Macro- and microplastic pollution in shore and bed sediments of limnic ecosystems – Case study in the model catchment of Lake Tollense, Mecklenburg-Western Pomerania, Germany

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Abstract

Environmental pollution by plastics is a major impact of human activity on earth. Increasing plastic production rates, mismanagement of plastic waste and longevity of this material intensified the problem in the last decades. Nowadays, plastic application covers a broad spectrum ranging from everyday objects to specialized equipment in medicine. As a result, input pathways for plastics into the environment and their respective impacts in ecosystems are manifold. Social, economic, and ecologic consequences arise from the environmental plastic pollution. The latter are predominantly entanglement of organisms in plastic items and ingestion of plastic particles by organisms. Due to these adverse effects, plastic pollution in different ecosystems and ecosystem compartments needs to be addressed in research.

Research on plastic pollution increased in the last decades mainly focusing on marine environments. Plastic debris accumulates in marine basins but is transported to the oceans by rivers. Lakes may serve as (temporary) sinks on this way. As a result, comprehensive data recognizing plastic pollution in lakes is necessary to increase data reliability and improve knowledge on underlying processes for plastic input, transport, and retention mechanisms. Only recently, microplastic research in freshwaters increased and lakes around the world have been investigated with respect to their plastic pollution in different ecosystem compartments. However, comparison between existing studies is hampered by the diversity in methods concerning sampling and laboratory analyses. The further development of methods in plastic research requires harmonization of protocols and/or the possibility to compare between different approaches through systematic conversions.

In this context, the presented PhD project is to be classified as part of the project MICROLIM, which analyzes microplastic concentrations at Lake Tollense, Mecklenburg-Western Pomerania, Germany. The major aims of the overall project are to budget microplastics in the model catchment of Lake Tollense and to gain information on its sources, transport pathways, and sinks. Therefore, different lake compartments are investigated concerning their plastic pollution. This PhD project thereby considers plastic pollution in the pedosphere. Lakeshore and lakebed sediments were sampled and analyzed with respect to their microplastic (<5 mm) abundances and a monitoring of anthropogenic litter (≥5 mm including macroplastics) at Lake Tollense shores was conducted, each on a semi-annual basis over a two- or three-year period, respectively. At the same time, applied methods were evaluated to contribute to the method development in plastic research.

Two microplastic identification approaches, relying on Nile Red staining and varying in resolution, were assessed with respect to their comparability. The lower resolution UV light photobox approach has proven suitable for the analysis of large microplastic particles (>630 µm). It is easy to implement, inexpensive, and broadly applicable. Therefore, this approach, in combination with a provided computational conversion of results towards the high-resolution method, qualifies for the deployment in microplastic monitoring procedures, where large sample numbers and volumes need to be analyzed in a short time. In parallel, first tests showed that the application of unmanned aerial vehicles can facilitate macroplastic, or in general anthropogenic litter monitoring at lakeshores. However, further improvements in remote image analysis are necessary to apply the approach on a regular basis.

The investigation of Lake Tollense shore and bed sediments verified a ubiquitous pollution by plastics resulting in mean abundances of 0.2 ± 0.1 anthropogenic litter items per m² lakeshore area and 1,410 ± 822 microplastic particles per kg dry sediment at the lakeshore as well as 10,476 ± 4,290 microplastics per kg dry sediment at the lakebed. The analysis of distribution patterns across the lake, between lakeshore segments of varying exposition and use intensities, and on the small-scale level, within lakeshore segments, showed a predominantly similar dispersion of large and small plastic items and particles. Nevertheless, factors influencing the input and distribution of anthropogenic litter and microplastics differed noticeably. In synthesis of both categories, a complex network of influencing factors and mechanisms was determined for the input and dispersion of plastics in sediments at Lake Tollense. This comprises lake related conditions (i.a. hydrology and sedimentary characteristics), external environmental influences (i.a. meteorologic and biotic factors), and human activity (i.a. population density, recreation, agriculture). Furthermore, this study verified the retention capability for microplastics by lake sediments.

Study results largely correspond to outcomes of former studies considering (converted) concentrations of anthropogenic litter and microplastics and especially controlling factors for their occurrence and distribution. The data obtained for the pedosphere by this study, combined with data on microplastic pollution of other spheres at Lake Tollense within the greater framework of the MICROLIM project, supports a more reliable determination of sources, pathways, sinks, and influencing factors for the plastic pollution at lakes in general. Furthermore, the enhancement in data availability is essential for future risk assessments of plastic pollution in limnic ecosystems.

Zusammenfassung

Die Umweltverschmutzung durch Plastik ist eine wesentliche Auswirkung menschlichen Handels auf der Erde. Steigende Plastikproduktionsraten, Missmanagement in der Plastikmüllverarbeitung und die Langlebigkeit des Materials haben das Problem in den letzten Jahrzehnten verschärft. Der Einsatz von Plastik deckt heutzutage ein breites Spektrum von Alltagsobjekten bis hin zu spezialisierten medizinischen Anwendungen ab. Dementsprechend vielfältig sind auch die Eintragspfade für Kunststoffe in die Umwelt sowie ihre jeweiligen Auswirkungen in den Ökosystemen. Aus der Umweltverschmutzung durch Plastik ergeben sich soziale, ökonomische und ökologische Folgen. Letztere umfassen insbesondere das Verfangen von Organismen in Plastikobjekten und die Aufnahme von Plastikpartikeln durch Organismen. Aufgrund dieser nachteiligen Auswirkungen besteht die Notwendigkeit, die Plastikverschmutzung in verschiedenen Ökosystemen sowie deren Kompartimenten zu erforschen.

Die Forschung zur Plastikverschmutzung hat in den letzten Jahrzehnten, mit einem Fokus auf die marine Umwelt, zugenommen. Plastikmüll sammelt sich in marinen Becken, wobei der Transport in die Ozeane über Flüsse erfolgt. Seen können auf diesem Weg als (temporäre) Senken dienen. Folglich werden umfassende Daten benötigt, die die Plastikverschmutzung in Seen bilanzieren, um die Verlässlichkeit der Daten zu erhöhen und das Wissen bezüglich zu Grunde liegender Prozesse für Plastikeinträge, -transport und -akkumulation zu verbessern. Erst kürzlich hat die Erforschung von Süßgewässern zugenommen und Seen auf der ganzen Welt wurden im Hinblick auf ihre Plastikbelastung untersucht. Studien betrachteten dabei unterschiedliche Kompartimente, unter anderem die Pedosphäre. Ein Vergleich zwischen verschiedenen Studien ist durch eine Vielfalt unterschiedlicher Methoden für die Probenahme und Laboranalyse erschwert. Die weitere Entwicklung von Methoden zur Plastikuntersuchung erfordert eine Harmonisierung von Methodenprotokollen und/oder Möglichkeiten zur Umrechnung zwischen verschiedenen Ansätzen.

In diesen Kontext ist das vorliegende Promotionsprojekt als Teil des umfassenderen MICROLIM-Projekts einzuordnen. Das Projekt MICROLIM untersucht Mikroplastikkonzentrationen am Tollensesee in Mecklenburg-Vorpommern, Deutschland. Die wesentlichen Ziele des Gesamtprojekts bestehen in der Stoffhaushaltsbilanzierung von Mikroplastik im Modelleinzugsgebiet des Tollensesees sowie der Informationsgewinnung bezüglich dessen Quellen, Transportwegen und Senken. Um dies zu erreichen, wird die Plastikverschmutzung in unterschiedlichen Kompartimenten des Sees untersucht. Das Promotionsprojekt beschäftigte sich

dabei mit der Plastikverschmutzung in der Pedosphäre. In einem halbjährlichen Rhythmus wurden über die Dauer von zwei bzw. drei Jahren Sedimentproben am Seeufer und vom Seegrund entnommen und im Hinblick auf ihre Mikroplastikkonzentrationen (<5 mm) untersucht sowie ein Monitoring an Strandabschnitten bezüglich anthropogener Abfälle (≥5 mm, welche Makroplastik integrieren) durchgeführt. Gleichzeitig wurden die angewandten Methoden evaluiert, um zur Methodenentwicklung in der Plastikforschung beizutragen.

Zwei Methoden zur Mikroplastikidentifizierung, die auf Färbung mit Hilfe von Nilrot basieren und sich hinsichtlich ihrer Auflösung unterscheiden, wurden in Bezug auf ihre Vergleichbarkeit untereinander getestet. Die Methode mit geringerer Auflösung, die UV-Licht Photobox, zeigte sich als geeignet für die Analyse größerer Partikel (>630 µm) und qualifiziert sich durch eine leichte Implementierung, geringe Kosten und vielfältige Anwendbarkeit. In Kombination mit der bereitgestellten Umrechnung zur Anpassung der Ergebnisse an die hochauflösende Methode, eignet sich dieser methodische Ansatz für den Einsatz in Monitoringverfahren, bei denen große Probenanzahlen bzw. -volumina in kurzen Zeiträumen analysiert werden müssen. Parallel zeigten erste Tests, dass das Monitoring von Makroplastik, bzw. allgemeiner von anthropogenem Abfall, an Stränden durch den Einsatz von Drohnen erleichtert werden kann. Weitere Verbesserungen in der fernerkundungsbasierten Bildauswertung sind jedoch nötig, um den methodischen Ansatz regelmäßig anwenden zu können.

Die Untersuchung von Seeufer- und Seegrundsedimenten am Tollensesee ergab eine omnipräsente Belastung mit Makro- und Mikroplastik, die sich in Häufigkeiten von im Mittel 0,2 ± 0,1 anthropogenen Müllobjekten pro m² Uferrandabschnitt und 1.410 ± 822 Mikroplastikpartikeln pro kg Trockensediment am Seeufer, sowie 10.476 ± 4.290 Mikroplastikpartikeln pro kg Trockensediment am Seegrund ausdrückt. Die Analyse der Verteilungsmuster innerhalb bzw. entlang des Sees, zwischen Uferrandabschnitten unterschiedlicher Exposition und Nutzungsintensitäten, sowie auf kleinräumiger Ebene, innerhalb der Ufersegmente, zeigte eine überwiegend ähnliche Streuung von großen und kleinen Plastikobjekten und -partikeln. Dennoch unterschieden sich die Einflussfaktoren für den Eintrag und die Verteilung von anthropogenem Abfall und Mikroplastik. In der Synthese beider Kategorien konnte ein komplexes Wirkungsgefüge von Einflussfaktoren und Mechanismen für den Eintrag und die Ausbreitung von Plastik in Sedimenten des Tollensesees ermittelt werden. Dieses beinhaltet seebezogene Einflüsse (u.a. Hydrologie und Sedimenteigenschaften), externe Umweltfaktoren (u.a. meteorologische und

biotische Faktoren) und menschliches Handeln (u.a. Bevölkerungsdichte, Freizeitbeschäftigungen, Landwirtschaft). Darüber hinaus wies die Studie das Retentionspotenzial für Mikroplastikpartikel in Seesedimenten nach.

Die Ergebnisse der Studie entsprechen im Hinblick auf (umgerechnete) Konzentrationen für anthropogene Abfälle sowie Mikroplastik und insbesondere auf die Kontrollfaktoren für deren Auftreten und Verteilung weitgehend Befunden früherer Studien. Eine Kombination der in dieser Studie für die Pedosphäre gewonnenen Daten mit den Daten der Mikroplastikbelastung anderer Umweltkompartimente am Tollensesee im größeren Rahmen des MICROLIM Projektes wird eine verlässlichere Bestimmung von Quellen, Pfaden, Senken und Einflussfaktoren für die Plastikbelastung in Seen ermöhlichen. Außerdem ist die Verbesserung der Datenverfügbarkeit essenziell für zukünftige Bewertungen von Risiken, die von der Plastikbelastung limnischer Ökosysteme ausgehen.

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List of abbreviations and symbols

$3Na_2WO_4 \cdot 9WO_3 \cdot H_2O$	Sodium polytungstate
AL	Anthropogenic litter
$CaCl_2$	Calcium chloride
DFG	Deutsche Forschungsgemeinschaft
DGPS	Differential Global Positioning System
FTIR	Fourier-transform infrared spectroscopy
H_2O_2	Hydrogen peroxide
HCl	Hydrochloric acid
HNO_3	Nitric acid
K(HCOO)	Potassium formate
KOH	Potassium hydroxide
$Li_2O_{13}W_{4\text{-}24}$	Lithium metatungstate
LiDAR	Light detection and ranging
MPSS	Microplastic Sediment Separator
MSFD	Marine Strategy Framework Directive
NaCl	Sodium chloride
NaClO	Sodium hypochlorite
NaI	Sodium iodide
NaOH	Sodium hydroxide
OSPAR	Oslo and Paris conventions
PA	Polyamide
PE	Polyethylene
PET	Polyethylene terephthalate
PP	Polypropylene
Py-GC-MS	Pyrolysis gas-chromatography mass-spectrometry
UAV	Unmanned aerial vehicle
UNEP	United Nations Environment Programme
UV	Ultraviolet
WWTP	Wastewater treatment plant
$ZnCl_2$	Zinc chloride

1. Introduction

Impacts of human activity on earth are manyfold which gave occasion to assign a new geological age - the "Anthropocene" (Crutzen and Stoermer, 2000). Environmental pollution is one of multiple results of human activity caused by the introduction of substances or energy into the environment where these act crucially hazardous towards organisms and ecological systems and necessitate the evaluation of quantity and effect of pollutants (Holdgate, 1979). In this respect, plastic materials introduced into the environment are considered a kind of environmental pollution. Environmental plastic pollution is raising concern and is considered a major problem which requires intensive research (Wright and Kelly, 2017; GESAMP, 2015, 2010; Thompson et al., 2009; UNEP, 2005). Incorporated into soils, plastics can form clearly defined layers, serving as a marker horizon of the Anthropocene (Corcoran et al., 2018; Zalasiewicz et al., 2016). Some authors even refer to a "Plasticene" as large volumes of plastics were and are produced and introduced into the environment since the 1950s (e.g., Campanale et al., 2020; Reed, 2015).

Plastic materials are composed of polymers. The term polymer derives from the Greek word referring to "many" and describes large molecules that are made of many repeated and by polymerization bonded monomers (Ehrenstein, 2001). Nowadays, these monomers are mainly manufactured from fossil-based materials, predominantly oil and gas (Brandsch and Piringer, 2008). Natural polymers were used by humans even before the beginning of the 20th century (Brydson, 1999). Bakelite was the first synthetic plastic discovered by Leo Hendrik Baekeland in 1909 (Corcoran et al., 2018). From the 1930s on, the production and variety of synthetic polymers steadily increased as more polymers were invented (Brydson, 1999). By 2019, plastic production rates reached a sum of 368 million tons (PlasticsEurope, 2020). Chemical attributes of polymers differ and influence characteristics of the final plastic product (e.g., McKeen, 2017; Ehrenstein, 2001). For example, the polymer crystallinity affects the specific density of plastics (McKeen, 2017) which is of importance for plastic materials introduced into the environment as it controls their flotation and/or settlement which in turn effects the degradation (Lambert and Wagner, 2018). Polymer characteristics are further modified to match specialized application requirements which is typically achieved by blending of polymers or by additives (Lambert and Wagner, 2018; Biron, 2016). Common additives include fillers for reinforcement, stabilizers to reduce ultraviolet (UV) degradation, plasticizers to increase flexibility, and the addition of color (McKeen, 2017). The diversity of synthetic polymers in combination with various available additives allows a broad application range for plastic products. Furthermore, favorable

characteristics such as their light weight, durability, and corrosion resistance, in combination with low costs, made plastic deployment common in multiple fields spanning from everyday life to various industrial branches to medicine (PlasticsEurope, 2020; Ryan, 2015; Ehrenstein, 2001).

From the variety of application segments, plastic packaging accounts for nearly 40% of the plastic demand in Europe (PlasticsEurope, 2020). Even though the single use of plastic packaging products decreased in the last decades, around one-fifth still ended up in European landfills in 2018 (PlasticsEurope, 2020). Landfills, and in general insufficient waste management as well as untreated wastewater, can be considered as terrestrial input pathways for plastics (Barnes et al., 2009; UNEP, 2005). Terrestrial sources further include traffic, population-related inputs, industry as well as agriculture and construction (Bertling et al., 2018). As plastic products are ubiquitously utilized, degradation of plastic items, abrasion, and accidental or purposeful loss may lead to plastic pollution in proximity to these activities (Bertling et al., 2018; Lambert et al., 2014; Barnes et al., 2009; UNEP, 2005). In contrast, marine plastic inputs result from shipping and fishing activities as well as from offshore oil and gas extraction platforms and aquaculture (Fig. 1; GESAMP, 2010; UNEP, 2005; Pruter, 1987). Considering their source, small plastic particles, so called microplastics (<5 mm; Arthur et al., 2009), are further classified into primary and secondary microplastics. Primary microplastics are produced in the respective size range. They may be preproduction pellets, textile fibers or abrasion particles originating from personal care products or industrial sandblasting (Andrady, 2011). Contrastingly, secondary microplastics emerge from the fragmentation of larger plastic items due to degradation induced by abiotic and biotic factors (Lambert et al., 2014).

Once in the environment, plastics will not remain stationary but can be transported to other environmental compartments (Fig. 1). Wind can induce transport on land, depending on wind velocity, microplastic particle size, shape, and landcover (Allen et al., 2019; Rezaei et al., 2019; Lambert and Wagner, 2018). Surface runoff transports plastics into streams and rivers where they move with the general flow depending on flow conditions and morphology of the stream (Lambert and Wagner, 2018; Horton et al., 2017a; Nizzetto et al., 2016). Two- and three-dimensional currents as well as wind-driven currents and Stokes drift can transport plastics within lakes and oceans (Isobe et al., 2014; Law et al., 2014, 2010; Eriksen et al., 2013). Wind and wave activity, as well as tidal inundation result in the wash up of plastics at shores where they accumulate and may be remobilized (Smith and Markic, 2013; Barnes et al., 2009; UNEP, 2005;

Debrot et al., 1999). The specific plastic particle density determines whether it floats or submerges in water bodies. Densities for common polymers range between 0.85 g/cm³ for polypropylene (PP) and 1.39 g/cm³ for polyethylene terephthalate (PET; Lambert and Wagner, 2018). The formation of hetero-aggregates and biofilms can increase densities and can cause even low-density polymers to partially or completely sink within the water column (Woodall et al., 2014; Lobelle and Cunliffe, 2011; Morét-Ferguson et al., 2010). Flood events can resuspend particles settled in river and shore sediments, and enable further transport (Hurley et al., 2018b; Nizzetto et al., 2016), whereas the accumulation in deep ocean sediments is considered as long-term sink for environmental plastics (Rochman, 2018; Woodall et al., 2014).

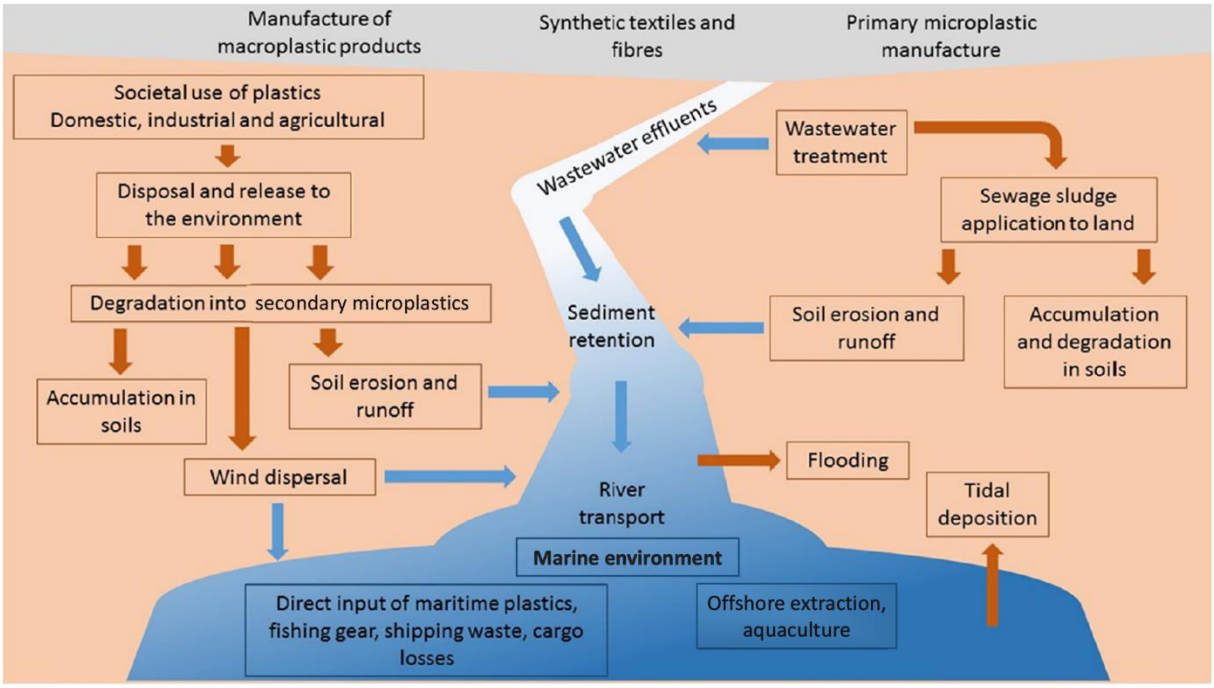

Figure 1: Input and transport pathways for microplastics in the environment according to Horton et al. (2017b) (modified).

Organisms may interact with plastics in the environment, which can cause entanglement and ingestion (GESAMP, 2015; Kühn et al., 2015; UNEP, 2005; Derraik, 2002). The entanglement of different species within larger plastic items has widely been demonstrated (e.g., Ryan, 2018; Allen et al., 2012; Laist, 1997). Especially discarded nets and fishing gear cause entanglement, often labeled as ghost fishing (Galgani, 2015; Gregory, 2009; Breen, 1990), and may result in restrictions in mobility, impairment of health and/or death (GESAMP, 2010; Laist, 1997). Several studies further verified the ingestion of plastic particles by species of various size and habitats, considering either environmental samples (e.g., Su et al., 2018; Foekema et al., 2013; Laist, 1987) or laboratory exposure experiments (e.g., Scherer et al., 2017; Cole et al., 2013; Browne et al., 2008). The latter showed that ingestion of plastic particles can lead to reduced

food consumption, weight and fecundity, and may result in the translocation of smallest particles, inflammatory effects and/or death (Naidoo and Glassom, 2019; Besseling et al., 2014, 2013; von Moos et al., 2012; Browne et al., 2008). Toxic effects are not only related to the plastic particle itself but are further evoked by additives and absorbed persistent organic pollutants (Gunaalan et al., 2020; Lambert et al., 2014; GESAMP, 2010; Teuten et al., 2009). However, concentrations used in laboratory experiments often exceeded environmental concentrations and thus, studies may not provide environmentally realistic consequences (Scherer et al., 2018; Lenz et al., 2016). The transport of ingested plastic particles into species of higher order as well as the uptake of microplastics via drinking water or inhalation also poses a threat to human beings (e.g., Carbery et al., 2018; Prata, 2018; Revel et al., 2018; Rochman et al., 2015). But, research considering impacts of plastics and associated chemicals on the human metabolism is rare, so far (Prata et al., 2020; Revel et al., 2018; Galloway, 2015). Initial studies verified the presence of plastic particles in humans for example in the intestine (Schwabl et al., 2019) and placenta (Ragusa et al., 2021). Adverse effects to human health in form of irritation, oxidative stress, and inflammation have been demonstrated (e.g., Hwang et al., 2020; Schirinzi et al., 2017; Warheit et al., 2001), which are, again, often related to plastic additives and absorbed chemicals (Galloway, 2015; Thompson et al., 2009; Araújo et al., 2002).

Environmental plastic pollution does not only result in described ecological consequences but in social (e.g., decline of recreational value due to reduction in aesthetic of recreational areas) and economic effects (e.g., increased costs due to decline in tourism) as well (Gregory, 2009). The relevance and need of studying plastic occurrence and impact in all environmental compartments is underlined by the manifold consequences evoked by the pollution, which in turn emphasizes the need for plastic contamination prevention in the environment (Galgani, 2015). Plastic pollution increased due to multiple factors, such as the low degradation rates of this material and consequently long durability, ongoing non-sustainable use as well as inadequate and insufficient waste management (Barnes et al., 2009) in combination with the increasing plastic production rates (PlasticsEurope, 2020). Simultaneously, research interest in environmental plastic pollution steadily increased in the last decades (Lambert and Wagner, 2018; GESAMP, 2015; Wright et al., 2013; Thompson et al., 2004). So far, research studies focused on plastic pollution in marine habitats, whereas freshwater ecosystems were less represented (Blettler and Wantzen, 2019; Blettler et al., 2018). As plastics are introduced into marine environments by i.a. terrestrial freshwater drainage (Wagner et al., 2014; GESAMP, 1990; Pruter, 1987), the investigation of inland waters concerning their plastic pollution is of similar im-

portance. Recently increasing numbers of studies investigating lakes or rivers show the ubiquitous pollution of these waterbodies even in remote areas (Zhang et al., 2016; Free et al., 2014) and underline the necessity to quantify and analyze plastic pollution and coherent factors in freshwater ecosystems.

