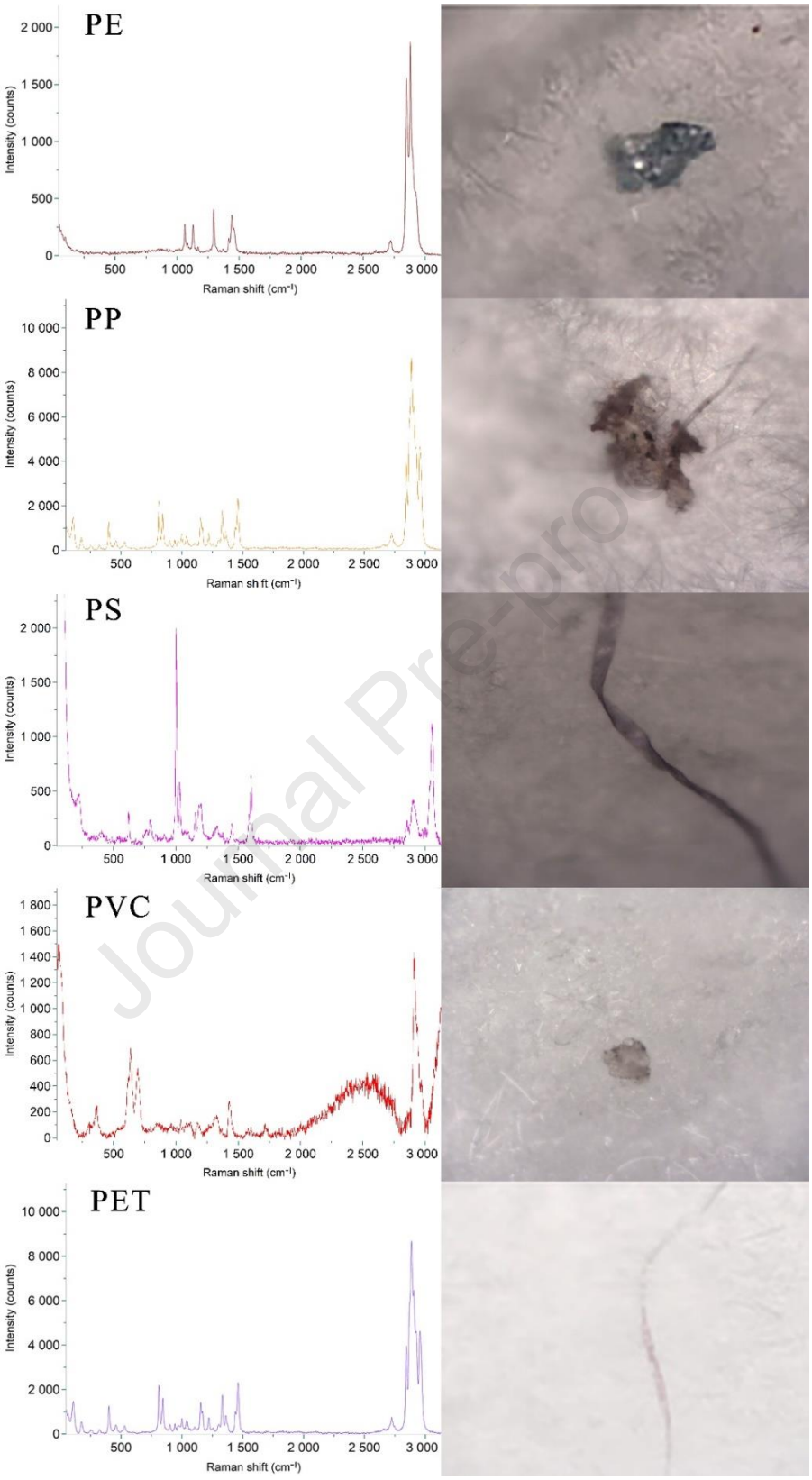
767



768

769 **Fig. 6**

770



771

772 **Fig. 7**

Highlights

- MP abundance was 4.5–11.3 times higher in ice-free than ice-covered lake water
- MP abundance in ice was 2.74–8.14 times higher than in the underlying water
- The spatial distribution of MPs in lake waters differed seasonally
- Seasonal ice formation significantly affected the relative shapes of MPs in water
- Seasonal ice significantly affected the spatial distribution of small MPs in water

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Author Statement

Yu Liu: Conceptualization, Methodology, Validation, Investigation, Writing - Original Draft, Writing - Review & Editing

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Sheng Zhang: Project administration

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Wenbao Li: Investigation

Biao Sun: Validation

Shengnan Zhao: Formal analysis

Dingwen Yao: Investigation, Validation, Resources

Jussi Huotari: Writing - Review & Editing

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Declaration of interests

☒ 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.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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