The MICROLIM project, "Matter budget of microplastics in limnic ecosystems: sources, flow paths and sinks of microplastic particles in the model catchment area of Lake Tollense, Mecklenburg-Western Pomerania", considers the plastic pollution in limnic ecosystems. The project, funded by the Deutsche Forschungsgemeinschaft (DFG), involves the analysis of plastic pollution in different compartments, namely hydrosphere, pedosphere, atmosphere, and biosphere at the example of Lake Tollense, Mecklenburg-Western Pomerania, Germany. The project aims to quantify macro- and microplastics in the ecosystem and to analyze underlying processes. The presented PhD project, which is part of the overall MICROLIM project, contributes to the development of methods in macro- and microplastic research and covers the analysis of plastic occurrence in the pedosphere. In a first step, methodical evaluations concerning the identification of microplastics in environmental samples and the remote analysis of macroplastics on lakeshores was implemented as standard protocols in microplastic research are missing (Ivleva et al., 2017; Galgani, 2015; Van Cauwenberghe et al., 2015). The main focus was on the quantification of macro- and microplastics in lakeshore and lakebed sediments from Lake Tollense and in tracing transport and accumulation mechanisms. The prime objectives were to provide data on plastic pollution in lake sediments and to increase the knowledge on factors influencing the input and fate of plastics in sediment compartments of limnic ecosystems.

The thesis is presented in a cumulative structure. It is based on three individual publications in peer-reviewed journals and includes a joining introduction and a comprehensive discussion. In the following, the state of research in plastic pollution research is presented in chapter 2. A more detailed description of the MICROLIM project in general and the PhD project in particular is given in chapter 3, outlining the study objectives. Chapter 4 presents the three individual publications and personal contributions to these. In a comprehensive way, the publications are discussed in chapter 5, followed by an overall conclusion and perspective for future research. A description of materials and methods is excluded from the thesis itself as these are described in detail in the respective publications which can be found in the appendix.

2. Plastic pollution research – State of research

In the early 1970s, first findings of plastics in marine environments were published (Carpenter et al., 1972; Carpenter and Smith, 1972). Plastic pollution research became more important since then, especially as of the early 2000s, reflected by a strong increase in publications (Ivleva et al., 2017; Ryan, 2015). Pollution by large plastic items was in focus in early studies, but research interest gradually turned towards smaller plastic particles in the environment (Koelmans et al., 2015; Thompson et al., 2004). Size classifications of environmental plastics commonly comprise nano-, micro-, meso-, and macroplastics. However, no unique definitions are available for these terms (Hartmann et al., 2019). Common upper size limits for microplastics are 5 mm (Arthur et al., 2009) and the more intuitive 1 mm (Andrady, 2015; GESAMP, 2015). The lower limit for microplastics can be drawn at 1 µm, which simultaneously is the upper limit for nanoplastics (GESAMP, 2015; Browne et al., 2007). The term mesoplastics is infrequently used but often refers to plastic particles between 5 mm and 25 mm whereas macroplastic items are defined as items larger than 5 mm or 25 mm (Hartmann et al., 2019; GESAMP, 2015; Koelmans et al., 2015; Wagner et al., 2014; MSFD, 2013; Browne et al., 2007). Microplastics may be categorized according to their shape into fragments/irregular particles, filaments/fibers, films, granules/spheres, foams, and pellets (Hartmann et al., 2019; Hidalgo-Ruz et al., 2012). Regardless of the size and shape of particles, studies investigated plastic pollution in various environmental compartments. These considered atmospheric deposition (e.g., Klein and Fischer, 2019; Dris et al., 2015), ice (e.g., Bergmann et al., 2017; Obbard et al., 2014), sea- and freshwater (e.g., Lenaker et al., 2019; Eriksen et al., 2013), shore and bed sediments (e.g., Cunningham et al., 2020; Thompson et al., 2004), and soils (e.g., Harms et al., 2021; Fuller and Gautam, 2016) as well as plastics taken up by diverse species (e.g., O'Connor et al., 2020; Cole et al., 2013).

An imbalance concerning the number of published studies on plastic pollution in different environments exists. Plastic pollution in and plastic impacts on organisms from marine environments have been more intensively studied compared to freshwater environments (Blettler and Wantzen, 2019; Blettler et al., 2018; Lambert and Wagner, 2018). However, rivers may serve as input pathways for plastics into the oceans (Horton et al., 2017b; Lebreton et al., 2017; Mani et al., 2016; Nizzetto et al., 2016; Gasperi et al., 2014; Wagner et al., 2014) and lakes are expected to be at least temporary storage basins for plastics on this transport pathway (Li et al., 2020; Bordós et al., 2019; Turner et al., 2019; Vaughan et al., 2017; Imhof et al., 2013). This underlines the importance to likewise investigate plastic abundance in freshwaters. Recently,

lakes around the world were investigated concerning their macro- and microplastic pollution increasing the number of studies available for freshwater ecosystems. Plastic concentrations are highly variable when comparing different studies which can be explained by, on the one hand, methodical differences between studies and, on the other hand, by differences in study area and catchment characteristics (Prata et al., 2019a). The following chapters briefly address the variety in methods in plastic pollution research for macro- and microplastic sampling and analysis, respectively. Chapter 2.3 covers the current state of the art for plastic research in lakes summarizing major findings and concentration ranges of recent studies.

2.1. Macroplastic sampling and analysis

In a marine context, several anthropogenic litter (AL) monitoring programs, which include macroplastics, have been developed, e.g., by the United Nations Environmental Programme (UNEP), the Marine Strategy Framework Directive (MSFD), or more regional by the OSPAR (Oslo and Paris) Commission or British Marine Conservation Society (Galgani et al., 2019; Nelms et al., 2017). Partially, monitoring programs incorporate citizen science to reduce costs, increase efficiency, and enlarge spatial coverage of data while simultaneously increasing the public awareness for environmental plastic pollution (Nelms et al., 2017; Zettler et al., 2017; Hidalgo-Ruz and Thiel, 2015). Comparable litter monitoring programs relying on citizen science are less common for lakes and are mainly established at the Laurentian Great Lakes (Adopt-a-Beach^{TM} program, Great Canadian Shoreline Cleanup, Earthwatch Institute's Freshwater Watch; Vincent et al., 2017; Driedger et al., 2015; Hoellein et al., 2015). Similarly, an AL study at Lake Malawi in Africa relied on data collected by volunteers (Mayoma et al., 2019). In contrast, several studies conducted more local investigations at lakes around the world (see chapter 2.3). In this case, trained and experienced staff commonly performed surveys resulting in smaller but more reliable data sets compared to the ones gained by citizen science (GESAMP, 2019; Hidalgo-Ruz and Thiel, 2015). Still, differences in sampling strategies hamper the comparison between results. Whereas some studies investigate litter at limnic shore segments, others even consider intertidal and submerged areas (e.g., Egessa et al., 2020; Ngupula et al., 2014). Studies analyzing shore segments may sample the drift line (e.g., Faure et al., 2015), transects of a specific width (e.g., Blettler et al., 2017; Free et al., 2014; Hoellein et al., 2014), (random) quadrats of a specific area (e.g., Chapman, 2019; Corcoran et al., 2015; Imhof et al., 2013) or the complete area of a segment (e.g., Czarkowski et al., 2016; Hoellein et al., 2015). Commonly, a visual identification and manual collection of AL at shores is performed in the field (e.g., Dalu et al., 2019; Blettler et al., 2017; Free et al., 2014). However, techniques collecting spectral

data can enhance the analysis of litter items by semi-automatic or even automatic remote analysis. For example, image data provided by unmanned aerial vehicles (UAV) or by webcams (e.g., Merlino et al., 2020; Haseler et al., 2019; Kako et al., 2018; Martin et al., 2018) were analyzed with respect to litter occurrence. Ge et al. (2016) also utilized light detection and ranging (LiDAR) for this purpose. Studies mostly concentrated on marine shores whereas the deployment of remote methods was only recently tested at freshwater shores (e.g., Jakovljevic et al., 2020; Geraeds et al., 2019). Even though harmonization efforts have been provided for AL monitoring, the decision for one protocol or the other depends on various factors, like the type of shore, the intention (analyzing standing stock or temporal variations), and time and financial resources (GESAMP, 2019).

Units used for the presentation of macroplastic or AL concentrations at shores depend on data processing and vary between studies, which further prevents a direct comparison (Hoellein et al., 2014). Abundances can be expressed as number of items or its relative frequency without any relation to shore parameters (e.g., Chapman, 2019; Dalu et al., 2019; Ge et al., 2016). Furthermore, studies report items per shore length (e.g., Blettler et al., 2017; Free et al., 2014). To incorporate a second dimension and improve comparability, results are commonly presented as items per area (e.g., Vincent et al., 2017; Czarkowski et al., 2016; Corcoran et al., 2015). Additionally, litter weight can be determined to present concentrations in mass, also related to length or area of the investigated shore segment (e.g., Chapman, 2019; Dalu et al., 2019; Faure et al., 2015).

2.2. Microplastic sampling and analysis

In the following, the description of microplastic sampling and analysis focuses on sediment samples as these were in focus of this PhD study as well. However, laboratory methods for samples of different compartments largely overlap. Microplastic investigations regarding lake sediments can consider lakeshore or lakebed sediments. Studies investigating lakeshore sediments either sampled quadrates of various areas and within different zones of beaches (e.g., Egessa et al., 2020; Xiong et al., 2018; Zhang et al., 2016) or took samples from greater depths via cores of different diameters (e.g., LfU, 2019; Dean et al., 2018; Imhof et al., 2018). Similarly, different devices such as grabs or corers were used for sampling lakebed sediments (Yang et al., 2021; Stock et al., 2019). Sampling area, volume and especially depth, depend on the sampling method chosen (Prata et al., 2019a). After sampling, laboratory processing is necessary for the analysis of microplastics in samples and comprises three major steps: (i) digestion

of biogenic organic material, (ii) segregation of plastic and sediment particles, and (iii) microplastic identification. Applied methods for these steps vary greatly between studies since no standard protocol has been defined yet, which hampers the comparison between results (Ivleva et al. 2017, van Cauwenberghe et al. 2015). Likewise, lower size limits for microplastic particles, often set by additional preprocessing steps like sieving, considerably vary between studies and influence resulting concentrations (Yang et al., 2021; Koelmans et al., 2020; Prata et al., 2019a). Therefore, validated and standardized protocols are highly needed to harmonize methods and resulting concentrations for microplastics (GESAMP, 2019; Prata et al., 2019a). In the following, the summary of applied methods in microplastic analysis focuses on the three named major laboratory steps - digestion, segregation, and identification - despite variances in possible additional preprocessing steps (e.g., drying, sieving, filtering of samples).

Digestion protocols in microplastic analysis

On the one hand, reagents used for digestion in microplastic analysis need to be efficient in degrading biogenic organic material to decrease the amount of interfering material. On the other hand, they must not weaken or even eliminate polymers (Pfeiffer and Fischer, 2020; Lusher et al., 2017). Diverging protocols have been tested based on several chemicals at different concentrations including acids, bases, oxidizers, and enzymes (e.g., Prata et al., 2019a; Stock et al., 2019; Munno et al., 2018). In case of acids, nitric acid (HNO_3) and hydrochloric acid (HCl) were proposed. HNO_3 showed high efficiency in digesting organic material in samples but also degraded specific polymers (Catarino et al., 2017; Avio et al., 2015; Claessens et al., 2013). HCl efficiently digests carbonates only, but does not affect polymers when applied at low concentrations and temperatures (Pfeiffer and Fischer, 2020; Nuelle et al., 2014). For bases, potassium hydroxide (KOH) is preferred over sodium hydroxide (NaOH) as synthetic polymers are more resistant and the efficiency of digestion is still decent (Munno et al., 2018; Dehaut et al., 2016; Foekema et al., 2013). Hydrogen peroxide (H_2O_2) is commonly applied as oxidizing reagent in the digestion process (Hurley et al., 2018a) causing no to little polymer degradation when applied at low concentrations and for longer time frames instead (Frias et al., 2018; Avio et al., 2015; Nuelle et al., 2014). To increase efficiency and reduce reaction time, wet peroxide oxidation with Fenton's reagent was applied, combining H_2O_2, an iron catalyst (Fe(II)) and the addition of heat (e.g., Tagg et al., 2017; Masura et al., 2015). However, Munno et al. (2018) showed that temperatures above 60 °C may degrade microplastics. The application of the oxidizing agent sodium hypochlorite (NaClO) was proposed as well (Collard et al., 2015; Sørensen et al., 2013) and was successfully used in studies in combination with H_2O_2 (e.g., Hengstmann

et al., 2018; Tamminga et al., 2018) or KOH (e.g., Enders et al., 2017; Strand and Taivora, 2016). Furthermore, enzymes have been applied for digestion in biota samples (e.g., Catarino et al., 2017; Löder and Gerdts, 2015; Cole et al., 2014). Their non-toxicity and the absence of degradation effects towards polymers are advantageous (Stock et al., 2019). Disadvantageously, enzyme protocols are laborious and expensive (Thiele et al., 2019; Hurley et al., 2018a; Lusher et al., 2017).

Microplastic separation from sediment samples

For sediment samples, the separation of microplastic and sediment particles is required to improve microplastic identification. Separation techniques are commonly based on the differences in density between sediment (2.65 g/cm³; Hidalgo-Ruz et al., 2012) and polymer particles (<1.4 g/cm³ for raw polymers; Lambert and Wagner, 2018). Therefore, saline solutions of intermediate density are applied, causing the settling of sediment while microplastics stay suspended or float (Quinn et al., 2017; Hidalgo-Ruz et al., 2012). The majority of studies utilized saturated sodium chloride (NaCl) solutions (Prata et al., 2019a). This solution is advantageous due to low costs and low toxicity (Frias et al., 2018; Löder and Gerdts, 2015; Nuelle et al., 2014). However, its relatively low density (~1.2 g/cm³) prohibits the separation of polymers with higher specific densities (Quinn et al., 2017; Van Cauwenberghe et al., 2015). To increase extraction efficiencies, solutions with higher densities like sodium polytungstate ($3Na_2WO_4 \cdot 9WO_3 \cdot H_2O$; 1.4 g/cm³) and sodium iodide (NaI; 1.6-1.8 g/cm³) have been deployed (e.g., Zhao et al., 2015; Nuelle et al., 2014; Claessens et al., 2013; Corcoran et al., 2009). The application of zinc chloride ($ZnCl_2$; 1.5-1.7 g/cm³) and calcium chloride ($CaCl_2$; 1.3-1.35 g/cm³) was successful in extracting microplastic particles from sediment samples as well (e.g., Coppock et al., 2017; Crichton et al., 2017; Stolte et al., 2015; Liebezeit and Dubaish, 2012). Furthermore, Zhang et al. (2016) utilized potassium formate (K(HCOO); ~1.6 g/cm³) whereas Masura et al. (2015) recommended lithium metatungstate ($Li_2O_{13}W_{4\text{-}24}$; ~1.6 g/cm³) for density separation. Even though listed solutions enable the extraction of high-density polymers, they still differ in their extraction efficiencies and costs and toxicity is increased compared to the application of NaCl solutions (Frias et al., 2018; Quinn et al., 2017).

Further approaches for the separation of microplastic and sediment particles, like an air induced overflow system (Nuelle et al., 2014) or an elutriation column (Claessens et al., 2013), still rely on density differences but benefit from a flow direction. Biological research applied the process of elutriation for the separation of low-density meiofauna from high-density sand particles

(Southwood and Henderson, 2000), which originates from an upward fluid or gas stream, typically water (Claessens et al., 2013). Elutriation is a cheap and efficient method but requires preceding sieving of samples (Kedzierski et al., 2016). For microplastic analysis, the principle of elutriation was refined and adjusted (Kedzierski et al., 2017, 2016; Claessens et al., 2013) and successfully implemented (e.g., Hengstmann et al., 2018; Zhu, 2015). Similarly, the Microplastic Sediment Separator (MPSS) uses the upward flow of $ZnCl_2$ solution to separate microplastics from large volumes of sediment (Imhof et al., 2012). Additionally, the separation of microplastic and sediment particles by oil was tested (Lechthaler et al., 2020; Crichton et al., 2017; Karlsson et al., 2017) as well as by magnetism (Grbic et al., 2019). These approaches do not rely on density differences, but on surface characteristics of plastic particles.

Microplastic identification

Early studies on microplastics in the environment commonly performed visual inspection for the final identification of particles (Lusher et al., 2017; Hidalgo-Ruz et al., 2012). Visual inspection is relatively cheap and independent of specialized equipment, but is considered subjective, error prone, and is limited to large particles >1 mm, as identification reliability decreases below this size limit (Prata et al., 2019a; Lusher et al., 2017; Löder and Gerdts, 2015; Hidalgo-Ruz et al., 2012). Still, visual identification, with the naked eye or a microscope, is the most commonly applied method (Prata et al., 2019a). However, chemical analysis of particles is highly recommended and often subsequently conducted to assure proper microplastic identification (Frias et al., 2018; Lusher et al., 2017; Dekiff et al., 2014; Hidalgo-Ruz et al., 2012). For polymer verification, spectroscopic approaches, especially Fourier-transform infrared (FTIR) spectroscopy (e.g., Frias et al., 2018; Löder and Gerdts, 2015; Thompson et al., 2004), or (μ)Raman spectroscopy (e.g., Song et al., 2015; Van Cauwenberghe et al., 2013; Imhof et al., 2012) are commonly applied. These methods use the excitation and detection of molecule vibrations resulting in characteristic FTIR or Raman spectra for polymer identification (Käppler et al., 2016). Major advantageous of spectroscopic approaches are a reliable, non-destructive polymer identification, the possibility to directly measure particles on filters, a low particle size limit in case of μRaman spectroscopy, and that these processes offer a high potential for automatization (Lusher et al., 2017; Käppler et al., 2016; Lenz et al., 2015). Disadvantages are associated with insufficient spectrum qualities when analyzing chemically weathered particles, interfering fluorescence induced by biological sample residues for Raman spectroscopy, large timeframes for analysis, and high costs for devices (Käppler et al., 2016; Löder and Gerdts, 2015; Norén et al., 2014; Imhof et al., 2012; GESAMP, 2010). Besides spectroscopic analyses, studies conducted pyrolysis gas-chromatography mass-spectrometry (Py-GC-MS) for polymer

verification (e.g., Fischer and Scholz-Böttcher, 2019, 2017; Nuelle et al., 2014). The method determines chemical characteristics of single particles or bulk samples related to mass by measuring thermal degradation products, but its destructive mode and missing information on particle numbers limits its application (Prata et al., 2019a; Frias et al., 2018; Fries et al., 2013).

To aid visual inspection and overcome subjective identification, Andrady (2011, 2010) proposed a staining approach using the fluorescent dye Nile Red (9-diethylamino-5H-benzo[α]phenoxazine-5-one). This approach was further developed and evaluated for microplastic identification and relies on the adsorption of the dye to the polar surface of polymers (Nel et al., 2021; Prata et al., 2019b; Stanton et al., 2019; Erni-Cassola et al., 2017; Maes et al., 2017; Tamminga et al., 2017; Shim et al., 2016). Varying details in the staining protocol (i.a. choice of solution, concentrations, incubation time, wavelength), preprocessing (i.a. choice of digestion approach), and particle characteristics (i.a. shape, size, weathering) influence the staining and identification process and can result in differences in staining efficiencies for microplastics and co-staining of residual organic material (Nel et al., 2021; Prata et al., 2019b; Stanton et al., 2019; Erni-Cassola et al., 2017; Maes et al., 2017; Tamminga et al., 2017; Shim et al., 2016). Nile Red staining is inexpensive, readily accessible, and facilitates fast and easy identification of microplastics for large sample numbers (Nel et al., 2021; Miller et al., 2017; Tamminga et al., 2017; Song et al., 2014). Major drawbacks comprise co-staining of residual organic material, the difficulty to detect specific polymers, and missing information on polymer composition still requiring subsequent chemical analyses (Prata et al., 2019b; Stanton et al., 2019; Erni-Cassola et al., 2017; Miller et al., 2017; Tamminga et al., 2017; Shim et al., 2016).

2.3. Current research in limnic ecosystems

The preceding chapters presented the variety in field and laboratory methods for macro- and microplastic analysis in sediment samples without addressing a specific ecosystem as methods largely overlap. In the following, the description concerning the state of the art focuses on limnic ecosystems as a lake was also in focus of this PhD study and results are hardly comparable to other ecosystems due to deviating environmental influences.

Lakes around the world were investigated for their plastic pollution even in remote areas e.g., the Tibetan Plateau or the Alps (Pastorino et al., 2021; Zhang et al., 2016; Free et al., 2014). Studies intensively analyzed the pollution at the Laurentian Great Lakes in North America (e.g., Hendrickson et al., 2018; Ballent et al., 2016; Corcoran et al., 2015) also addressing models concerning plastic sources and transport pathways (e.g., Cable et al., 2017; Hoffman and Hittinger, 2017). Furthermore, freshwater lakes on other continents, in South America, Asia, and

Africa (e.g., Egessa et al., 2020; Yuan et al., 2019; Blettler et al., 2017) were investigated considering their macro- and microplastic concentrations. Concerning Europe, early lake studies analyzed the plastic pollution at Lake Garda (Imhof et al., 2013) and at Swiss lakes (Faure et al., 2012). Recently, European plastic research in freshwaters increased and lakes were analyzed in various countries, including the UK (e.g., Turner et al., 2019), Spain (e.g., Gil-Delgado et al., 2017), Germany (e.g., LfU, 2019), Italy (e.g., Imhof et al., 2018), Finland (e.g., Uurasjärvi et al., 2020), Poland (e.g., Kaliszewicz et al., 2020), Hungary (e.g., Bordós et al., 2019), and Russia (e.g., Zobkov et al., 2020). Research of plastic concentrations in different lake compartments focused on the analysis of water and sediments, followed by biota (Cera et al., 2020). A higher number of studies analyzed microplastic abundances compared to macroplastic concentrations (Schwarz et al., 2019; Blettler et al., 2018). Comprehensive data sets on macroplastics or AL are provided for the Laurentian Great Lakes as beach litter monitoring programs have already been established here (Vincent et al., 2017; Hoellein et al., 2015).

Nearly all lake studies verified ubiquitous pollution with macro- and/or microplastics, but with strong variations in concentrations, independent of the analyzed compartment. Several recent studies investigated microplastic concentrations in lakebed sediments (e.g., Lenaker et al., 2021; Baldwin et al., 2020; LfU, 2019), whereas lakeshore sediments were more frequently investigated in early lake studies (e.g., Faure et al., 2015; Imhof et al., 2013; Faure et al., 2012). The comparison between studies for both types of sediment samples shows a considerable variation in microplastic abundance. For example, Su et al. (2016) detected a maximum of 235 particles/kg sediment in lakebed samples from Taihu Lake whereas Dong et al. (2020) found a maximum concentration of more than 7,700 particles/kg in Donghu Lake bed sediments. Both lakes are located in China. Simultaneously, a more than three times lower mean value for microplastic abundance in shore sediments was reported for Lake Chiusi, Italy, with 112 particles/kg (Fischer et al., 2016) compared to average concentrations in lakes in Finland (396 particles/kg; Scopetani et al., 2019). The difference in microplastic concentrations in lakeshore sediments expressed as particles per area is even greater. Concentrations range, for example, from on average 3,508 to 17,068 microplastic particles/m² for Lake Garda, Italy, and Bavarian Lakes in Germany, respectively (LfU, 2019; Imhof et al., 2018). Similarly, the average abundance of macroplastics or AL at shorelines varies considerably between, for example, 3 items per m² at Lake Zurich, Switzerland (Faure et al., 2015) and up to 483 items per m² at Lake Garda, Italy (Imhof et al., 2013). In terms of mass per area, average concentrations vary between 0.1 g per m² for Lake Lewisville, United States, and 26 g per m² for Lake Maggiore, Switzerland (Chapman, 2019; Faure et al., 2015).

Water surface concentrations are reported in particles per area or particles per volume. For both units, reported concentrations largely differ. Concentrations ranged from $2.0 * 10^4$ to $6.8 * 10^6$ particles/km² or rather 1.9 to $3.4 * 10^4$ particles/m³ for different investigated lakes around the world (Alfonso et al., 2020; Yuan et al., 2019; Su et al., 2016; Free et al., 2014). Only a few studies analyzed microplastics in the water column. Lenaker et al. (2019) reported 10-fold lower maximum concentrations in the water column of Lake Michigan, United States (4.3 particles/m³), compared to abundances at Bavarian lakes, Germany (44 particles/m³; LfU, 2019). Less common studies on microplastic occurrence in biota samples still detected particles in gastrointestinal tracts of different fish species ranging between frequencies of 22% at Lake Ziway, Ethiopia, to 55% at Lake Victoria, Tanzania (Merga et al., 2020; Biginagwa et al., 2016). Furthermore, 44% of all investigated faeces samples from water bird species at Spanish lakes contained microplastics (Gil-Delgado et al., 2017). Concentrations in mussels, for example, ranged between 0.2 and 12.5 particles/g wet weight in Asian clams in Taihu Lake, China (Su et al., 2018).

Large variations in plastic abundances between limnic plastic studies stem from different factors: On the one hand, they are influenced by selected methods for sampling and analysis as described before, on the other hand, variations result from differences in the study area and environmental conditions. A major environmental control for plastics in limnic ecosystems is the input of plastics via tributaries and surface run-off (Zobkov et al., 2020; Lenaker et al., 2019; Dris et al., 2018; Vaughan et al., 2017; Su et al., 2016; Zhang et al., 2016). Furthermore, studies verified environmental factors such as wind, currents, and circulation patterns as impact factors for the spatial and temporal distribution of plastics in lakes (Egessa et al., 2020; Imhof et al., 2018; Ballent et al., 2016; Fischer et al., 2016; Eriksen et al., 2013; Imhof et al., 2013). Lake attributes (e.g., surface area, morphology) and respective substrate characteristics were moreover identified to contribute to the accumulation and dispersion of plastics (Imhof et al., 2018; Ballent et al., 2016; Free et al., 2014; Zbyszewski et al., 2014). Additionally, human activity aspects, including population density, degree of tourism, and the presence of industry and agriculture in the catchment area have to be considered (e.g., Lenaker et al., 2021; Xiong et al., 2018; Wang et al., 2017; Zbyszewski and Corcoran, 2011).

3. Plastic pollution in lake sediments - Case study at Lake Tollense

A case study considering macro- and microplastic pollution was conducted at Lake Tollense, Mecklenburg-Western Pomerania, Germany, since plastic pollution research in freshwaters is inadequately represented. The case study is comprised in a DFG funded project called MICRO-LIM. In the following, the conception and objectives of the MICROLIM project are briefly described before the presented PhD project is integrated into the superior MICROLIM project and is further characterized by defining its specific study objectives. The description of applied methods for sampling and analysis is not thematized in the thesis as these are explained in detail in the individual publications (see chapter 4 and appendix).

3.1. The MICROLIM project

The project MICROLIM deals with the budgeting of microplastics in limnic ecosystems by identifying sources, fluxes, and sinks of microplastic particles in the model catchment of Lake Tollense (project title: "Matter budget of microplastics in limnic ecosystems: sources, flow paths and sinks of microplastic particles in the model catchment area of Lake Tollense, Mecklenburg-Western Pomerania"). As lakes may serve as (temporary) sinks on transport pathways of microplastics from terrestrial sources to the ocean and since lakes show much smaller dimensions and complexity compared to marine ecosystems, they are predestined to study sources, transport mechanisms and retention processes at a smaller scale. On this basis, two major aims were defined for the MICROLIM project: i) Microplastic budgeting in a model lake catchment by quantification and evaluation of fluxes and retention rates for the compartments hydrosphere, pedosphere, atmosphere, and biosphere, and ii) synthesis and modelling of the microplastic budget in limnic catchments. For an exemplary analysis, Lake Tollense in Mecklenburg-Western Pomerania, in the northeast of Germany, was selected (Fig. 2). The following specific lake characteristics were decisive when choosing the study area to facilitate the analysis of relevant processes influencing plastic accumulation and distribution: surface area, size of catchment area, adequate water depth, shape factors, lake morphology, access to bank border segments of relevant expositions, and kind and intensity of utilization. Table 1 presents a selection of characteristics for Lake Tollense.

Mecklenburg-Western Pomerania is largely covered by a young moraine area formed by the last glacial Weichselian period (LUNG MV, 2015). The Tollense basin is situated within this landscape, characterized by ground moraines with proximity to the terminal moraine, and was formed as a tunnel valley by flowing melt waters under the ice cover (STALU MS, 2013). The u-shaped moraines edge the southern part of the basin and partly flank the western and eastern

shore of Lake Tollense (Nixdorf et al., 2004). The study area is located in the temperate climate zone with warm summers and no dry season according to Köppen (1936). An annual mean temperature of 9.1 °C and an annual precipitation of 580 mm was measured at the climate data station closest to Lake Tollense (Trollenhagen, linear distance ca. 12 km) between 1991 and 2020 (DWD, 2021). As a consequence of the geological origin and climatic conditions, clayey sands dominate in the Tollense basin and sand and clay cambisols as well as luvisols and colluvial anthrosols predominantly formed (LUNG MV, 2020, 2015). The potential natural vegetation in the Tollense basin is primarily composed of beech forests with slightly differing peculiarities and small areas of alluvial forests in the south of Lake Tollense (STALU MS, 2013). Circa one fifth of the Lake Tollense catchment area is still covered by forests whereas more than half of the area is utilized as arable land (Nixdorf et al., 2004).

Table 1: Lake characteristics of Lake Tollense according to Nixdorf et al. (2004).

Characteristic	**Specification**
Surface area	17.9 km²
Volume	316 * 10^6 m³
Mean water depth	17.6 m
Maximum water depth	31.2 m
Catchment area size	515 km²
Catchment area land cover	56% agriculture, 23% forest, 6% grassland

Lake Tollense is a dimictic lake, characterized by a simple, sparsely dissected shape aligned in a southwest-northeast direction. Lake Tollense is largely groundwater fed but additionally receives water by tributaries (i.a., Gätenbach, Nonnenbach, Krickower Bach, Wustrower Bach and Liepskanal), which mainly enter the lake in the south (Nixdorf et al., 2004). The only effluent leaves Lake Tollense in the north and ultimately drains into the Baltic Sea after its confluence with the Peene (STALU MS, 2013). Lake Tollense receives drainage from a small wastewater treatment plant (WWTP) via the Lieps nature reserve in the south whereas a larger WWTP directly drains into its effluent. Anthropogenic influence is very distinct at Lake Tollense allowing a comparison concerning utilization prevalence. The city of Neubrandenburg (63,400 inhabitants in 2020; LAiV, 2020) is located at the northern shore resulting in higher population and building densities and increased offers for recreational activities (e.g., fishing, boating, bathing). A nature reserve touches the southern shoreline, which is further characterized by large forest stretches with agricultural areas in the hinterland.

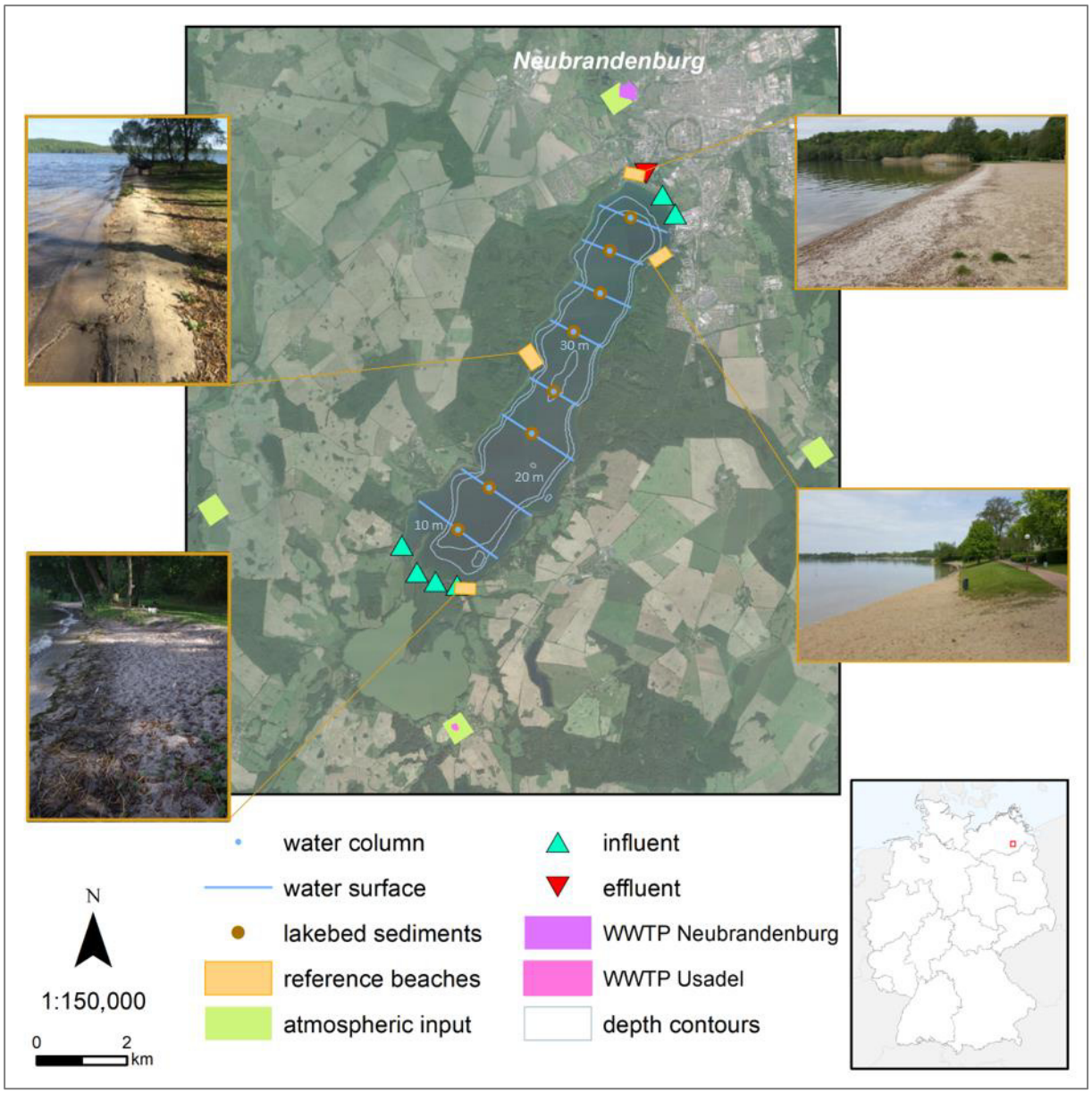

Figure 2: Overview of the study area at Lake Tollense in Mecklenburg-Western Pomerania, Germany, with respective sampling points and areas for different compartments and information on lake hydrology characteristics. Projection: Transverse Mercator. Coordinate system: WGS_1984_UTM_Zone_32N. Satellite image: ArcGIS Imagery ©ESRI. Bathymetry: MLUMV (2017).

The MICROLIM project investigates microplastic occurrence in the Lake Tollense catchment in water surface and water column samples from the lake and its tributaries, in shore sediments at four bank border segments accompanied by macroplastic monitoring, and in sediments from the lakebed, on a semi-annual basis. Additionally, atmospheric deposition is investigated at test plots around Lake Tollense allocated to the four cardinal directions (Fig. 2). Methods used for macro- and microplastic sampling and following laboratory analyses are adapted to the respective compartment samples and were successfully tested and applied in former investigations. Fundamental data (e.g., topographic parameters, aerial images, meteorological parameters, sediment parameters, plastic production and exploitation information) are simultaneously gathered.

3.2. The PhD project and study objectives

This PhD project is integrated into the larger MICROLIM project and considers the investigation of plastic concentrations in the pedosphere at Lake Tollense. It provides data concerning pollution levels, which add, in particular, to the overall balancing of microplastics in the model catchment and, in general, to the greater data set of plastic pollution in lake sediments worldwide. Therefore, AL (≥5 mm; including macroplastic) monitoring as well as microplastic (<5 mm) samplings were conducted semi-annually (March and September) at Lake Tollense shorelines over a three- and two-year period, respectively. Additionally, lakebed sediments were sampled parallel to shoreline sediments to analyze microplastic abundances. Besides the major aim of plastic pollution quantification and evaluation in lake sediments, methodological aspects were considered in the conducted study. In the following, principal research questions in combination with specific sub-questions are phrased and shortly explained, forming the basis of this PhD study.

How can applied methods for the quantification of macro- and microplastics in lake sediments be further enhanced?

Standardized methods are missing in plastic research so far and their enhancement is a continuous process (Ivleva et al., 2017; Van Cauwenberghe et al., 2015). Methods adapted in this study are based on successfully applied approaches for macro- and microplastic quantification, but were still constantly evaluated and partly adjusted during the on-going investigation.

> *How might different methods for microplastic identification affect results and are they comparable to each other?*
>
> The identification of microplastics on filters is an essential processing step within microplastic analysis of environmental samples. Two different approaches based on Nile Red staining and photographing of microplastic particles, which mainly differ in the respective resolution, are contrasted (publication I). The classification of comparability between different approaches is important to further estimate harmony in results and to value any under- or overestimation of concentrations. Transformations between different methodical approaches should be developed to finally enable comparisons.

Can the deployment of unmanned aerial vehicles facilitate anthropogenic litter monitoring at lakeshores?

Regular monitoring of AL at lakeshores requires human and time resources for visual search and identification of items. The deployment of UAVs and a subsequent remote analysis of aerial images may help to reduce the effort in the field as already shown for marine beaches (e.g., Martin et al., 2018). Aerial images taken of Lake Tollense beach segments by UAV are evaluated by applying remote image analysis to gain information on the viability of UAVs in AL monitoring at bank border segments of lakes (publication II).

What is the abundance of anthropogenic litter including macroplastic at sandy bank borders of Lake Tollense?

The quantification of AL at sandy lakeshore segments of Lake Tollense is a major goal of the conducted study (publication II). AL monitoring at specific lakes can contribute to data availability for limnic ecosystems. Since the degradation of macroplastics serves as a source for secondary microplastics (Gregory and Andrady, 2003), an investigation of these is essential for a subsequent microplastic analysis. Of course, the assessment of possible AL sources is of equal importance to facilitate the prevention of plastic pollution at lakeshores in future.

Are there differences in the spatial distribution of anthropogenic litter items and which factors influence it?

The analysis of the spatial distribution of AL items may indicate point sources and impacting factors for transport and accumulation. Sandy lakeshore segments with varying expositions and use intensities are considered to examine the spatial distribution at the lake level. Additionally, analysis of AL dispersion within individual lakeshore segments provides insights into small-scaled spatial distribution patterns.

Are there differences in the seasonal distribution of anthropogenic litter items and which factors influence it?

An AL monitoring at different times in the year helps to assess seasonal variations in pollution levels. The examination of temporal distribution patterns can further indicate sources, sinks, and transport pathways for AL as well as impacting factors.

What is the abundance of microplastics in lakeshore and lakebed sediments of Lake Tollense?
The quantification of microplastics in different sediment compartments, namely shore and bed sediments, at Lake Tollense is the second major goal of this study (publication III). Sediment related pollution by microplastics should be evaluated to determine potential sources, fluxes, and retention mechanisms. Results will contribute to the budget balancing of microplastics at Lake Tollense and to the data availability for lake environments in general.

Are there differences in the spatial distribution of microplastics and which factors influence it?
Again, the analysis of spatial variations in microplastic concentrations in lake sediments may indicate sources and impacting factors for transport and distribution. Concentration differences between sediment compartments (lakeshore vs. lakebed sediments) provide insights into transport and aggregation patterns due to diverging sedimentation regimes. Variations between sandy lakeshore segments with varying expositions and use intensities can picture microplastic distribution on a larger (lake) level, and the analysis of multiple sampling points within these segments can account for small-scale variations in microplastic abundances.

Are there differences in the seasonal distribution of microplastics and which factors influence it?
Seasonal variations in sedimentary microplastic concentrations are assessed by samplings conducted at different times in year. Again, the investigation can point to sources and factors that control microplastic abundance and distribution in lake sediments.

Do lake sediments act as a (temporary) sink for microplastics?
The question of (temporary) storage of microplastics in limnic basins represents another aspect of this study. Previous research identified a so-called missing sink for microplastics in the environment. The missing part is related to the discrepancy between plastic inputs from production, recycling, and waste management and identified environmental concentrations. The investigation concerning accumulation, retention, and remobilization of microplastics in lake sediments will provide data to allow predictions on storage mechanisms of limnic ecosystems.

4. Overview of original publications

This cumulative dissertation comprises three publications. In a first step, results from the comparison of methods used for the microplastic identification process were published in the peer-reviewed journal *Environmental Monitoring and Assessment* (Springer). Furthermore, abundances, distribution, and methodical evaluations resulting from the AL monitoring conducted at shores of Lake Tollense are presented in a second publication in *Environmental Research* (Elsevier). Results on microplastic abundances and distribution at Lake Tollense are subject of a third paper, also published in the peer-reviewed journal *Environmental Research* (Elsevier). In the following, abstracts of and personal contribution to the publications are presented. The papers themselves are attached in the appendix.

4.1. Publication I

Hengstmann, E., Fischer, E.K., 2019. Nile red staining in microplastic analysis - Proposal for a reliable and fast identification approach for large microplastics. Environ. Monit. Assess. 191, 612. https://doi.org/10.1007/s10661-019-7786-4

Abstract:

A variety of methods concerning the identification of microplastics in environmental samples exist. While visual identification is often used, implemented easily, and cost-efficient but implying biased results, spectroscopic or chromatographic approaches are reliable but time-consuming and need specific equipment. Nile red staining is an available alternative and complement method for identifying microplastics. In this study, Nile red staining and subsequent photographing in a UV light photobox was tested on its reliability and feasibility. The approach was compared with a second identification process using again staining but a fluorescence microscope. Selected identified microplastic particles were analyzed by μ-Raman spectroscopy to prove their polymeric origin. The results show that the presented approach is faster compared with the use of a fluorescence microscope or μ-Raman spectroscopy. Furthermore, it is cost-effective as well as accurate for large microplastics > 0.63 mm and, therefore, may be applied when large sample volumes need to be analyzed.

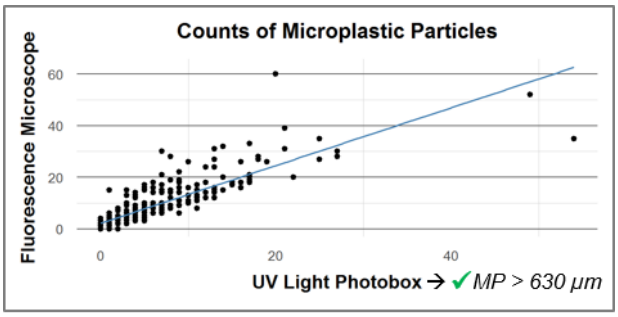

Figure 3: Graphical Abstract for publication I.

Author contribution:

My personal contribution comprises the idea and conceptualization of the study as well as method development. Furthermore, the investigation and formal analysis was conducted by me including sampling, sample processing, identification of microplastics in samples using the two different approaches and statistical evaluation with the focus on the comparison of described methods. I drafted the manuscript and visualized results in graphics. The co-author Elke K. Fischer provided resources for sampling campaigns and laboratory analyses as well as supervision of the overall project. Additionally, she contributed to the final manuscript by reviewing and editing.

4.2. Publication II

Hengstmann, E., Fischer, E.K., 2020. Anthropogenic litter in freshwater environments – Study on lake beaches evaluating marine guidelines and aerial imaging. Environ. Res. 189, 109945. https://doi.org/10.1016/j.envres.2020.109945

Abstract:

Studies on macroplastic pollution in freshwater systems are rare compared to the marine environment. Nevertheless, freshwater systems are worthy to be equally investigated as they are pathways of plastic to the ocean and lakes may act as (temporary) sinks. The aim of this study was to identify sources for plastics and influences on its distribution in a limnic environment. Anthropogenic litter (>5 mm) was monitored semi-annually over a three-year period at four sandy bank border segments of Lake Tollense in Mecklenburg-Western Pomerania, Germany. The selected beaches represent different expositions and vary in their level of anthropogenic activity. Considering all six samplings, mean abundance of anthropogenic litter is 0.2 ± 0.1 items/m² or 130.9 ± 91.0 items/100 m beach length. The averaged mass of anthropogenic litter is 0.5 ± 1.0 g/m² or rather 218.7 ± 284.6 g/100 m. Plastic consistently is the predominate material (72%) and cigarette butts are the most found items. A higher pollution by anthropogenic

litter is found at the end of tourist season unveiling the impact of anthropogenic activity on litter abundance. Additionally, litter transport via tributaries into the lake plays a role.

Testing the detection of anthropogenic litter via aerial images taken by unmanned aerial vehicles resulted in good recovery rates when minimizing the flight height. Furthermore, the analysis of anthropogenic litter distribution displayed on the images showed litter accumulation areas at the border of sandy beach areas. The deployment of marine guidelines in a freshwater environment did work well, however, small changes in the protocol are suggested for future lake beach studies dealing with anthropogenic litter pollution.

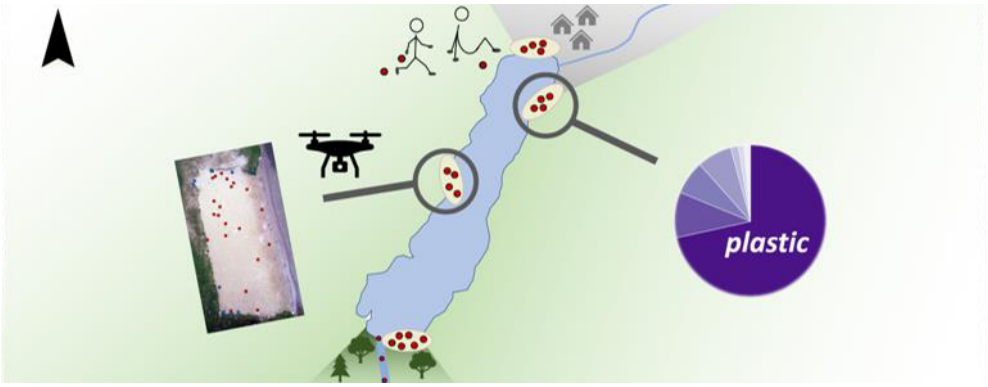

Figure 4: Graphical Abstract for publication II.

Author contribution:

My personal contribution comprises the idea and conceptualization of the case study at Lake Tollense considering sample sites, numbers of samples and sampling methods. Furthermore, the investigation and sampling in field was conducted by me followed by the formal analysis and the statistical evaluation of results. Furthermore, I conducted a method development concerning remote image analysis for macroplastic identification. I drafted the manuscript and visualized results in graphics. The co-author Elke K. Fischer also contributed to the idea and conceptualization of the case study at Lake Tollense and provided necessary resources for sampling and laboratory analyses as well as supervision of the overall project. Additionally, she contributed to the final manuscript by reviewing and editing.

4.3. Publication III

Hengstmann, E., Weil, E., Wallbott, P.C., Tamminga, M., Fischer, E.K., 2021. Microplastics in lakeshore and lakebed sediments – External influences and temporal and spatial variabilities of concentrations. Environ. Res. 197, 111141. https://doi.org/10.1016/j.envres.2021.111141.

Abstract:

Microplastics have been predominantly studied in marine environments compared to freshwater systems. However, the number of studies analyzing microplastic concentrations in water and sediment within lakes and rivers are increasing and are of utmost importance as freshwaters are major pathways for plastics to the oceans. To allow for an adequate risk assessment, detailed knowledge concerning plastic concentrations in different environmental compartments of freshwaters are necessary. Therefore, the major aim of this study was the quantification and analysis of temporal and spatial distribution of microplastics (<5 mm) in freshwater shore and bed sediments at Lake Tollense, Mecklenburg-Western Pomerania, Germany. Likewise, it addresses the hypothesis that lakes may serve as long-term storage basins for microplastics. Concentrations were investigated semi-annually over a two-year period at four sandy bank border segments representing different expositions and levels of anthropogenic influence. In addition, lakebed samples were taken along the longitudinal dimension of Lake Tollense. Mean microplastic abundances were 1,410 ± 822 particles/kg DW for lakeshore sediments and 10,476 ± 4,290 particles/kg DW for lakebed sediments. Fragments were more abundant compared to fibers in both sediment compartments. Spatial and temporal variation was especially recognized for lakeshore sediments whereas microplastic abundances in lakebed sediments did not differ significantly between sampling points and sampling campaigns. This can be related to long-term accumulation at the lakebed. Lower microplastic abundances were found within the intertidal zone at lake beaches where constant wave action reduces accumulation. Increased microplastic abundances were recognized at the beach with least anthropogenic influence but in proximity to a tributary, which may serve as microplastic input pathway into Lake Tollense due to its catchment comprising mainly agricultural areas. Furthermore, spatial variations in microplastic concentrations were related to the abundance of macroplastic items at beaches and correlated with pedologic sediment characteristics, namely the content of organic matter.

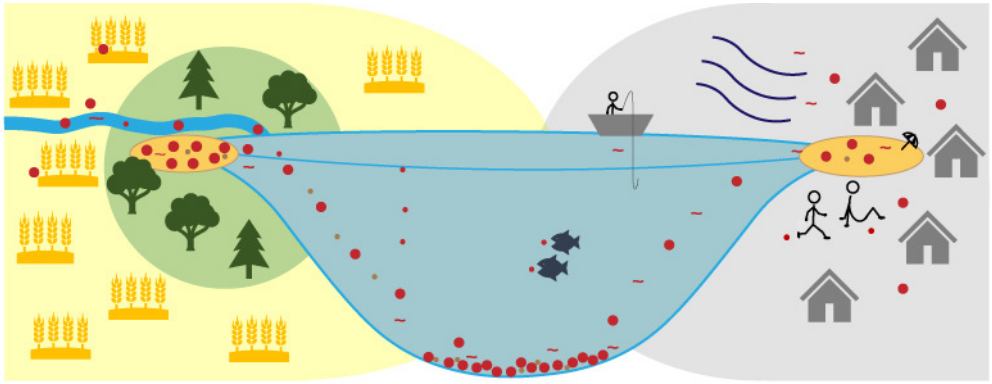

Figure 5: Graphical Abstract for publication III.

Author contribution:

My personal contribution comprises the idea and conceptualization of the case study at Lake Tollense considering sample sites, numbers of samples and sampling methods as well as laboratory methods for microplastic analysis. The investigation and formal analysis were largely conducted by me including regular sampling in field and laboratory analysis of samples (processing, microplastic analysis, pedologic analysis). I performed the statistical evaluation of results and drafted the manuscript. Additionally, I compiled all diagrams and maps. The co-authors Elke K. Fischer and Matthias Tamminga also contributed to the idea and conceptualization of the case study at Lake Tollense. Matthias Tamminga was further involved in the investigation in the field. The methodical development in microplastic identification, resulting in a software tool for the semi-automatic image analysis identifying microplastic particles, was carried out by Esther Weil (Justus-Liebig-Universität Gießen) and Paul C. Wallbott (Fraunhofer Institute for Intelligent Analysis and Information Systems). Further contribution was provided by Elke K. Fischer in terms of resources for sampling and laboratory analyses as well as supervision of the overall project. Additionally, the final manuscript was reviewed and edited by Elke K. Fischer and Matthias Tamminga.

5. Synthesis

This dissertation comprising the three publications listed in chapter 4 focused on the general quantification and evaluation of plastics in the environment, more precisely on the occurrence and distribution of plastic particles in sediments of Lake Tollense, as well as on methodical developments. In this chapter, the relevance of respective outcomes is expressed, and results of individual publications, referred to as publication I to III, will be intertwined and discussed in relation to each other, to the phrased research questions, and to the MICROLIM project.

The further development of microplastic identification methods or rather the comparison of different approaches thematized in publication I contributes towards the methodological development in microplastic research. Similarly, tests considering aerial image analysis for AL monitoring at lakeshores described in publication II contribute to macroplastic method developments. Due to a relatively young and highly dynamic research field, methods for the investigation of plastic pollution in different environmental compartments are regularly improved and still need further development (Ivleva et al., 2017; Wagner et al., 2014). The evaluation of two different microplastic identification methods for environmental samples presented in publication I highlights the easy application of the first approach using a UV light photobox in combination with a digital camera to image microplastic particles stained by Nile Red. This approach is less reliable for small particles (≤630 µm) compared to the fluorescence microscope due to lower resolution of the resulting images. However, a UV light photobox is easier to implement, as it does not require expensive equipment. The photobox itself is made of wooden boards, and UV illumination and digital cameras are widely available at comparably low costs today, facilitating the described microplastic identification process. For monitoring programs considering environmental plastic pollution, simple, cost-effective, standardized, and consistent methods are necessary to provide practicability of multiple studies at several locations and various points of time with reasonable effort (GESAMP, 2019; Cheshire et al., 2009). Such monitoring programs were and are developed worldwide comprising repeated investigations in different compartments to create comprehensive and reliable data sets over space and time and verify developments in concentrations induced by temporal variations or anti-littering measurements (GESAMP, 2019; Cheshire et al., 2009). In a European context, the MSFD requires member states to implement monitoring programs to achieve “good environmental status” also considering plastic pollution (European Commission, 2018). Varying lower size limits for microplastics were proposed for monitoring programs (e.g., GESAMP, 2019; Masura et al., 2015). In this regard, the UV light photobox approach may be commonly applied for the detection of large

microplastic particles (at least >630 µm) whereas methods with higher resolution like the fluorescence microscope can be implemented for the detection of smaller microplastics only if needed and time- and cost-efforts are adequate.

Publication I considering microplastic identification method developments further provides a computational conversion between results gained by the UV light photobox and by the higher resolution fluorescence microscope to project concentrations from one method to the other. The implementation of the UV light photobox for monitoring studies should be followed by extrapolation of resulting microplastic concentrations based on the provided conversion to increase comparability to other studies, especially for smaller microplastics. Methodical variability in microplastic research studies often hampers the comparison of results. Harmonization efforts are promoted (e.g., Primpke et al., 2020; GESAMP, 2019; Rochman et al., 2017) but have not reached a final level. Therefore, conversions between different approaches are highly needed and have already been proposed by, e.g., Koelmans et al. (2020) in terms of computational tools to transfer microplastic concentrations between different analyzed size ranges of particles and for the alignment of results presented in different units. In this regard, the developed computational transfer between the two microplastic identification methods, the UV light photobox and the fluorescence microscope, is of utmost importance and was already successfully applied for the microplastic analysis in Lake Tollense shore sediments presented in publication III. For the first sampling campaign, microplastic particles in samples were identified by the UV light photobox. The fluorescence microscope was applied for subsequent samplings. Therefore, raw results were not comparable between the first and following samplings. Based on the developed regression model for computational conversion, microplastic abundances were aligned to allow consideration of all samplings in the final evaluation.

Tests concerning the implementation of UAVs for the detection of AL at lake beaches (publication II) showed that this approach can also be considered an effective monitoring approach in a limnic context, in case of further improvement of remote image analysis. Other authors have utilized images taken by UAVs, LiDAR technology or webcams to detect AL items at shorelines as well (e.g., Bao et al., 2018; Ge et al., 2016; Kataoka et al., 2012). In the presented study, evaluation of UAV data was complicated by low resolution of images or the need for manual input by the processor. However, semi-automatic or even automatic image analysis in relation to litter detection was improved recently for example by deep-learning approaches (e.g., Politikos et al., 2021; Gonçalves et al., 2020a; Martin et al., 2018). The implementation of remote approaches by nowadays affordable UAVs can provide simplified monitoring of litter

at shorelines requiring less effort in the field and covering greater areas (Papakonstantinou et al., 2021; Gonçalves et al., 2020a, 2020b).

The semi-automatic approach for litter detection via UAV was not implemented on a regular basis in this study. Continuing tests and improvements in image quality and image analysis are necessary to facilitate the process. Still, aerial images were utilized to locate AL items more precisely within the beach segments investigated by the combination with Differential Global Positioning System (DGPS) data recorded in the field. As described in publication II, AL was predominantly dispersed across the beach segments and only partly accumulated in the transition zone from sandy to vegetated areas and to an even lower degree at the high tide line. Former studies verified an accumulation of items in the back of beaches as well as at the strand line (e.g., Blettler et al., 2017; Hengstmann et al., 2017; Hoellein et al., 2014). The distribution of AL items can be compared to microplastic concentrations at different sampling points within the beach segments to analyze accordance or discordance concerning the small-scale allocation of macro- and microplastics. For microplastics, significantly higher concentrations were found at the sampled high tide line and in elevated shore areas compared to the intertidal zone (see publication III). Therefore, both, microplastics and AL items, are primarily dispersed across the analyzed beach segments with low accumulation rates at the high tide line or in the transition zone. In contrast, Corcoran et al. (2015) showed that small particles (<10 cm) accumulated at the strand line trapped by organic material, whereas larger items were evenly dispersed at Laurentian Great Lakes beaches. The difference in size class limits might result in diverging observations concerning distribution patterns. Despite the divergence in size, a correlation to organic content in sediments was similarly verified for microplastic concentrations in lakeshore sediments of Lake Tollense. In general, a relation to sediment characteristics, especially organic content and partly sediment grain size distribution, was demonstrated for microplastic abundances in Lake Tollense sediments.

This PhD study further verified a coinciding trend for AL items and microplastics considering variability at the lake level, between investigated beach segments at Lake Tollense. For both types of plastic debris, South beach showed the highest concentrations and East beach was least polluted, as described in publication II and III. Physical, chemical, and biological induced fragmentation of large plastic items into several smaller microplastic particles (Helm, 2020; Lambert et al., 2014; Andrady, 2011) is one reasonable explanation for the relation between the two variables. The comparison of AL and microplastic composition partially confirms this assumption. Major polymers identified for microplastic particles in lakeshore and lakebed sediments

are polyethylene (PE), polyamide (PA), PP and PET. Owing to the frequent utilization of PE and PP in plastic packaging materials and the common application of PA and PET in fishing and textile materials, these can be designated as major sources for microplastic particles (e.g., PlasticsEurope, 2020; Koelmans et al., 2019; Scopetani et al., 2019; Dris et al., 2018). For AL, cigarette butts and filters were the most frequently found items which are made from neither of the polymers mentioned above, but from cellulose acetate. The high number of cigarette butts and filters points to *in situ* littering due to recreational beach activities. However, AL items were also classified as unidentifiable plastic pieces as well as bottle caps, foil wrappers or sweet packaging. These items may or are very likely made of PE or PP, respectively, emphasizing the formation of microplastics from large plastic items. Only a few textile items were identified in the AL monitoring. Consequently, microplastic fiber input at lakeshores is less related to fragmentation of larger plastic items compared to fragments. Again, recreational activities serve as input pathways via the release of fibers from synthetic clothes worn by beach visitors (Scopetani et al., 2019; Turner et al., 2019).

Not yet discussed deviations in distribution and composition between AL and microplastics, result from divergent input and transport pathways for the two categories which were identified in publication II and III. Besides the generation of microplastics via fragmentation of large plastic items, they are further introduced into Lake Tollense by tributaries. Especially the Nonnenbach in proximity to South beach seems to function as an input pathway and particles are subsequently washed ashore. Agricultural areas may serve as a source for microplastics (Dris et al., 2018; Rochman, 2018; Wagner et al., 2014) in the catchment area of the Nonnenbach. Contrastingly, AL abundance is primarily influenced by *in situ* human activity and wind exposition. The difference concerning impact factors is further underlined when considering the seasonal variations of AL versus microplastic concentrations. AL abundances were significantly higher in September compared to March indicating again the strong impact of *in situ* anthropogenic activity which was confirmed by other authors before (e.g., McCormick and Hoellein, 2016; Hoellein et al., 2015). The touristic season at Lake Tollense lasts from May to September. Whereas samplings in March were conducted well before the start of touristic activity, samplings in September were carried out at the end of the tourist season. Contrastingly, microplastic concentrations at Lake Tollense shores were higher in March compared to September. Lake and tributary hydrology as well as wind are more crucial factors for seasonal microplastic variations.

Lake characteristics and hydrology further influence the retention capability for microplastics in lake sediments (e.g., Imhof et al., 2018; Ballent et al., 2016; Free et al., 2014). Lakeshore sediments are, to a certain degree, able to retain microplastics, but particles may still be re-suspended by waves and high water levels. Lakebed sediments provide better retention conditions for microplastics due to low water dynamics at the lake bottom. For the settlement and incorporation of microplastics in lakebed sediments, biotic factors are of further importance. The formation of biofilms on the surface of microplastic particles can induce the sinking of even low-density polymers through the water column (e.g., Kaiser et al., 2017; Chubarenko et al., 2016; Andrady, 2011), enabling the settling and accumulation of particles at the lake bottom. Therefore, lakes seem to serve as a storage basin for microplastics on their general transport from terrestrial sources to the marine final sink, as demonstrated in publication III. The storage characteristic of lakes was further verified by other recent studies (Li et al., 2020; Bordós et al., 2019; Turner et al., 2019; Vaughan et al., 2017).

From the synthesis of publication II and III, a complex causal network explaining the plastic occurrence and distribution in lake sediments may be derived. Summarizing, this network includes the following key elements:

- Lake and tributary related hydrology,
- Lake morphology, especially depth, shape, and shore structure,
- Wind (direction and velocity) with resulting wind-driven currents and waves,
- Surface runoff from the catchment (urban and rural),
- Sediment characteristics, especially organic content,
- Biotic factors, especially formation of biofilms,
- Anthropogenic activity, including proximity to urban areas, presence, and intensity of agriculture in the catchment area, frequency and type of recreational activities, and counter measurements for beach pollution.

In addition, atmospheric input plays a considerable role especially for microplastic occurrence as shown by former studies (Allen et al., 2019; Klein and Fischer, 2019; Dris et al., 2018). However, atmospheric inputs were not considered in this PhD study. Within the MICROLIM project, atmospheric deposition is analyzed with respect to microplastic concentrations and will provide information on this input parameter at Lake Tollense. Moreover, the evaluation of biotic influence factors on the occurrence of microplastics in limnic ecosystems is included in the MICROLIM project. First hints towards microplastic ingestion by biota in Lake Tollense based on results from microplastic analysis in lakebed sediments in publication III have to be verified

by the analysis of microplastic concentrations in the biosphere. Considering the influence of lake and tributary hydrology on macro- and microplastic accumulation in lakes, the analysis of the hydrosphere can give further insights. Water samples from Lake Tollense and its tributaries were investigated for its microplastic pollution simultaneously to the analysis of sediment samples but are not within the scope of this dissertation. The MICROLIM project will summarize the comprehensive datasets gained for all environmental compartments in multiannual investigations at Lake Tollense and will give a synthesis concerning microplastic concentrations in the model lake catchment.

Concentration levels in the pedosphere and assumptions concerning impact factors derived in this PhD study from repeated AL monitoring and microplastic analysis in shore and bed sediments of Lake Tollense in a two- or three-year period contribute to the evaluation of sources, pathways, and sinks for macro- and microplastic in the model catchment of Lake Tollense. Thereby, decisive data is provided to accomplish the major aim defined for the MICROLIM project, budgeting microplastics in the model lake catchment. To summarize, this study and the overall MICROLIM project contribute to the data availability considering plastic pollution in limnic ecosystems which is highly needed to improve data reliability and understanding of processes within the ecosystem.

6. Conclusion and perspective

The presented PhD project encompasses an evaluation of potential methods for macro- and microplastic monitoring and, at the same time, an investigation of macro- and microplastic pollution in sediments in the model catchment of Lake Tollense. These two aspects contribute to the development of methodical approaches in the field of plastic research and provide substantial datasets by multiannual and repeated sampling adding to the data availability concerning freshwater plastic pollution. Plastic pollution data for freshwaters is of importance as these ecosystems were less studied compared to marine ecosystems in the past decades. The widespread pollution of Lake Tollense sediments by macro- and microplastics influenced by a set of diverse and crucial factors underline the need for comprehensive analysis of the plastic pollution in lakes. Within the greater framework of the MICROLIM project, results concerning plastic concentrations in the pedosphere will be combined with results from other compartments, namely hydrosphere, atmosphere, and biosphere to provide a more profound overview on plastic pollution in Lake Tollense. Thereby, the network of major influencing factors on plastic abundance and dispersion in the study area determined by this PhD project will be expanded, enabling the budgeting of microplastics in the lake catchment. Furthermore, an input-output model for microplastic particles in Lake Tollense based on the available baseline concentrations and in- and output parameters will be established to improve the understanding of sources, transport pathways, and sinks of plastics in the model catchment.

The importance for investigating lake ecosystems with respect to their plastic pollution in general is further emphasized by retention capabilities of lake sediments verified in this study. The presence and storage of microplastics in sediments provide availability of particles to different organisms. Research on effects of plastics on freshwater organisms needs to be expanded by in field and laboratory studies as well as research on possible adverse effects on human health. Furthermore, effect thresholds need to be considered to classify the severity of plastic pollution in lakes. In a marine context, regulatory limits for plastic pollution are just discussed and should be likewise assigned to freshwater ecosystems as these serve as major input pathways for microplastics into the oceans. The determination of plastic concentrations in different freshwater compartments like the pedosphere is a first step in a right direction. A plausible risk assessment is not possible before comprehensive data on pollution levels in freshwater ecosystems and reliable data on plastic impacts are available. Only then, concentration thresholds can be assigned for different compartments and appropriate measures can be adopted to counteract environmental plastic pollution.

List of publications

Original publications with reference to the cumulative dissertation

Hengstmann, E., Fischer, E.K., 2019. Nile red staining in microplastic analysis - Proposal for a reliable and fast identification approach for large microplastics. Environ. Monit. Assess. 191, 612. https://doi.org/10.1007/s10661-019-7786-4

Hengstmann, E., Fischer, E.K., 2020. Anthropogenic litter in freshwater environments – Study on lake beaches evaluating marine guidelines and aerial imaging. Environ. Res. 189, 109945. https://doi.org/10.1016/j.envres.2020.109945

Hengstmann, E., Weil, E., Wallbott, P.C., Tamminga, M., Fischer, E.K., 2021. Microplastics in lakeshore and lakebed sediments – External influences and temporal and spatial variabilities of concentrations. Environ. Res. 197, 111141. https://doi.org/10.1016/j.envres.2021.111141.

Further publications not related to the dissertation

Hengstmann, E., Gräwe, D., Tamminga, M., Fischer, E.K., 2017. Marine litter abundance and distribution on beaches on the Isle of Rügen considering the influence of exposition, morphology and recreational activities. Mar. Pollut. Bull. 115, 297–306. https://doi.org/10.1016/j.marpolbul.2016.12.026

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Hengstmann, E., Fischer, E.K., 2018. Macro- and microplastics in sediments from Lake Tollense, Germany. In: Baztan, J., Bergmann, M., Carrasco, A., Fossi, C., Jorgensen, B., Miguelez, Q., Pahl, S., Thompson, R.C., Vanderlinden, J-P. (Eds.), 2018. MICRO 2018. Fate and Impact of Microplastics: Knowledge, Actions and Solutions. 414 pp. MSFS-RBLZ. ISBN 978-84-09-06477-9. CC-BY-NC-SA.

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Appendix: Original publications

Publication I

Hengstmann, E., Fischer, E.K., 2019. Nile red staining in microplastic analysis - Proposal for a reliable and fast identification approach for large microplastics. Environ. Monit. Assess. 191, 612. https://doi.org/10.1007/s10661-019-7786-4

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Nile red staining in microplastic analysis—proposal for a reliable and fast identification approach for large microplastics

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Abstract A variety of methods concerning the identification of microplastics in environmental samples exist. While visual identification is often used, implemented easily, and cost-efficient but implying biased results, spectroscopic or chromatographic approaches are reliable but time-consuming and need specific equipment. Nile red staining is an available alternative and complement method for identifying microplastics. In this study, Nile red staining and subsequent photographing in a UV light photobox was tested on its reliability and feasibility. The approach was compared with a second identification process using again staining but a fluorescence microscope. Selected identified microplastic particles were analyzed by μ-Raman spectroscopy to prove their polymeric origin. The results show that the presented approach is faster compared with the use of a fluorescence microscope or μ-Raman spectroscopy. Furthermore, it is cost-effective as well as accurate for large microplastics > 0.63 mm and, therefore, may be applied when large sample volumes need to be analyzed.

Electronic supplementary material The online version of this article (https://doi.org/10.1007/s10661-019-7786-4) contains supplementary material, which is available to authorized users.

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Keywords Microplastics · Identification · Monitoring · Nile red staining

Introduction

Microplastics, being introduced into the environment via direct input pathways or by weathering and degrading of larger plastic objects (Barnes et al. 2009; Andrady 2015; Geyer et al. 2017), became a concerning environmental pollutant, which is enhanced by increasing global production rates (PlasticsEurope 2018). As a widely spread pollutant (Galgani 2015), the impacts of microplastics may be severe due to ingestion of particles by organisms including the subsequent uptake and accumulation of Persistent Organic Pollutants (e.g., Anbumani and Kakkar 2018; Wang et al. 2018).

Microplastics are widely defined as plastic particles < 5 mm (Arthur et al. 2009); however, other definitions are used. Recently, 1 mm was progressively proposed as an upper size limit for microplastics (e.g., GESAMP 2015; Hartmann et al. 2019). For the analysis of microplastics in environmental samples, several different protocols have been applied. Also, the final step of identifying microplastics varies greatly between studies. While some studies solely determine microplastics visually (e.g., Morét-Ferguson et al. 2010; Dris et al. 2015; Mason et al. 2016; Pinheiro et al. 2019), with or without microscope, others use staining protocols (e.g., Fischer et al. 2016; Shim et al. 2016; Erni-Cassola et al. 2017; Maes et al. 2017), while others apply

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spectroscopic or chromatographic methods (e.g., Käppler et al. 2016; Fischer and Scholz-Böttcher 2019; Primpke et al. 2019). The visual inspection of microplastics is a simple and fast method; however, it may be inaccurate, especially for smaller microplastics, and is dependent on the researcher (Hidalgo-Ruz et al. 2012; MSFD 2013; Löder and Gerdts 2015). In contrast, (μ-)Raman spectroscopy and (μ-)Fourier transform infrared (FTIR) spectroscopy as well as pyrolysis-gas chromatography are accurate, though, these methods require expensive equipment and are time-consuming (Lusher et al. 2017; Primpke et al. 2017; Silva et al. 2018; Zarfl 2019). Identification methods combining the two aspects of, on one hand, being fast, simple, and inexpensive and, on the other hand, being accurate, reliable, and independent are highly needed in microplastic research. This is especially true for monitoring approaches of microplastics in the environment covering large sample numbers and volumes in short times. These may lead to high costs and time-consuming laboratory analyses. Technical and methodological improvements in microplastics analysis may counteract these problems (MSFD 2013).

The lipophilic dye Nile red has been introduced to the analysis of microplastics by Andrady (2010) and has since been adapted and modified by other authors (Song et al. 2014; Shim et al. 2016; Erni-Cassola et al. 2017; Maes et al. 2017; Tamminga et al. 2017) providing an alternative identification method for microplastics.

In this study, we analyzed the staining method via Nile red proposed by Tamminga et al. (2017) using the proposed UV light photobox as well as a fluorescence microscope. The comparison between the two approaches is presented and suggestions for using these in monitoring approaches are expressed.

Materials and methods

For the comparison of two identification methods in microplastic analysis, 44 environmental sediment samples which were taken in September 2017 from Lake Tollense, Mecklenburg-Western Pomerania, Germany, were considered. Additionally, 12 blank samples were run parallel during the laboratory processes and were as well considered for the comparison presented here. A detailed photo documentation of the applied Nile red staining protocol and subsequent photographing is provided in the Supplementary Material (SM; S3).

Sample preparation

The preparation of sediment samples included the destruction of organic material and the fractioning of the sample as well as a density separation step. Blank samples were processed equally except for starting with an empty beaker instead of with 50 ml sediments.

To remove the organic matter, H_2O_2 (30%; 60 ml, 7 days at room temperature) was added to the environmental and blank samples. Hydrogen peroxide is deployed for the destruction of organic matter in soil (DIN ISO 11277) and in microplastic analysis (e.g., Nuelle et al. 2014). After removing the first reagent via wet sieving (0.063 mm), NaClO (6–14%; volume ratio 1:3, 24 h at room temperature) was added to remove residual organic matter (Collard et al. 2015). Subsequently, the sample was fractionated into the following size classes that are adapted to the subsequent elutriation process for density separation and are derived from divisions used in pedologic (DIN EN ISO 14688-1) as well as microplastic research: > 1.0–5.0 mm, > 0.63–1 mm, > 0.3–0.63 mm, > 0.2–0.3 mm, > 0.063–0.2 mm (see Hengstmann et al. (2018) for a detailed description). The residuals of the elutriation process were transferred onto filters (VWR, qualitative filter paper 413, 5–13 μm particle retention) via a stainless-steel filter funnel (Sartorius Stedim) and left to dry in closed Petri dishes for 48 h at room temperature. In cases of high material loads, the residuals were split onto more than one filter.

In total, 287 filters (see Table 1) were further processed to compare the UV light photobox and fluorescence microscopic approach. The filters were stained with 1 ml of a Nile red solution (1 mg/ml Nile red in chloroform according to Tamminga et al. 2017), covered with a watch glass and dried for 24 h at room temperature.

UV light photobox

All filters were photographed in the UV light photobox. This box was developed for the purpose of microplastic identification and is made of wood. A circular hole was drilled into the central wooden board to allow a replicable position of the sample. UV lights (Omnilux UV 18W G13, 365 nm) were installed in the upper level of the box to have top light conditions. A second circular hole was drilled into the cover for placing the camera. A picture of the UV light photobox is provided in the SM (S1).

Table 1 Number of filters representing different size fractions used in the study. Note that only one blank sample contained material in the largest size fraction

Size fraction	No. of filters from environmental samples	No. of filters from blank samples	Total no. of filters
> 1.0–5.0 mm	42	1	43
> 0.63–1 mm	42	12	54
> 0.3–0.63 mm	43	12	55
> 0.2–0.3 mm	53	12	65
> 0.063–0.2 mm	58	12	70
Total	238	49	287

With a digital camera (Pentax K-30, exposure time 2″, ISO 100, resolution 2420 × 2343), one picture per filter was taken after focusing manually. Subsequently, each picture was evaluated using the software Photoshop (Adobe, Version CS5) and microplastics were counted by shape as fragments and fibers. Microplastic particles appear pink under UV light when stained with Nile red in chloroform (Tamminga et al. 2017) and were compared with formerly photographed reference particles (virgin pellets by Good Fellow and post-consumer plastics of different polymers as well as biogenic material). Since samples were previously sieved into different size fractions, only particles in the respective size range were counted on the filter photos.

Fluorescence microscope

All filters were also photographed with a fluorescence microscope (AxioLab A.1, Zeiss, TRITC HC filterset (AHF), × 2.5 magnification) with external illuminant. A camera (Canon EOS 80D, exposure time 1″, ISO 500, resolution 1 × 1 μm) is combined with the microscope. Using these settings, a rectangular of 6 × 4 mm of the filter is presented in one picture. To cover the complete filter, circa 160 partly overlapping photos have to be taken with an *x*- and *y*-offset of 5 and 3 mm, respectively. However, if no stained particle was visible in the preview, no photo was taken in order to reduce the number of pictures to be analyzed. The surface of the filter was manually focused before taking the picture.

Again, each picture was evaluated using Photoshop and counting microplastic fragments and fibers according to the procedure described for the UV light photobox. Microplastic particles appear yellow under the fluorescence microscope when stained with Nile red in Chloroform.

Spectroscopic analysis

To gather data on the chemical composition and to ensure that only microplastic particles were considered, selected microplastic particles (n = 130) were collected from the filters after being photographed with both methods and were analyzed with a μ-Raman spectroscope (DXR2xi Raman Imaging Microscope, Thermo Fisher Scientific). The particles were placed between object slides and spectra were recorded with a laser power of 5–8 mW, an exposure time of ~ 10 Hz and at least 500 scans. The preceding staining via Nile red may induce fluorescence. Therefore, particles were first photo-bleached at 10 mW before recording the actual spectrum according to Tamminga et al. (2019).

Statistical analysis

Statistical analyses were performed using R statistics (R Core Team 2017, Version 3.4.2) in an R Studio environment (RStudio Team 2016, Inc., Version 1.1.383). None of the data was normally distributed (Shapiro-Wilk test). Spearman's Rho was calculated to compare the results of both approaches. A linear trend was visible for the relation between the two datasets, and therefore, linear regression models were computed for each size fraction and for fragments and fibers separately. Statistical outliers were excluded when the Cook's distance was > 1 before running the regression.

Results

In total, 1731 particles (822 fragments and 909 fibers) were counted in the UV light photobox, while the number of particles counted on microscopic photos was 2552 particles (1282 fragments and 1270 fibers). Figure 1 shows the results comparing the two methods according to the different size classes and shapes.

The number of microplastics per filter increases with decreasing size class. Simultaneously, the difference between the counts in the photobox and under the microscope increases. For fragments, this increase is more pronounced than for fibers.

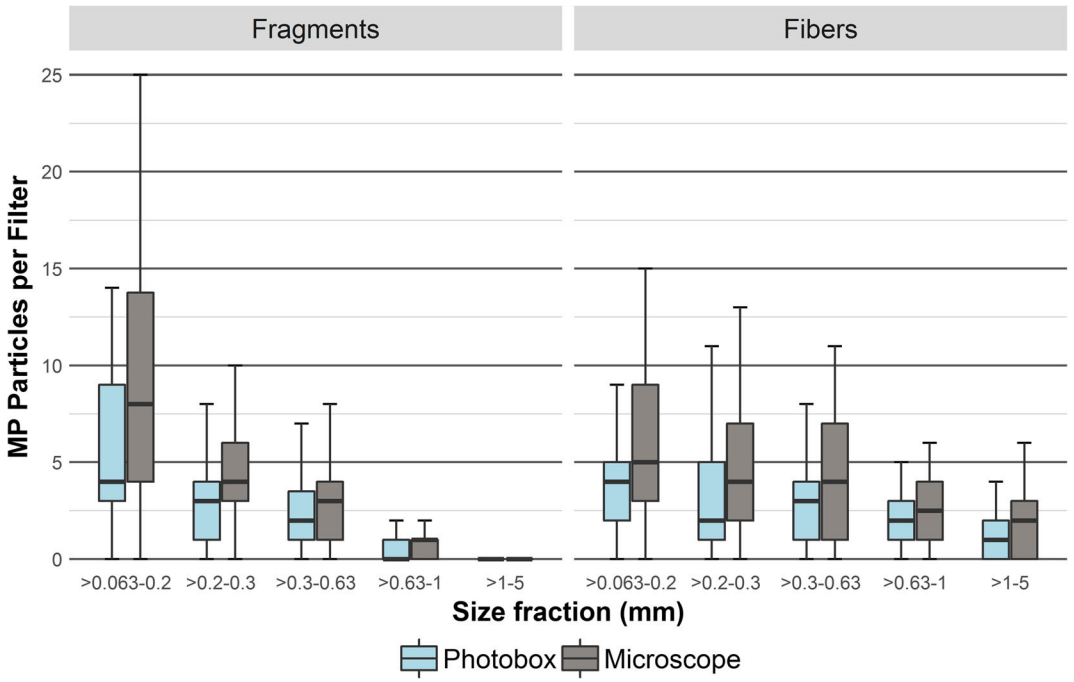

Fig. 1 Comparison of counts of fragments and fibers between the two identification approaches used considering different size fractions (outliers are not presented)

Correlations between the number of particles in the UV light photobox and under the fluorescence microscope were tested for each size class and shape, respectively. The correlation coefficients were predominately > 0.7 showing high correlations for all tested relationships (see S2 in the SM).

Scatterplots with regression lines were produced for the different size classes for fragments and fibers separately. These are shown in Fig. 2a and b. All regression models display a strong explained variability according to Cohen (1988; $r^2 > 0.26$). For fragments, the slope of the regression line is nearly 1 for all size classes, differing significantly from zero ($\alpha = 0.001$). The same is true for fibers except for the smallest size class. In contrast, the *y*-offset greatly differs between the models for different size classes. It increases with decreasing size of particles and is close to zero for the size classes > 0.63–1.0 and > 1.0–5.0 mm for fragments. Again, a similar trend is visible for fibers, except for the smallest size class and *y*-offsets are at least 0.6. Statistical evaluation showed that the *y*-offsets significantly differ from zero for all size classes except for > 1.0–5.0 mm concerning fragments and > 0.063–0.2 mm considering fibers ($\alpha = 0.01$). The statistical analysis concerning the quality of the regression proved that the models work very well for the two smallest size classes, both for fragments and fibers. For the two largest size classes, however, the distribution of residuals is not normal or homoscedasticity of residuals is not given. For the size class > 0.3–0.63 mm, residuals are only approximately normally distributed.

Figure 3 shows an example of a polymer fragment identified by the UV light photobox and the fluorescence microscope. It was furthermore analyzed via μ-Raman spectroscopy revealing that the fragment is made of PET. The spectrum of the analyzed particle, even though it has an unstable baseline, matches the PET reference spectrum very well (see Fig. 3d). The unstable baseline of the particle spectrum is expected to be caused by weathering changes in PET structure for microplastics from environmental samples and the preceding staining process with Nile red resulting in fluorescence. Similarly, 130 of all identified particles were analyzed for their chemical composition via μ-Raman spectroscopy, corresponding to ca. 7.5% of the particles counted in the photobox and ca. 5% considering particles from the microscope. 91.5% of these particles were

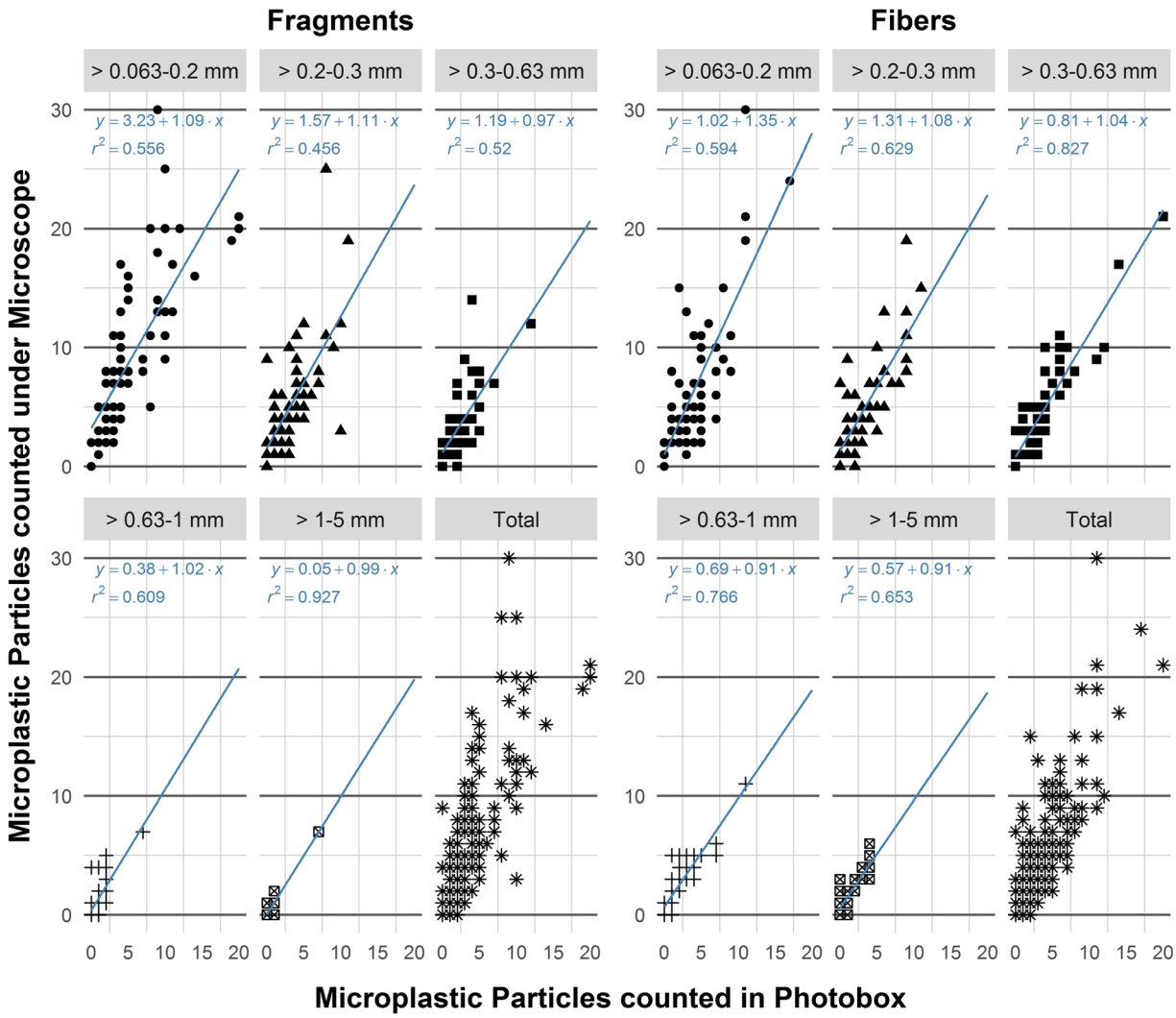

Fig. 2 Scatterplots with regression models considering the differences in particle counts between the two applied methods for different size classes for fragments and fibers

confirmed to be polymers; 3.1% were also of synthetic origin and represented pigments. Nearly 4% of all particles were organic particles, especially cellulose, and less than 2% were minerals. Nearly half of the plastic particles were made of polyethylene terephthalate (PET), especially fibers, followed by polyethylene (PE; 40%, including copolymers). Only small fractions were made of polyamide (PA), polypropylene (PP), or were classified as phenoxy resins. The polymers polystyrene (PS) and polyacrylonitrile (PAN) were even less represented with only 1%, respectively, whereby PS is evidently altered by the staining process due to the use of chloroform (Tamminga et al. 2017).

Discussion

The results clearly show that the UV light photobox approach is more reliable for bigger size classes. For these, the particle counts are close to the microscope results. Mean differences between the two methods are less than half a particle for fragments as well as fibers for the size classes > 0.63–1.0 and > 1.0–5.0 mm. Additionally, over 90% of identified particles were confirmed as plastic particles by μ-Raman spectroscopy regardless of the specific size class. Therefore, it can be stated that for larger microplastics (> 0.63–5.0 mm), corresponding to the grain size of coarse sand and larger according to DIN EN ISO 14688-1, the staining via Nile red and

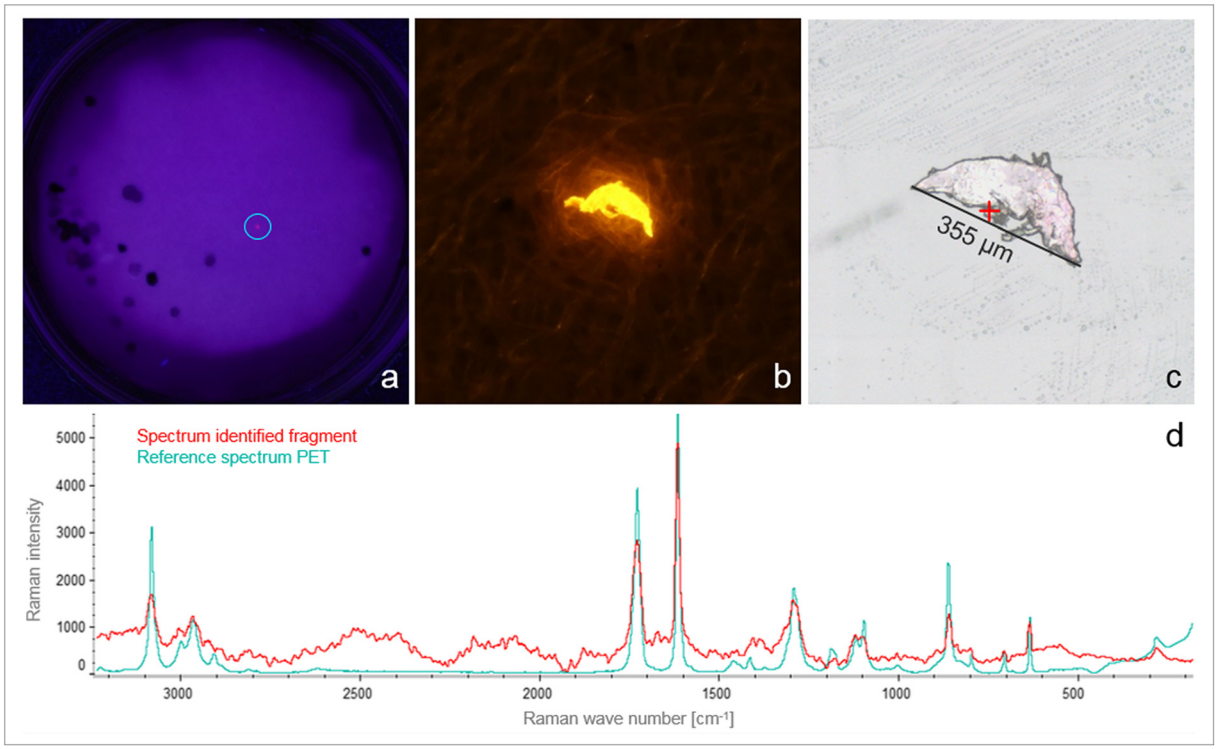

Fig. 3 Identified plastic fragment in the UV light photobox (**a**, within blue circle), under the fluorescence microscope (**b**), and under Raman Imaging Microscope (**c**); the spectrum (**d**) is the result of μ-Raman analysis of the shown particle compared with a reference spectrum of PET

subsequent photographing in the UV light photobox is a suitable method to identify microplastics in environmental samples. The visual detection limit is restricted and often defined as 1 mm (Hidalgo-Ruz et al. 2012; Song et al. 2015; MSFD 2013) or 500 μm (Löder and Gerdts 2015; Laforsch 2017). However, when microplastics are only identified visually, a potentially severe bias remains due to subjectivity, a dependence on quality and magnification of (microscopic) pictures and interfering materials from the sample matrix (Löder and Gerdts 2015; Song et al. 2015). Using Nile red, the misidentification can be reduced (Erni-Cassola et al. 2017; Maes et al. 2017; GESAMP 2019), especially due to the independence concerning the color of particles (Lenz et al. 2015). Yet, the identification using microscopy may still be time-consuming. To achieve a faster result for larger microplastics, the previously described method of using a UV light photobox provides a good alternative. Large microplastics can be easily and efficiently detected in one picture per filter. Especially, when monitoring microplastics in environmental samples, this may be very helpful. A monitoring program needs to deliver accurate data on microplastic accumulation in ecosystems in a short time and with low costs (MSFD 2013; González et al. 2016; GESAMP 2019). The identification via Nile red in a UV light photobox is reliable (for microplastics > 0.63 mm), fast, and does not require expensive equipment as presented in this study. Even for monitoring studies using citizen science, the involvement of non-scientists for acquiring scientific data (Bonney et al. 2009), this method may be applicable with minor effort. Citizen science applied in microplastic research often leads to great numbers of samples from a wide spectrum of geographical areas (e.g., Forrest et al. 2019). Screening such samples for the occurrence of microplastics can be easily shortened with the described method. The evaluators should be trained with reference samples, though, before they analyze environmental samples. In addition, cross-checking between the two described identification methods and Raman results can improve the reliability of the enumeration of particles on images.

Still, for smaller size classes, the Nile red staining protocol was proven to work reliably since more than 90% of all particles were verified as polymers or synthetic products using μ-Raman spectroscopy. However, for

small size classes, the UV light photobox is not satisfactory and does produce non-reliable data due to low resolutions of filter photos. Here, the implementation of a microscope with a magnification of at least × 2.5 (for particles > 0.063 mm) is necessary. This study also demonstrated that the number of microplastic particles recognized in the UV light photobox may be extrapolated to be comparable with microscopic results. Regression models can be used to statistically project the counts from the photobox towards a more accurate quantification of microplastics in environmental samples.

Conclusion

To conclude, the implementation of Nile red and a UV light photobox, where filters containing environmental samples may be photographed in total, proved to be a reliable and fast identification method for microplastic analysis for particles > 0.63 mm. The limit of 0.63 mm is derived from pedologic analyses. Further tests need to be performed to confirm whether this lower size limit may be decreased to 0.5 mm to match other size limits in microplastic analysis.

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Compliance with ethical standards This article does not contain any studies with human participants or animals performed by any of the authors.

Conflict of interest The authors declare that they have no conflict of interest.

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Supplementary Material

Nile Red Staining in Microplastic Analysis – Proposal for a reliable and fast Identification Approach for large Microplastics

Journal: Environmental Monitoring and Assessment

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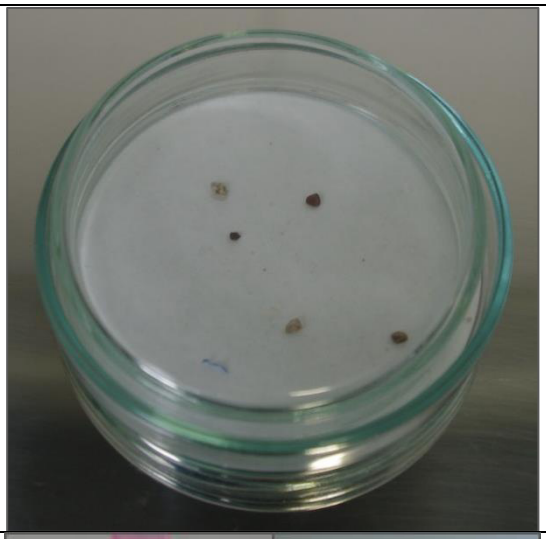
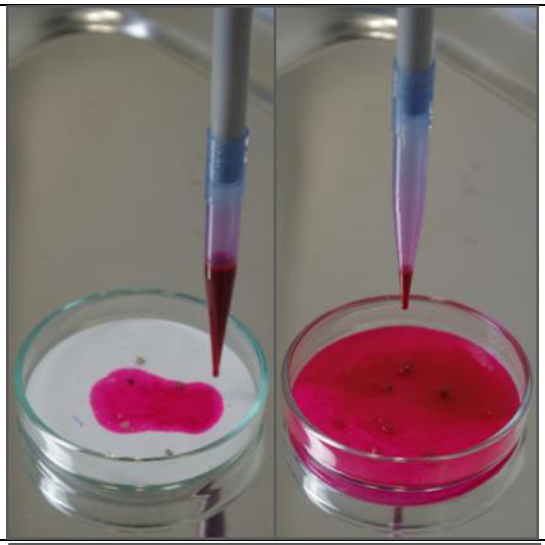
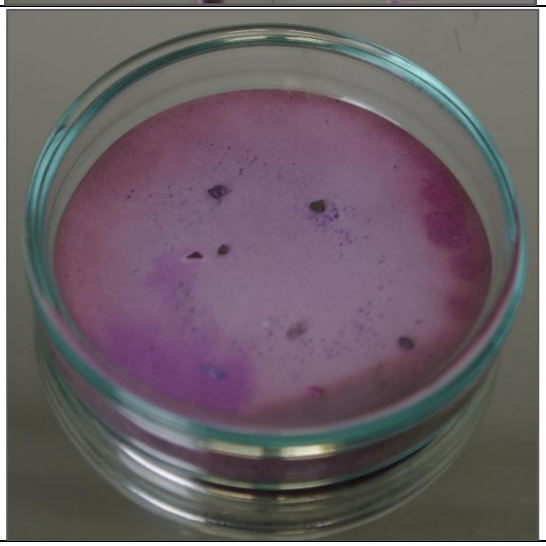
S1: Pictures of the UV light Photobox. (a) front view, (b) upper level with sample, (c) top view. The lowest level provides the possibility to install bottom light illumination but is not actually required for the recording of microplastics. The second level includes a whole to place the sample replicably and is equipped with UV light illumination from above. At the top, the camera is installed directly above the filter.

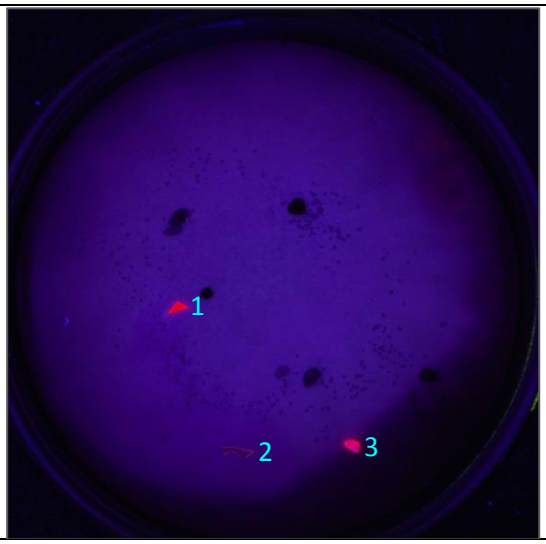
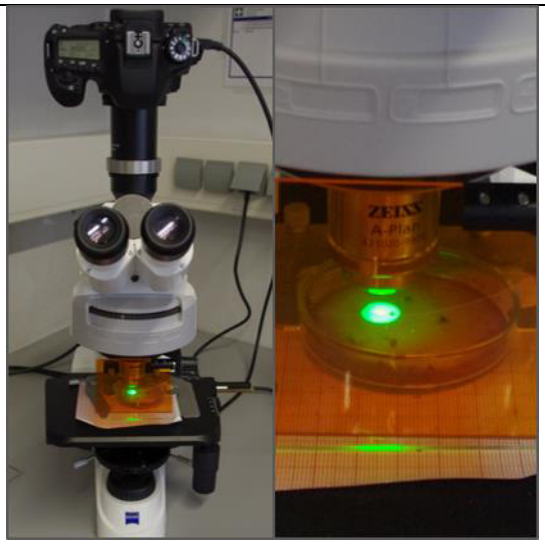
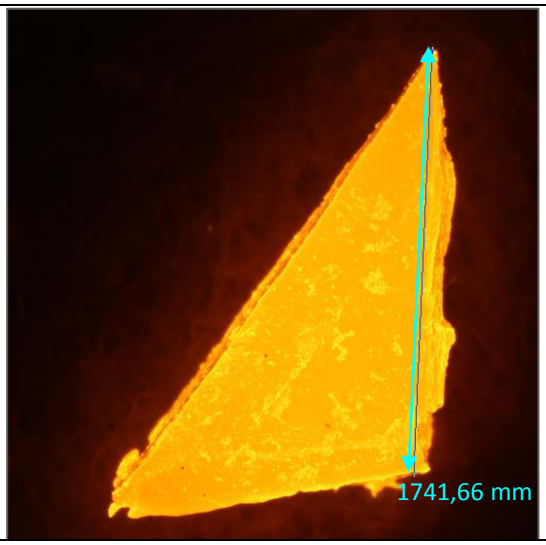
S2: Correlations between the number of particles in the UV light photobox and under the fluorescence microscope according to size class and shape.

	Size Class				
Shape	> 1.0 – 5.0 mm	> 0.63 - 1 mm	> 0.3 - 0.63 mm	> 0.2 - 0.3 mm	> 0.063 - 0.2 mm
Fragments	0.75	0.73	0.74	0.69	0.82
Fibers	0.76	0.87	0.89	0.74	0.69

S3: Step-by-step documentation with photoguide of the staining process using Nile Red and the UV light Photobox and/or a fluorescence microscope according to Tamminga et al. (2017). If you cite this method please additionally refer to this original publication (*Tamminga, M., Hengstmann, E., Fischer, E.K., 2017. Nile Red Staining as a Subsidiary Method for Microplastic Quantification: A Comparison of Three Solvents and Factors Influencing Application Reliability. SDRP Journal of Earth Sciences & Environmental Studies 2. https://doi.org/10.15436/JESES.2.2.1*).

Sample preparation The sample is transferred onto paper filters (VWR, qualitative filter paper 413, 5-13 µm particle retention), placed in glass petri dishes and dried for at least 48 hours at room temperature.	
Staining solution preparation 1 mg Nile Red ($C_{26}H_{18}N_2O_2$, ultrapure, *Roth Art.No.: 7726.3*) is used per 1 ml Chloroform ($CHCl_3$, stabilized with about 0.6% ethanol, *AnalaR NORMAPUR*) and these are mixed in a volumetric flask. The solution is transferred to brown glass bottles for storage.	

Staining Filters are stained with 1 ml of Nile Red solution in two sequences of 0.5 ml each.	
Drying Filters are dried, covered with watch glass, for at least 24 hours at room temperature.	
for MP > 630 µm	
Photographing in the UV light Photobox Filter is photographed in the UV light Photobox with a digital camera (*here: Pentax K-30*) using remote release. The following camera settings are applied: - exposure time 2'' - ISO 100 - minimum resolution of 2420x2343 One picture per filter is taken.	

Counting particles Each filter photo is evaluated using an image analysis software. Particles are counted at a zoom step of 150 % distinguishing different shapes of microplastics.	
<u>for MP ≤ 630 µm</u> *(also possible for larger particles)*	
Photographing with fluorescence microscope Filter is photographed under fluorescence microscope (AxioLab 2.0, Zeiss, a specifically mounted light source (AHF) and TRITC HC filterset, 2.5x magnification) with a digital camera (*here: Canon EOS800*) with the following settings: - exposure time 1” - ISO 500 - minimum resolution 1x1 µm. Several pictures per filter are taken using specified y- and x-offsets to create partly overlapping pictures.	
Counting particles All filter photos are evaluated using an image analysis software and its measuring tool at a zoom step of at least 25 %. Particles are counted and measured (1 pixel = 1 µm if resolution is 1x1 µm) distinguishing different shapes of microplastics.	

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Anthropogenic litter in freshwater environments – Study on lake beaches evaluating marine guidelines and aerial imaging

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ABSTRACT

Studies on macroplastic pollution in freshwater systems are rare compared to the marine environment. Nevertheless, freshwater systems are worthy to be equally investigated as they are pathways of plastic to the ocean and lakes may act as (temporary) sinks. The aim of this study was to identify sources for plastics and influences on its distribution in a limnic environment. Anthropogenic litter (>5 mm) was monitored semi-annually over a three-year period at four sandy bank border segments of Lake Tollense in Mecklenburg-Western Pomerania, Germany. The selected beaches represent different expositions and vary in their level of anthropogenic activity. Considering all six samplings, mean abundance of anthropogenic litter is 0.2 ± 0.1 items/m^2 or 130.9 ± 91.0 items/100 m beach length. The averaged mass of anthropogenic litter is 0.5 ± 1.0 g/m^2 or rather 218.7 ± 284.6 g/100 m. Plastic consistently is the predominate material (72%) and cigarette butts are the most found items. A higher pollution by anthropogenic litter is found at the end of tourist season unveiling the impact of anthropogenic activity on litter abundance. Additionally, litter transport via tributaries into the lake plays a role.

Testing the detection of anthropogenic litter via aerial images taken by unmanned aerial vehicles resulted in good recovery rates when minimizing the flight height. Furthermore, the analysis of anthropogenic litter distribution displayed on the images showed litter accumulation areas at the border of sandy beach areas. The deployment of marine guidelines in a freshwater environment did work well, however, small changes in the protocol are suggested for future lake beach studies dealing with anthropogenic litter pollution.

1. Introduction

Macro- and microplastic research has become an established field in environmental research and environmental plastic pollution is already considered a critical problem (UNEP, 2014). Plastic has been produced since the mid-1950s and its production rates steadily increased up to 359 million tons in 2018 (PlasticsEurope, 2019). Due to its longevity and low decomposition rates (Andrady, 2015), in combination with mismanaged waste and littering the plastic pollution of the environment is also increasing.

When investigating the plastic pollution of ecosystems, studies distinguish between micro- and macroplastics, commonly drawing the line at 5 mm (e.g. Arthur et al., 2009; Moore, 2008). More detailed size classifications are available, for example distinguishing between micro- (<5 mm), meso- (5–25 mm) and macroplastics (>25 mm) (Wagner et al., 2014; MSFD 2013). The size of plastic influences the impact plastic litter can have on organisms. Small plastic particles are expected to be ingested more often by aquatic organisms, while larger items may lead to entanglement of organisms (e.g. Andrady, 2011; UNEP, 2005; Thompson et al., 2004).

Considering published studies on macroplastic occurrence, studies on freshwater ecosystems are less frequent, compared to marine studies (Blettler et al., 2018; Dris et al., 2015). This is also true for studies about the impacts of macroplastic on aquatic organisms (Blettler and Wantzen, 2019). Beach cleaning programs were established for different marine regions that regularly collect data on beach litter in coastal areas (e.g. MARLIN, 2014; OSPAR, 2010; Marine Litter Watch by the European Environment Agency). The dominance of marine macro- and microplastic pollution studies established the term “marine debris” for persistent items manufactured and discarded by humans in the marine or coastal environment (UNEP, 2005). This term, however, points to marine environments as source areas and, therefore, cannot be applied to freshwater environments. With this in mind, Hoellein et al. (2014) suggested using the term anthropogenic litter (AL) instead, since it doesn’t indicate a specific environment, but includes different ecosystems, and still distinguishes human made material from natural debris.

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Freshwater studies on AL pollution may investigate the water surface layer (e.g. Faure et al., 2015; Eriksen et al., 2013), bottom (e.g. Turner et al., 2019; Ngupula et al., 2014) or beach sediments (e.g. Dalu et al., 2019; Zbyszewski et al., 2014; Faure et al., 2012). Most studies analyzing these freshwater compartments focus on microplastic abundances while data on macroplastic occurrence is lacking (Blettler et al., 2018). Only few litter monitoring programs exist for freshwater systems (e.g. Adopt a beach program and Great Canadian Shoreline Cleanup at the Laurentian Great Lakes, RIMMEL by the European Commission).

The current state of research for plastic pollution in freshwater environments reveals the need to compile data on AL, especially macroplastic pollution in lakes, and to develop similar monitoring programs as established in marine environments.

We analyzed the occurrence of AL, including macroplastic (>5 mm), at four beaches at Lake Tollense, Mecklenburg-Western Pomerania (MV), Germany, over a three-year period to i) gather data on AL in freshwater environments, specifically lakes, ii) investigate abundance and distribution of AL at lake beaches, iii) test AL detection via image analysis using an unmanned aerial vehicle (UAV) and Geographic Information Systems (GIS) and iv) develop a customized protocol for lake beaches similar to marine protocols.

2. Materials and methods

2.1. Study area

The study was conducted at Lake Tollense in MV in the northeast of Germany. Lake Tollense was chosen due to its simple, longitudinal shape. Beach segments covering different expositions and varying anthropogenic influences are available. The Tollense basin, including Lake Tollense, is a residue of ice-age melt waters (STALU MS, 2013). Lake Tollense has a length of 10 km and a width of 2.4 km at its maximum, resulting in a surface area of 17.9 km^2. The depth averages 17.3 m while the maximum depth is 31.2 m. The catchment area covers 515 km^2 and is dominated by agriculture and forests (Nixdorf et al., 2004). Lake Tollense has several tributaries entering the lake, mainly in the northeast and south. The major influents are the Nonnenbach with 0.57 m^3/s in the southeast of the lake and the Gaetenbach (0.55 m^3/s) in the northeast. The only effluent, the Tollense, is located in the north (Nixdorf et al., 2004) and flows into the Peene at Demmin which drains into the Baltic Sea.

Four bank border segments were selected being characterized by sandy substrate and varying expositions. The sample sites are shown in

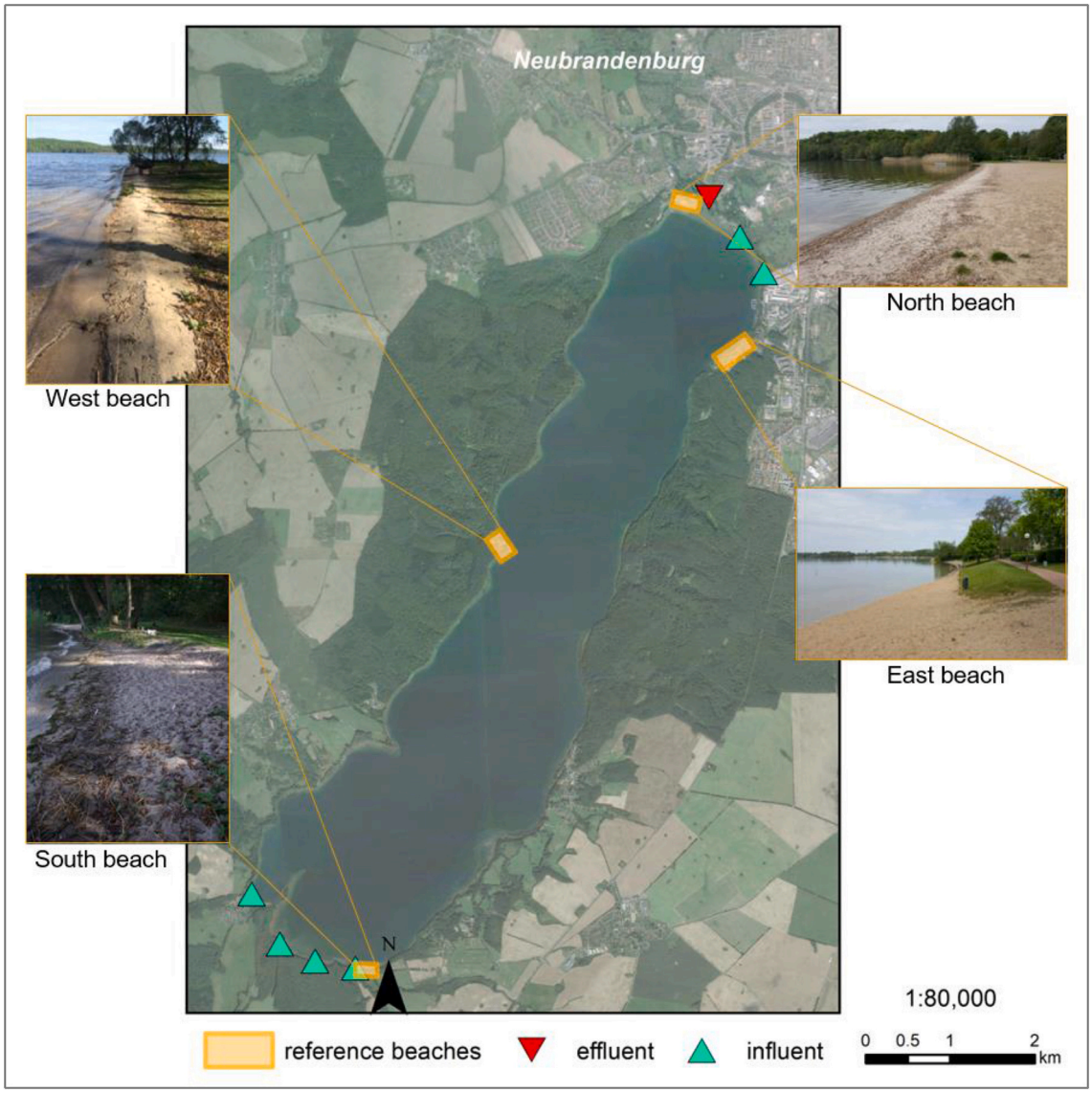

Fig. 1. Investigated beaches at Lake Tollense with locations of tributaries. Transverse Mercator. Coordinate system: WGS_1984_UTM_Zone_32N. Sattelite image: ArcGIS Imagery.

Fig. 1. The most northern beach, called North beach in the following, is located closest to the city of Neubrandenburg (ca. 1.5 km straight-line distance from the city center) and faces south-southeast. Neubrandenburg is the third most-populated city (ca. 64,000 in 2019) in MV and covers an area of 8611 ha (LAiV (Landesamt für innere Verwaltung Statistisches Amt), 2019). The analyzed North beach, composed of three smaller sandy subsegments, has a total area of 1253 m^2 on average. For one subsegment, obstacles (beach volleyball field and climbing frame) were present so that the investigated beach area was restricted to a width of ca. 14 m. The area of all investigated beaches varied between sampling times (see Table 1) due to differing water levels and wind conditions. Due to its proximity to Neubrandenburg, North beach is frequently visited by inhabitants and tourists and is used for sunbathing, bathing and sport activities. Furthermore, a children's playground is located close by. Two jetties of different lengths stretch into the water close to this beach.

The investigated beach at the eastern border of the lake is located within the outskirts of Neubrandenburg (ca. 2.5 km straight-line distance from city center). Still, the beach is frequently used as a sunbathing and bathing spot by inhabitants and tourists. A restaurant is located close to this beach. The beach segment in front of the restaurant was excluded from this study. The analyzed segment faces west and has an average area of 1218 m^2.

The beach at the southern end of the lake is located within a nature reserve. Trees split the beach segment into two sections covering a total area of 76 m^2 on average. The beach, which faces northwest, is impacted by the stream Nonnenbach entering the lake at this point and by a pier where a ferry runs on Lake Tollense from May to the beginning of October (neu.sw, 2020). The closest village is Nonnenhof (ca. 1 km straight-line distance).

The West beach is located on the western border of Lake Tollense on a small headland and campground area. It is used for sunbathing and bathing by campers and day visitors in the summer months. Furthermore, boat mooring is available here. The beach is segmented in three parts of which two are facing in a north-northeastern direction, the third one faces southeast. All segments together cover an average area of 518 m^2. The closest village is Neu Rhäse (ca. 2 km straight-line distance).

2.2. Data acquisition

Data acquisition in the field was conducted semi-annually in March and September beginning in September 2017 and ending in March 2020, resulting in six sampling dates. At the four beaches the same segment was therefore investigated three times in September (2017, 2018, 2019) and three times in March (2018, 2019, 2020). Sampling in March occurred before seasonal tourism and towards the end of winter storms. In contrast, September is the end of seasonal touristic activity and autumn storms are not yet occurring frequently. Furthermore, the longtime water level of Lake Tollense is highest in February and starts decreasing in March. In September, the water level starts to increase after its minimum in August (Pegelportal MV, 2020).

For litter monitoring on marine beaches, the OSPAR convention recommends, ideally, studying a 100 m-stretch of sandy beach not influenced by buildings and other litter collection programs (OSPAR, 2010). Finding sandy bank border segments of at least 100 m was not always possible at Lake Tollense. For North and East beach, the criterion was fulfilled, while South and West beach are shorter than recommended. South beach is not cleaned by other litter collecting programs, while beach litter at North and East is collected daily or weekly during tourist season from mid-May to mid-September. West beach is cleaned by the campground team during tourist season. All beaches are free from buildings.

Beaches were studied by two to three researchers looking for AL while moving in transects from the shoreline to the back of the beach and vice versa. Litter items were collected, classified and coordinates were registered using a D-GPS device (Trimble Geo 7 ×). Collected litter was transported to the laboratory in litter bags, dried at 40 °C for several days and weighed.

AL items at all beaches were recorded according to the litter list provided by MSFD (2013) which was developed for litter monitoring at marine beaches. This protocol distinguishes between different material groups and enables a detailed categorization of litter items on account of their function and/or origin. The protocol furthermore categorizes litter items into different size classes. Not only macro items (>25 mm) are included in the list but also meso items (5–25 mm). In some cases, micro items (pellets) may be identified. Litter items that were not assignable but could be identified, were classified into "others" and a comment was included describing the item. For each item, a tally list was compiled for each beach segment.

2.3. Aerial images

Aerial images of all beaches were recorded using an unmanned aerial vehicle (UAV; DJI Phantom II Vision+ and DJI Phantom IV Pro) including a camera (resolution 14 and 20 mega pixel) mounted via gimbal. Images were taken, with gimbal position at 90°, in different heights depending on the size of the beach. Poles were placed in all corners of the beach segment and the central point was labeled. These points were calibrated using the D-GPS device to be able to georeference images retrospectively via ArcGIS (©ESRI, Version 10.3). The gained images were used to locate litter coordinates. Furthermore, beach areas and strandline locations were estimated from these images again using ArcGIS.

Due to malfunction of the UAV, no aerial images could be taken at North, East and West beach in September 2018 and information on area and strandline location at these beaches at this time is missing but was approximated according to images from other sampling dates and macrolitter coordinate distribution.

Table 1
Information on characteristics of the four investigated beach segments at Lake Tollense. Width variation depends not only on seasonal changes but also on point of measurement.

Beach	Coordinates **(X, Y; UTM 32N)**	Length [m] **(min, max, mean)**	Width [m] **(min, max, mean)**	Area [m^2] **(min, max, mean)**	Orientation	characteristics
North	781045.19	126.7	3.8	1090.6	SSE	Close to Neubrandenburg;
	5941828.95	146.8	13.4	1569.0		High anthrop. use
		139.0	7.3	1253.2		
East	781612.24	154.2	3.5	766.6	W	Close to Neubrandenburg;
	5940052.37	163.0	18.1	1789.6		High anthrop. use
		158.9	9.5	1218.2		
South	776955.52	30.0	1.5	44.3	NW	In nature reserve, low anthrop. use
	5932780.79	38.4	5.3	109.0		
		33.8	3.6	75.6		
West	778851.30	80.6	0.8	292.1	NNE/SE	On campground, med. anthrop. use
	5937754.46	132.0	14.6	697.1		
		102.7	6.9	517.5		

Additionally, images via UAV were taken to test automatic and supervised image analysis for plastic detection in March 2019. The DJI Phantom IV Pro with the RGB-camera FC6310 by DJI (4K resolution) was used providing images with horizontal and vertical dimension of 5472 × 3078 pixels. Colored (red, blue, green, yellow, transparent) plastic shreds of various size were prepared in laboratory by cutting plastic folders manually. A total number of 70 squares were prepared: three of each color were 10 × 10 cm, four of each color were 5 × 5 cm and seven of each color were 2.5 × 2.5 cm in size. These were placed randomly within a definite area of 10 × 10 m at North beach. A smaller area of 5 × 5 m was separately established within the large quadrat. Images were taken in different heights (7 m, 10 m, 15 m, 20 m, 30 m, 50 m, 80 m) covering at least the 25 m^2 quadrat. These aerial images were analyzed via unsupervised and supervised classification using SAGA GIS (Version 7.6.2). The tool "K-Means Clustering for Grids" was selected for the unsupervised classification using a combined classification method based on Minimum Distance by Forgy (1965) and Hill-Climbing according to Rubin (1967). The number of iterations was set to 10 with no normalization prior to classification. For this method the number of targeted classes is set prior to the analysis. Here, 4, 10, 15 and 20 classes were specified. For the supervised classification for grids, the algorithm "Minimum Distance" was chosen to classify images. This algorithm uses the Euclidean distance from the cluster center to classify pixels (Tempfli et al., 2009). The threshold was set to "0" to avoid an "unclassified" class. Prior to the analysis, training areas were defined for all colors of plastic squares as well as the beach surface in general, organic material and shades.

2.4. Statistical analysis

Tally lists containing litter amounts were transferred into actual numbers in spreadsheets using Excel (© Microsoft Office, 2013). Two lists were compiled including a list containing "new" identified litter items and a list including all litter items not found. Statistical analyses were performed using R statistics (R Core Team, 2017, Version 3.4.2) in an R Studio environment (RStudio Team, 2016, Inc., Version 1.1.383). For testing significant differences between groups, the Kruskal Wallis test with post-hoc Dunn test was applied. Relations between anthropogenic litter abundance and wind data were tested using the Pearson correlation index.

Visualization was also done using R statistics in an R Studio environment including the library ggplot (Wickham, 2016) and ArcGIS for map visualizing.

3. Results

3.1. AL abundance and distribution

AL items were ubiquitous at Lake Tollense beaches. In total, 4346 items weighing 4.7 kg were identified at all beaches considering all sampling dates. Abundances varied between the beaches and sampling dates. In March 2019, a cluster of confetti pieces was found at East beach comprising 513 items and leading to an increased pollution by AL at this time and beach. Since this finding only occurred once during the monitoring period and was spatially restricted, these confetti items are treated as outliers and were excluded for the following presentation of results concerning numbers of litter items.

Litter items found at the individual beaches considering all sampling dates varied between 9 and 638 items with a mean (±standard deviation) of 160 ± 149 and a median of 128 items. North beach showed the highest occurrence of litter items while at South beach the lowest number of items was registered. Results for all beach segments and sampling times presented as AL per 100 m beach length and AL per m^2 are presented in the Supplementary Material SM1 and Fig. 2. Litter items per 100 m beach length varied between 26.1 and 441.8 at all beaches over all sampling dates with on average 130.9 ± 91.0 items/100 m (median: 110.3). North beach in September 2019 showed the highest pollution by AL, while the lowest abundance was registered for West beach in March 2020 (see SM1). The number of litter items per m^2 varied between 0.1 and 0.6 at all beaches over all sampling dates resulting in a mean abundance of 0.2 ± 0.1 items/m^2 (median 0.2). The highest and lowest abundance was registered for South beach in March 2020 and West beach in March 2020, respectively (see Fig. 2 and SM1).

In averaging the items identified over the sampling dates, North beach was most polluted by litter items related to beach length, while South beach showed the highest pollution related to area (see Table 2).

The highest weight of AL was found at South beach in March 2019, while the lowest amount was registered for East beach in September 2017 (see SM1) resulting in 218.7 ± 284.6 g/100 m (median 126.1) or 0.5 ± 1.0 g/m^2 (median 0.2). When averaging mass values over sampling dates, as visible in Table 2, South beach showed the highest accumulation of litter related to beach length and area, respectively. The lowest pollution is reported for East beach.

Comparing seasons, significantly higher amounts of litter were identified for samplings in September compared to March when considering raw counts ($p < 0.001$), items/100 m ($p < 0.01$) and items/m^2 ($p < 0.05$; see Table 2). Besides, no significant differences were verified between the four beaches except for values based on raw counts ($p < 0.001$). North and East beach showed significantly higher numbers of litter items compared to South and West beach, however, these beaches also cover the greatest areas.

When categorizing litter items according to their source material, plastic was the dominant type for all beaches at all sampling dates. In total, plastic items made up 71.5% of all items, followed by glass (9.7%), paper (7.4%) and metal (7.1%). Rubber, clothes, and wood items only accounted for 1%–2%. The plastic share varies between 42.2% for West beach in March 2019 and 90.74% for South beach in September 2018. Plastic was especially dominant in all sampling dates at the northern and eastern beach, while at the southern and western beach, materials such as glass, metal and especially paper partly constituted higher shares (see Fig. 3).

A list of the top ten items was compiled that considered all beaches and sampling times. Cigarette butts and filters occurred most frequently, constituting 39% of all items, followed by plastic and polystyrene pieces (0–2.5 cm), which represent 6% of all items. Glass or ceramic fragments (>2.5 cm) made up 5% of all items while items in position 4 to 10 constitute less than 5% each. The total of top ten most common items account for 77% of all items registered. At all beaches, cigarette butts and filters were the top item. Similarly, cigarette butts and filters were the items found most often on all sampling dates. The following items in

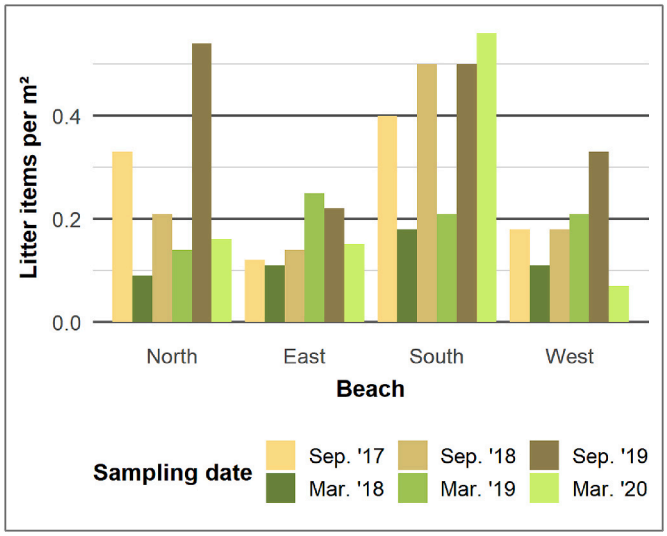

Fig. 2. AL abundance in items/m^2 according to beach and sampling date.

Table 2
AL quantities at the investigated beaches of Lake Tollense averaging over all sampling dates and AL quantaties for two seasons averaging over all beaches. Values display mean ± standard deviation and median. Letters a and b indicate significant differences between seasons.

		items/100 m	items/m^2	g/100 m	g/m^2
Beach	*North*	213.1 ± 132.0 184.4	0.2 ± 0.2 0.2	186.1 ± 115.3 147.4	0.2 ± 0.2 0.2
	East	129.7 ± 63.3 124.4	0.2 ± 0.1 0.1	80.7 ± 89.5 49.2	0.1 ± 0.1 0.1
	South	86.1 ± 40.4 91.0	0.4 ± 0.2 0.5	351.0 ± 494.0 111.1	1.4 ± 1.8 0.6
	West	94.8 ± 53.3 105.0	0.2 ± 0.1 0.2	257.0 ± 245.9 147.1	0.5 ± 0.4 0.3
Season	*March*	85.4 ± 51.7[a] 77.0	0.2 ± 0.1[a] 0.2	209.0 ± 349.2 109.3	0.6 ± 1.4 0.2
	September	176.4 ± 100.3[b] 152.2	0.3 ± 0.2[b] 0.3	228.4 ± 217.4 133.4	0.5 ± 0.5 0.2

the top-ten list vary slightly between single beaches and sampling dates. However, plastic/polystyrene pieces, glass pieces, or paper pieces, are often occurring.

Styrofoam (pieces) and industrial pellets (microplastic) were rarely found at the beaches of Lake Tollense with total numbers of 7 and 57 items, respectively, including all beaches at all sampling dates. North beach showed the highest pollution in these categories with on average 0.8 pellets and 8.7 styrofoam pieces per sampling.

For some identifiable items, no appropriate category existed in the MSFD list. Frequently occurring items that could not be assigned to a category were paint flakes (24), tissues (17), tags (14) and fireworks (11). In case of fireworks, a category exists for the paper material group. Fireworks made from artificial polymer materials (probably belonging to New Years rockets) were found, though. In some cases, e.g. for sticking plaster or threads, only related or generic categories were available. In contrast, several items included in the MSFD list were not found in our study. Items related to professional fishing and shipping were especially not present at Lake Tollense beaches.

Circa 5% of all AL items may be classified into source groups according to OSPAR (2007). Accordingly, "Public littering" (70%) is the dominant source for macrolitter at beaches of Lake Tollense, followed by fishing (15%) and shipping related sources (11%). "Sanitary and Medical waste" and "Galley waste" add up to 4%. The overall dominance of "Public littering" as source for AL is also true regarding the four beaches separately and considering the different sampling dates individually.

The distribution of AL within the beach area was analyzed using aerial images taken via UAV and geographical coordinates registered via D-GPS device (Fig. 4). No specific and recurring hot spots could be detected. However, AL seemed to accumulate along the border of the beaches where sandy substrate turns into vegetated areas (mostly meadows or reed/shrubs). A virtual buffer zone of 0.5 m was created in ArcGIS, covering the area at the border of the beaches. Within this zone, 2.8%–66.7% of items (mean: 24.8%) were registered. At the southern beach, the highest accumulation at the border of beach area was registered followed by the western beach. The North and East beach showed lower values (see Table 3). However, no significant differences were recognized between the beaches or in the variation between sampling dates.

Similarly, a zone for the (latest) strandline was created in ArcGIS according to visible contours on aerial images to estimate the accumulation of items at the strandline. In total, 0%–35.5% of items (mean 7.7%) were located at the strandline of the respective beach. The southern beach showed the highest accumulation rates at the strandline. At the eastern beach, the percentage of AL at the strandline was lowest (see Table 3). Again, no significant differences could be proven between beaches or sampling dates for strandline accumulation.

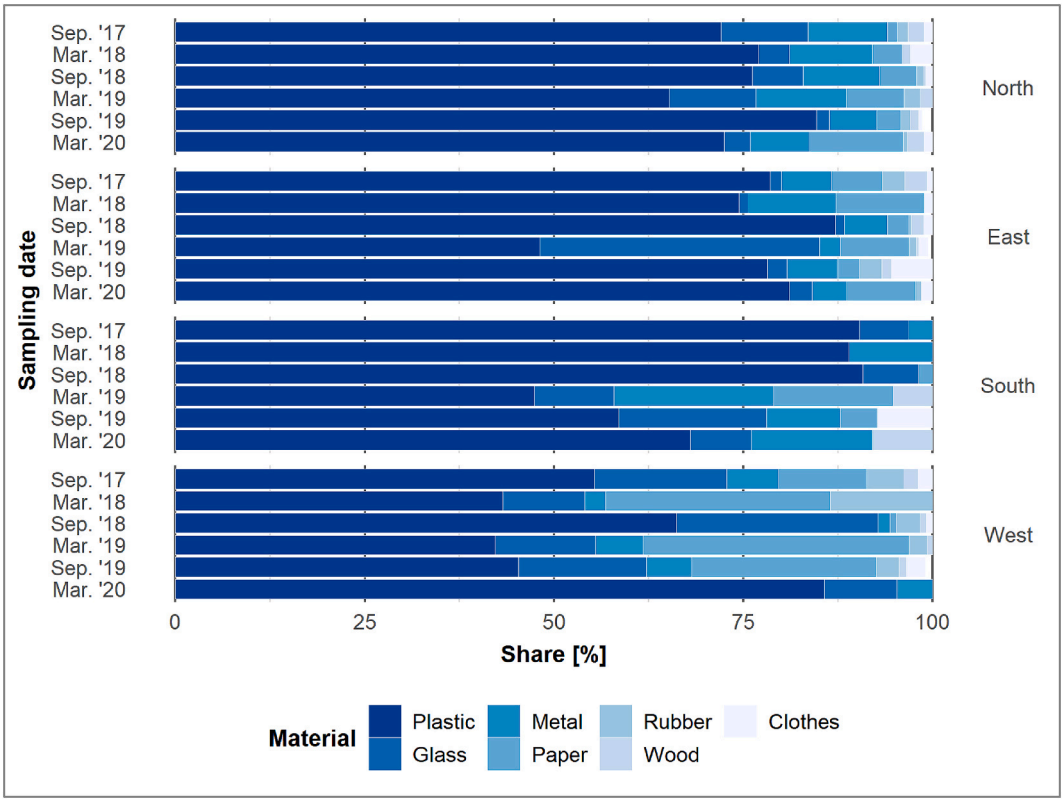

Fig. 3. Shares of different materials at the investigated beaches at Lake Tollense including all sampling times considering number of AL items.

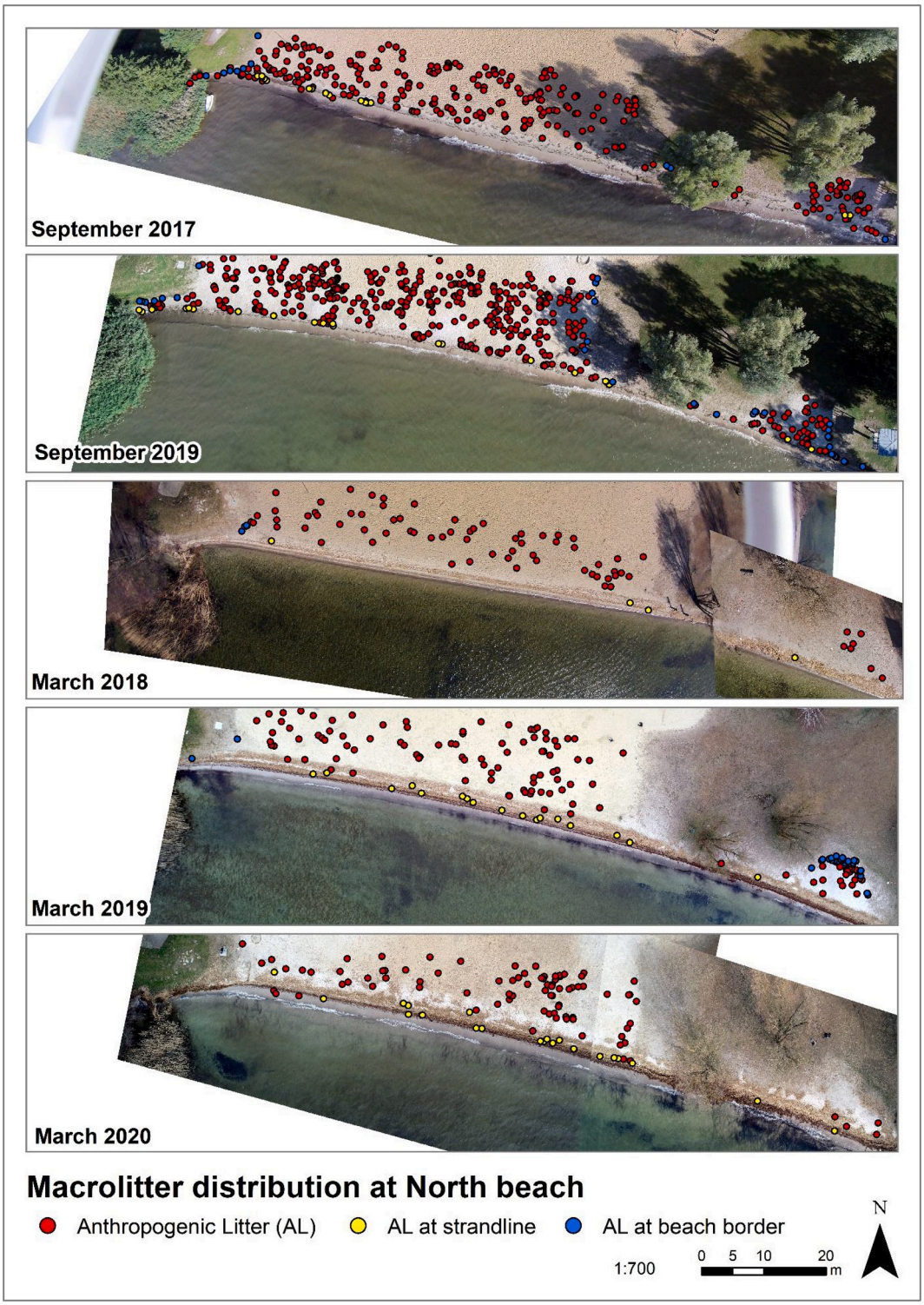

Fig. 4. Anthropogenic litter distribution visualized using aerial images taken by UAV and D-GPS coordinates for North beach at Lake Tollense.(Note: Due to malfunction of the UAV no aerial image is available for the sampling in September 2018). Projection: Transverse Mercator. Coordinate system: WGS_1984_UTM_Zone_32N.

The influence of wind velocity and direction on anthropogenic litter abundance was tested using climate data provided by DWD (2020). Wind velocity was averaged (mean and median) for two weeks and one week prior to the last sampling day. No significant correlations were registered between wind velocity and items/100 m, items/m^2 in total as well as items/m^2 at each beach for both time sequences. The mean wind velocity at North Beach showed the only consistent but not significant correlation within this analysis with $r = -0.64$ ($p = 0.17$) and $r = -0.65$ ($p = 0.16$), respectively.

3.2. Analysis of aerial images

Images taken by UAV in different heights covering the quadrat test areas on North beach, which were spiked with plastic shreds in March

Table 3
Accumulation rates for different zones for all beaches investigated at Lake Tollense. Values display mean ± standard deviation and median.

		Strandline accumulation (%)	Border accumulation (%)
Beach	*North*	7.1 ± 5.6	16.0 ± 3.8
		5.4	17.6
	East	1.7 ± 2.5	15.0 ± 6.7
		0.3	17.1
	South	14.0 ± 12.9	43.9 ± 18.1
		11.0	34.4
	West	6.6 ± 7.0	24.6 ± 7.0
		3.8	25.7

2019, were analyzed via SAGA GIS. First, an unsupervised classification was tested to detect the prepared plastic shreds within the 5 × 5 m quadrat. In using only four categories for the unsupervised classification, plastic pieces with colors blue, green and red, were partly visible. However, the smaller the size, the more difficult the correct detection. Yellow and transparent shreds were seldomly recovered due to "background noises" produced by the beach surface. In allowing 10 classes, a similar result was obtained with less background noise, though. The result was further improved when using 15 classes. Here, different colors were also detected within different classes (see Fig. 5). Furthermore, yellow and transparent particles, and smaller plastic shreds, were more visible. However, variances in beach surface were still distracting. Nevertheless, 11 out of 20 plastic shreds were easily visible in the classified aerial image. Eight plastic shreds were detectable in parts and only one transparent plastic square of 2.5 × 2.5 cm in size was not recognizable. When further increasing the number of classes to 20, results were partly impaired again (colors were not completely distinguishable anymore). Additionally, a higher number of classes increases the attribution of classes to shreds of different color or beach surface.

Subsequently, all images taken in different heights were analyzed via supervised classification. In Table 4, recovery rates for UAV images taken at different heights are presented according to the color and size of test plastic shreds. These show perfect recovery rates for the image taken at 7 m. At 10 m, recovery rates already start to decrease for transparent and the smallest plastic shreds. This trend continues for increasing heights with other colors being similarly affected in their recovery rates as well as larger plastic shreds. In general, results show, that classification accuracy decreases as the distance to the ground increases. The resolution of images consequently decreases and leads to an increased background noise in the classification which again hampers the detection of plastic particles. Transparent particles are especially affected. For heights of 50 m and 80 m, the green and red class also produces considerable background noise. The size of plastic particle also plays a

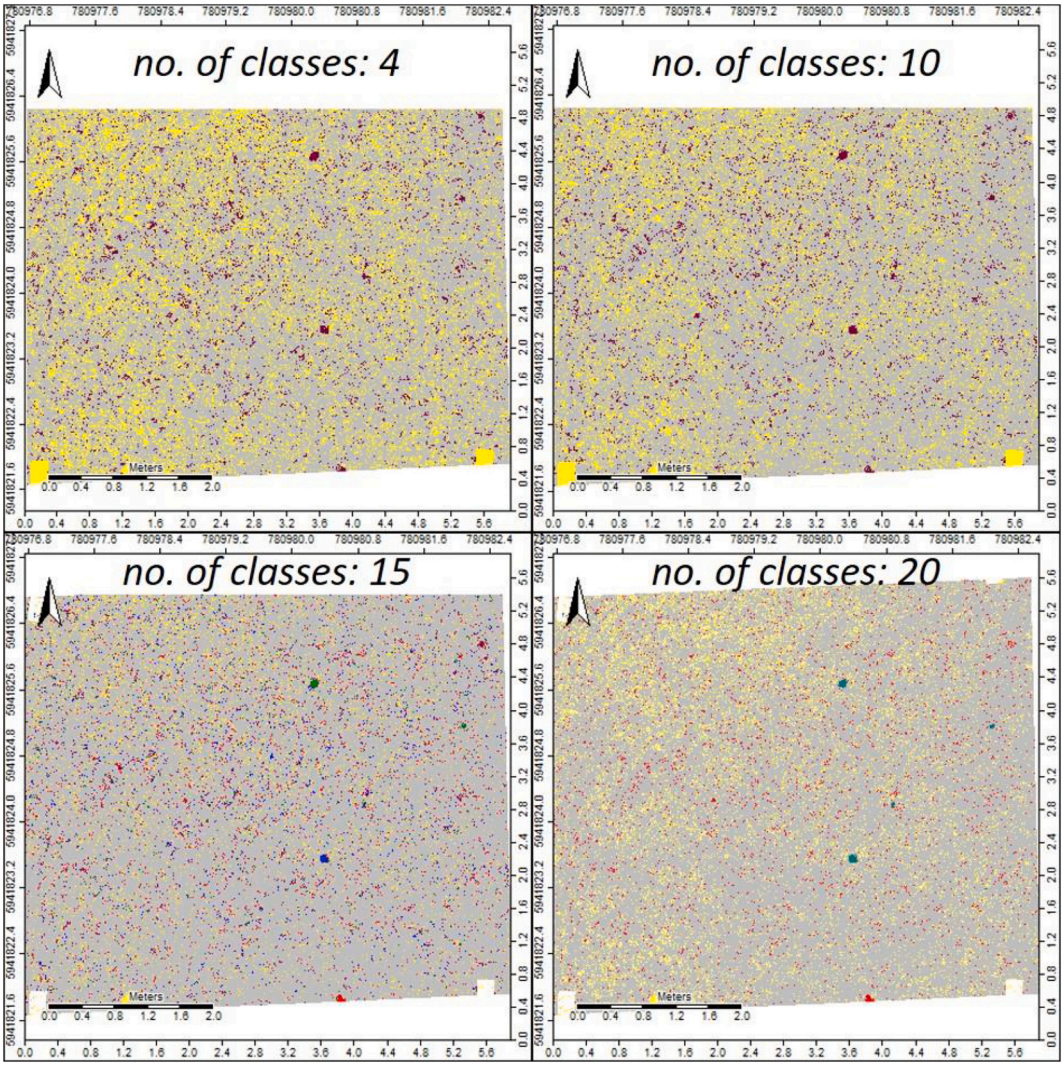

Fig. 5. Results of unsupervised classification with varying numbers of classes. Grey = beach surface, yellow = yellow/white/transparent plastic shreds, purple = red/green/blue plastic shreds for n = 4 and n = 10, green = blue and green plastic shreds for n = 20. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 4
Recovery rates of plastic shreds of different colors and sizes in aerial imaged taken at different heights. Percentages in parentheses present partly detectable plastic shreds. For heights 7 m and 10 m, only the small test area (5 × 5 m) was visible including 20 plastic shreds. 70 plastic shreds were distributed within the large test quadrat (10 × 10 m).

	color					size		
	blue	green	red	yellow	transparent	25 cm^2	50 cm^2	100 cm^2
7 m	100%	100%	100%	100%	100%	100%	100%	100%
10 m	100%	100%	100%	100%	67% (33%)	80% (20%)	100%	100%
15 m	79% (21%)	79% (21%)	100%	100%	57% (14%)	69% (23%)	95% (0%)	100%
20 m	57% (43%)	50% (21%)	71% (29%)	100%	43% (21%)	31% (46%)	95% (0%)	100%
30 m	50% (14%)	43% (7%)	43% (43%)	64% (36%)	14% (14%)	9% (34%)	65% (20%)	93% (0%)
50 m	36% (14%)	36% (14%)	36% (29%)	57% (29%	7% (7%)	6% (14%)	45% (35%)	87% (7%)
80 m	21% (21%)	7% (43%)	29% (21%)	36% (36%)	0% (14%)	0% (9%)	25% (50%)	53% (40%)

role in the detection accuracy. For images taken at low heights, even the smallest plastic shreds were distinguishable from the beach surface. In heights of 50 m and 80 m, only the largest plastic squares are unambiguously visible in the pictures.

4. Discussion

4.1. Litter abundances at Lake Tollense beaches

Different units in the presentation of results were used in this study to allow the comparison to former studies. Several studies use items per m^2 to present results while others have reported AL amounts per beach length or in mass of items (Hoellein et al., 2014). However, different units may also lead to varying interpretations of data (e.g. Hengstmann et al., 2017; Hoellein et al., 2014). The results for Lake Tollense beaches presented in mass reveal no distinctive pattern in AL distribution between beaches or seasons. In contrast, these results seem to highly depend on individual or few items showing high weights. Furthermore, abundances at beaches vary between results as items per m^2 and items per 100 m beach length. When measuring items per beach length, significant differences were proven between individual sampling dates, while this was not possible for items per m^2. The unit items per m^2 is more reliable, though, since a second dimension (the width of beach) is included. The width of the investigated beach may range greatly so that a comparison to other studies is hampered when using values related to length. Czarkowski et al. (2016) also determined varying width of analyzed shorelines and concluded that results should therefore be presented related to area.

Less significant differences between the beaches might be further attributed to seasonal changes in beach area when results are presented in items/m^2. The aerial dimension of the four beaches slightly varied between sampling dates and might influence relative AL occurrence. The dimension of the beach depends on water level and wind direction. However, no significant difference in beach area could be verified between sampling times.

Themes are visible in the data, although there were no significant differences between beaches for results related to beach length or area. On average South beach is more polluted than the other beaches in terms of items/m^2 and g/m^2. The inflow of the Nonnenbach into Lake Tollense might lead to an input and washing ashore of AL items at this beach. Former studies also found an influence of tributaries on the abundance of macro- and microlitter at lake beaches (Corcoran et al., 2015; Faure et al., 2015; Imhof et al., 2013). Rivers are considered as transport pathways for plastic to the oceans (e.g. Jambeck et al., 2015) but in smaller dimensions they might also transport plastic into lakes. Other characteristics of the southern beach (within nature reserve and great distance from Neubrandenburg) might favor a lower pollution due to lower visitor rates in contrast to especially North and East beach (Zbyszewski et al., 2014). The vicinity to highly populated areas may increase plastic abundances in the water body and on shorelines of freshwater bodies as verified by various studies (Blettler et al., 2017; Hoellein et al., 2015; Eriksen et al., 2013; Faure et al., 2012). This influence cannot be verified for South beach at Lake Tollense.

In contrast, West beach shows low contamination by AL in terms of items/100 m and items/m^2. As part of a campground, this beach is regularly cleaned to maintain the attractiveness of the beach for recreational use. Furthermore, the West beach segments are shielded from direct wind inputs due to their exposition. The dominant long-time wind direction during the sampling period was southwest (DWD, 2020). Imhof et al. (2013) equally concluded that dominant wind and current direction may have an impact on litter accumulation on shores, showing that the wind exposed beach at Lake Garda was more polluted. Other studies similarly confirmed the influence of wind on AL abundance at lake beaches (Corcoran et al., 2015; Hoellein et al., 2014). No significant correlation between wind and litter data could be verified in this study, though. A tendency was visible for North beach AL abundances only showing a negative correlation with wind velocity, which means that AL abundance decreases for stronger winds. With a prevailing wind direction from the southwest during sampling dates the North and East beach are probably most influenced by wind due to their orientation. North beach is flat, and the surrounding area is nearly unvegetated. Therefore, strong winds may transport AL from the North beach into the hinterland. This transport mechanism is hampered at the other beaches due to more vegetated areas and greater slopes in case of East beach.

In terms of mass, East beach was found to be least polluted by AL items despite its exposition, vicinity to Neubrandenburg and high recreational activity. Daily beach cleaning by lifeguards during the tourist season seems to have a severe impact on the results at this beach, especially on the mass of AL. Beach grooming activities were proven to alter AL abundances (Hoellein et al., 2015, 2014). However, these are not as effective for small litter items (Driedger et al., 2015) but may focus on large and heavy items due to easier identification. This is underlined by high percentages of cigarette butts at the East beach. These are small, sometimes fragmented and have a similar color as the sand, so that they are consequently more rarely removed during beach cleaning activities.

The influence of different seasons on AL abundance was analyzed by former studies and did not result in significant differences between sampling times in some cases (Czarkowski et al., 2016; Dalu et al., 2019). In contrast, other studies revealed a correlation between season and litter amounts. The relation might be attributed to varying visitor abundances in different seasons (Hoellein et al., 2015). The latter is in accordance with this study when using numbers of litter items, also related to beach length or area. Significantly higher abundances were registered for the end of tourist season (September) compared to March, when AL abundance was monitored before the start of visitor activities. Sampling bi-annually over a three-year period (resulting in six-samplings) offers the opportunity to detect large-scale temporal variations and the comparison of AL abundances between two seasons. Small-scale temporal variations may not be detected by this method. Investigations of AL at beaches with higher frequencies (e.g. bi-weekly) as performed by Vincent and Hoellein (2017) and McCormick and Hoellein (2016) are needed in this case. The mentioned studies also found higher AL contamination in fall (beginning of September to

mid-November) or summer (end of May to mid-September), respectively.

Plastic was found to be the dominant material of AL at beaches not only at Lake Tollense but also at other freshwater bodies (Winton et al., 2020; Chapman, 2019; Czarkowski et al., 2016; Driedger et al., 2015). Plastic has low degradation rates (Andrady, 2015) and may therefore persist on beaches for long time if not removed. Plastic items on beaches may lead to entanglement of or ingestion by organisms (e.g. Collard et al., 2018; Scherer et al., 2017). Ingestion of plastic particles depends on their availability, size and physiological traits of the ingesting organism (Bellasi et al., 2020). For organisms living in the area of Lake Tollense (i.a. European kingfisher, medium spotted woodpecker, beaver, firebellied toad, curt, roach according to STALU MS, 2013), small particles (i.e. microplastics) are more likely to be ingested (due to the relatively small size of organisms) than for large particles (i.e. macroplastics) investigated in this study. De Sá et al. (2018) stated that the most common size of ingested plastic particles was 800–1600 μm in field studies. When ingested, plastic particles may transport toxic chemicals as well as other contaminants absorbed to the particles into organisms and the food chain (Bellasi et al., 2020). Therefore, the high degree of plastic litter on freshwater beaches poses an environmental threat.

The top ten items in total accounted for 77% of all AL, showing that a small number of items dominates AL amounts, which is in line with former studies (Winton et al., 2020). At Lake Tollense beaches, cigarette butts and filters were the most commonly identified item, which is also a frequently occurring or even dominant AL item at other lake shorelines (Araújo and Costa, 2019; Hoellein et al., 2015, 2014). A reason for the high amount of cigarette butts at lake beaches may be the increased in-situ littering due to recreational use (Hoellein et al., 2014). They are easily washed ashore or transported onto shorelines by waves, currents, and wind. Additionally, cigarette butts have low decomposition rates in terrestrial environments (Araújo and Costa, 2019). Not only in freshwater ecosystems but also at marine shorelines cigarette butts constitute a great challenge (e.g. Ocean Conservancy, 2011). Given the high number of cigarette butts on freshwater shorelines, the possible environmental threats should not be underestimated. Possible impacts include the ingestion of cigarette filters by biota, the release of toxic compounds, and negative social and economic effects (Araújo and Costa, 2019).

Other top ten items included plastic and polystyrene pieces as well as paper and glass fragments, while other studies showed a high density of plastic bottles or bags at freshwater shorelines (Winton et al., 2020; Chapman, 2019; Blettler et al., 2017; Czarkowski et al., 2016; Free et al., 2014). The high number of plastic and also paper and glass pieces, points to the fact that degradation is in progress and macroplastic items become microplastics (by UV-radiation and mechanical processes according to Andrady (2011)) leading to a microplastic pollution of beach sediments and lake water, when being transported.

Here, we concentrate on the comparison of AL abundances to other freshwater studies to provide similar environmental conditions. Though, other authors already stated that AL densities are similar for freshwater and marine ecosystems (e.g. Vincent and Hoellein, 2017). Comparisons between studies on AL abundances are hampered due to different units and methods applied during sampling. For example, Zbyszewski et al. (2014) did not categorize AL items according to their material or former usage, but distinguished pellets, fragments, styrofoam and intact/near intact debris. Zbyszewski et al. (2014) found a high proportion of pellets as well as styrofoam on the shores of some of the Great Lakes. Conversely, these items were rarely present at Lake Tollense. This might result from increased inputs due to plastic manufacturing industries around the Great Lakes (Corcoran et al., 2015; Zbyszewski et al., 2014) which are absent in the Lake Tollense catchment. Plastic pollution due to domestic instead of industrial waste seems to prevail at Lake Tollense shores due to the rare occurrence of pellets (Blettler et al., 2017).

Table 5 shows the results of the present study on Lake Tollense compared to selected studies on AL on lake beaches. For example, comparing the results of Lake Tollense to lakes in Switzerland (Faure et al., 2015) and Italy (Imhof et al., 2013), the latter are more polluted than Lake Tollense. Lakes investigated in Switzerland have a greater surface area and volume than Lake Tollense and local differences in anthropogenic impact as well as different waste management systems may contribute to differences in plastic abundance (Dris et al., 2015). Furthermore, different sampling methods have been applied. Faure et al. (2015) focused on quadrats at the drift line and aimed for simultaneously analyzing microplastic particles while the current study at Lake Tollense was focused on the detection of AL across the whole beach area. In contrast, water bodies in Poland investigated by Czwarkowski et al. (2019) and Lake Michigan in the US (Hoellein et al., 2015, 2014) showed slightly lower abundances. Again, local conditions and varying methods play a role. Furthermore, Hoellein et al. (2014) already stated that Lake Michigan beaches seem to be less polluted than other freshwater beaches.

Concerning the distribution of AL across beaches, again, a comparison between studies is hampered, since some studies concentrate on specific areas like the strandline (e.g. Blettler et al., 2017; Faure et al., 2015), while some consider a transect with a specific width (e.g. Owens and Kamil, 2020; Blettler et al., 2017) and others investigate the complete beach or at least randomly selected areas of a beach (e.g. this study, Chapman et al., 2019; Imhof et al., 2013). Still, higher accumulations of AL within the strandline were reported by several authors (Blettler et al., 2017; Hoellein et al., 2014; Zbyszewski et al., 2014) suggesting that items are transported and deposited onto the beach via waves (Zbyszewski et al., 2014). Corcoran et al. (2015) observed that small items (<10 mm) seem to be accumulated within organic debris at the strandline (like pellets and fragments) while larger debris is found across the whole beach. The latter is in accordance with the findings from Lake Tollense where the AL items were dominantly distributed over the complete area of the beach. In contrast to an accumulation at the strandline, an accumulation at the edges of the beach could be observed, where grass and low vegetation seems to trap items. The transport of AL items by wind within the beach area seems to play a more crucial role than the transport of items onto the shore by waves.

4.2. Anthropogenic litter monitoring via UAV

The automatic image analysis via supervised and unsupervised methods in SAGA GIS showed that drone images (4K resolution) may be used for the detection of AL on beach surfaces. While the unsupervised analysis of the image taken in 7 m height showed a recovery rate of 55% with high certainty and 40% with low certainty, these values were improved for the supervised classification with 100% of plastic shreds (as small as 2.5 cm) being detectable (7 m height). Therefore, a supervised image analysis is preferable concerning accuracy of classification. However, supervised classifications also require more input parameters (operator knowledge), such as the compilation of training areas (Tempfli et al., 2009). Plastic shreds of known color, size and number could routinely be distributed on the beach area before taking images by UAV for later using these as training areas.

The height in which images are taken by UAV should not exceed 10 m for 4K resolution since result accuracy rapidly decreases above this height. Therefore, several pictures are needed to cover a complete beach. For example, 12 images are at least needed to cover the complete beach area of North beach (1200 m^2) since the image of 10 m height shows an area of ca. 100 m^2.

The camera used in this study only provides RGB-composite images. Cameras also covering the near infrared (NIR) spectrum could further improve the results of the image analysis. According to Masoumi et al. (2012), plastic objects show absorbance and reflectance characteristics within this spectrum. Anthropogenic plastic litter in beach sediment using NIR reflectance spectrometry was successfully detected by Driedger et al. (2015). Using LIDAR technology for AL detection is another possible and promising tool, showing accuracy rates of 75.4%

Table 5
Comparison of AL quantities at Lake Tollense and other lake studies on AL and/or macroplastics, (n.r. = not reported).

Paper	Country	Study area	Lake characteristics (area/mean depth/ volume)	No. of beaches	sampling method	Substrate	Size definition	Abundances
Present study	Germany	Lake Tollense	17.9 km^2/17.6 m/ 0.316 km^3	4	complete beach area	sand	>5 mm	mean: 0.2 ± 0.1 items/ m^2 (0.5 ± 1.0 g/m^2) 130.9 ± 91.0 items/ 100 m (218.7 ± 284.6 g/100 m)
Imhof et al. (2013)	Italy	Lake Garda	n.r.	2	random grid sampling	sand	>5 mm	8.3 and 483 ± 236 particles/m^2
Czarkowski et al. (2016)	Poland	5 water bodies (Mazuria)	n.r.	5	water/land junction	n.r.	n.r.	0.0049 to 0.1756 items/ m^2
Faure et al. (2015)	Switzerland	Lake Brienz	29.8 km^2/n.r./ 5170 × 10^6 km^3	3	quadrats of 0.3 × 0.3 m at drift line	sand, gravel	>5 mm	mean: 400 ± 510 particles/m^2 (16 ± 12 g/m^2) median: 190 particles/ m^2 (18 g/m^2)
	Switzerland	Lake Constance	539.0 km^2/n.r./ 48,530 × 10^6 km^3	3	quadrats of 0.3 × 0.3 m at drift line	sand, gravel	>5 mm	mean: 8 ± 11 particles/ m^2 (1.3 ± 2.3 g/m^2) median: 6 particles/m^2 (170 g/m^2)
	Switzerland	Lake Geneva	581.3 km^2/n.r./ 89,900 × 10^6 km^3	3	quadrats of 0.3 × 0.3 m at drift line	sand, gravel	>5 mm	mean: 35 ± 65 particles/m^2 (18 ± 40 g/m^2) median: n.r.
	Switzerland	Lake Maggiore	212.3 km^2/n.r./ 37,100 × 10^6 km^3	3	quadrats of 0.3 × 0.3 m at drift line	sand, gravel	>5 mm	mean: 28 ± 42 particles/m^2 (26 ± 54 g/m^2) median: 11 particles/ m^2 (780 g/m^2)
	Switzerland	Lake Neuchâtel	217.9 km^2/n.r./ 14,170 × 10^6 km^3	3	quadrats of 0.3 × 0.3 m at drift line	sand, gravel	>5 mm	mean: 17 ± 26 particles/m^2 (6.4 ± 13 g/m^2) median: 6 particles/m^2 (49 g/m^2)
	Switzerland	Lake Zurich	68.15 km^2/n.r./ 3770 × 10^6 km^3	3	quadrats of 0.3 × 0.3 m at drift line	sand, gravel	>5 mm	mean: 3 ± 6 particles/ m^2 (0.7 ± 1.4 g/m^2) median: 0 particles/m^2 (0 g/m^2)
Free et al. (2014)	Mongolia	Lake Hovsgol	2760 km^2/262 m/ 483 km^3	9	between water and wrack line	n.r.	n.r.	5.24 items/100 m (132 g/100 m)
Zbyszewski and Corcoran (2011)	US	Lake Huron	n.r./60 m/n.r.	7	every 10 m 1 m perpendicular to 60 m transect analyzed	n.r.	n.r.	range: 0 to 34 pieces/ m^2
Hoellein et al. (2014)	US	Lake Michigan	n.r.	3	400 m transects, within 50 m from water line	n.r.	>1 mm	mean: ca. 0.01 pieces/ m^2 (<1 g/m^2) (Fig. 1)
Zbyszewski et al. (2014)	US	Lake Erie	n.r.	10	every 10 m 1 m perpendicular to 60 m transect analyzed	sand, gravel, mud	n.r.	range: 0.56 to 3.70 pieces/m^2
	US	Lake St. Clair	n.r./3.4 m/n.r.	9	every 10 m 1 m perpendicular to 60 m transect analyzed	sand, gravel, mud	n.r.	range: 0.18 to 8.38 pieces/m^2
Hoellein et al. (2015)	US	Lake Michigan	58,000 km^2/85 m/ n.r.	5	citizen science; complete or parts of beaches	sand	>5 mm (ca.)	mean: 0.0092 items/m^2
Chapman (2019)	US	Lake Lewisville	n.r.	1	random grid sampling: 16 quadrats of 5 × 5 m	n.r.	>5 mm	0.213 items/m^2 (0.112 g/m^2)

and saving time and compared to manual observations of beach litter (Ge et al., 2016).

So far, UAV images in this study were only analyzed for the detection of prepared plastic particles. The detection of actual AL has not been tested, yet. However, it was already shown that using RGB-composites with a 4K resolution may lead to an accurate classification of prepared plastic particles. UAVs, including these camera characteristics, are widely available and easily deployable so that they might provide a cost- and time-efficient alternative compared to the manual beach litter monitoring. This was also confirmed by a study by Martin et al. (2018) using a UAV for analyzing AL at beaches. The authors showed that automatic detection by image analysis was 39 times faster than manual litter recording and that particles >4 cm were reliably tracked.

4.3. Anthropogenic litter monitoring at lake beaches

For the present study, the list for marine litter monitoring provided by the MSFD (2013) was used. Using a list developed for marine environments worked well, even though some categories are not at all present at the beaches investigated. On one hand, this may be attributed to missing sources of these items at the selected study area, on the other hand, specific activities leading to littering are not available in freshwater environments compared to marine environments. Therefore, the list was slightly adapted for lake beach litter monitoring using the results of the study at Lake Tollense as well as incorporating other lake studies.

Item categories not identified at beaches were removed from the list, while in other cases more detailed descriptions were necessary for specific items. The complete adapted list can be found in the Supporting Material (SM3). While, e.g. octopus and lobster fishing items were

removed from the list, as they are not typical for freshwater lakes, other lake studies showed, that general fishing related items may be included in AL on lake beaches (Egessa et al., 2020; Free et al., 2014; Ngupula et al., 2014). The same is true for shipping related items. Plastic manufacturing industries are situated around the Great Lakes (e.g. Corcoran et al., 2015; Hoellein et al., 2015) and pollution by pellets is high. Although these items are in most cases <5 mm they were kept within the list but were classified as microlitter items.

For Lake Tollense some items could not be attributed to a class due to no matching category. Therefore, the most frequently added annotations were converted into new categories in the list: paint flakes (material group: artificial polymers), tissues (paper/cardboard), fireworks (artificial polymers and metal), sticking plaster (artificial polymers), bread bag fastener (artificial polymers), bottle banderole (paper/cardboard), plastic foil (artificial polymers).

Furthermore, a consistent sampling method is needed for AL studies at lake beaches. Therefore, based on the OSPAR beach monitoring protocol (OSPAR, 2010) and the sampling method used at Lake Tollense the following suggestions are made for future sampling of lake beaches: Marine beaches often stretch over long distances and segments of 100 m are easily available (as recommended by OSPAR, 2010). Freshwater environment beaches, especially beaches with sandy substrate, are often smaller so that this prerequisite has to be dropped. However, whenever possible, a stretch of at least 100 m should be analyzed. Furthermore, it is important to define the area investigated. Measurements of beach length and beach width (every 10 m with subsequent averaging according to Hoellein et al. (2015)) should be performed to estimate the analyzed beach area. An alternative method to gain beach area is the analysis of UAV aerial images via GIS as done in this study. As suggested by OSPAR (2010), beaches should be free from buildings and accessible all year round. If possible, the whole beach area or length of beach should be investigated for AL, instead of concentrating the study on the strandline or other specific parts of the beach, to enable comparisons between studies and avoid over- or underestimation of AL.

During sampling at Lake Tollense, the movement of surveyors back and forth from the waterline, in small transects, was found to be an effective method. This walking strategy was also proposed by Owens and Kamil (2020) for riverbank litter monitoring. All visible AL should be recorded using the tally list, removed from the beach, and subsequently weighed. Items may later be categorized into broader groups, according to e.g. material. Additionally, information on influences on AL abundances should be recorded during sampling. These include obstacles within the sampled beach area, a description of the development of the back of the beach, weather conditions, information on regular beach cleaning programs and the number of surveyors.

5. Conclusion

Anthropogenic litter is ubiquitous at beaches of Lake Tollense in MV, Germany. Abundances depend not only on anthropogenic activity in vicinity of the beaches but are also influenced by inputs via tributaries and wind conditions. The high percentage of plastic within AL underlines the progressing pollution of the environment by this material. The threat plastic litter poses to aquatic organisms in lake environments is not yet fully understood but the results of this study show that impacts of plastic litter in beach lake environments need consideration. Similarly, regular monitoring of lake beaches should be established to further observe seasonal variations in AL abundances and possible impact factors. A monitoring program at beaches can be performed by manual litter monitoring or may be improved using aerial images taken by UAV with appropriate cameras which are widely available.

Authors contribution

Elena Hengstmann: Conceptualization, Investigation, Formal analysis, Writing - Original Draft, Visualization. Elke Fischer: Conceptualization, Resources, Writing - Review & Editing, Supervision.

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Declaration of competing interest

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

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Appendix A. Supplementary data

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Supplementary Material

Anthropogenic litter in freshwater environments – Study on lake beaches evaluating marine guidelines and aerial imaging

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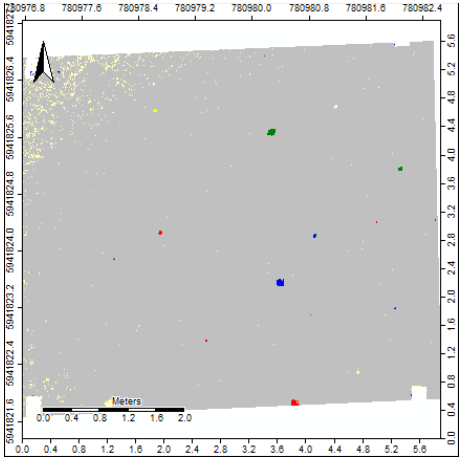
SM1: Anthropogenic litter abundances at the four investigated beaches at Lake Tollense for all sampling dates presented in different units.

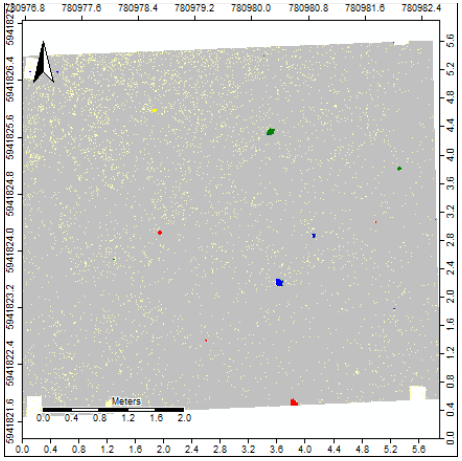
Unit	Time	North	East	South	West
items/100 m	**September 2017**	266.4	87.6	102.8	118.7
	March 2018	75.0	54.0	30.0	42.6
	September 2018	227.5	161.3	143.2	96.2
	March 2019	126.4	200.8	54.8	113.7
	September 2019	441.8	193.6	106.8	171.2
	March 2020	141.3	81.0	79.1	26.1
g/100 m	**September 2017**	398.7	16.4	63.0	570.2
	March 2018	73.0	29.3	17.1	137.0
	September 2018	155.8	39.1	512.7	566.9
	March 2019	124.3	257.1	1291.0	157.1
	September 2019	139.0	59.3	127.8	91.9
	March 2020	225.6	83.0	94.3	18.9
items/m²	**September 2017**	0.3	0.1	0.4	0.2
	March 2018	0.1	0.1	0.2	0.1
	September 2018	0.2	0.1	0.5	0.2
	March 2019	0.1	0.3	0.2	0.2
	September 2019	0.5	0.2	0.5	0.3
	March 2020	0.2	0.2	0.6	0.1
g/m²	**September 2017**	0.5	0.0	0.2	0.9
	March 2018	0.1	0.1	0.1	0.4
	September 2018	0.2	0.0	1.8	1.1
	March 2019	0.1	0.3	5.0	0.3
	September 2019	0.2	0.1	0.6	0.2
	March 2020	0.3	0.2	0.7	0.1

SM2: Results of image analysis considering UAV images from different heights and using supervised classification with training areas according to colored plastic shreds distributed on a test area at North beach in March 2019.

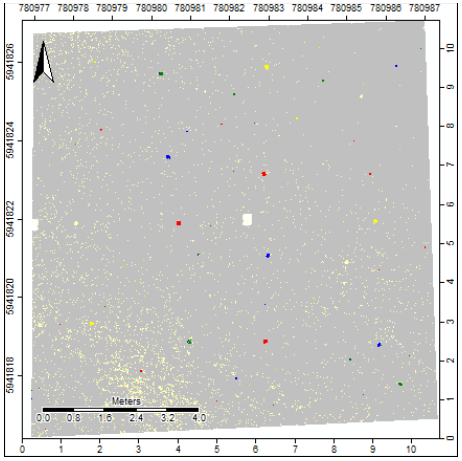
area 5x5 m – height 7 m



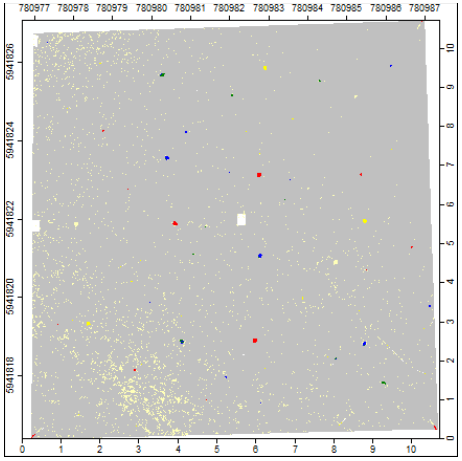
area 5x5 m – height 10 m



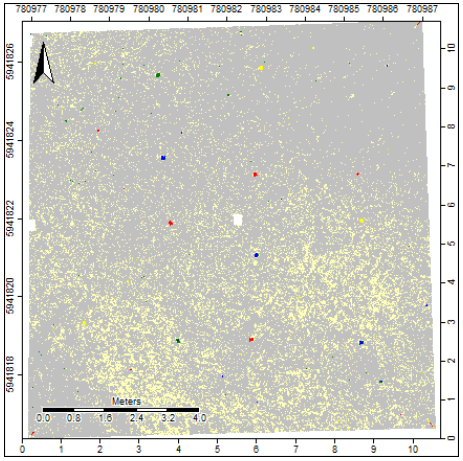
area 10x10 m – height 15 m



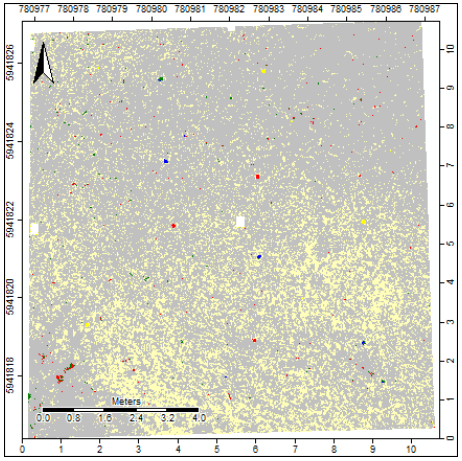
area 10x10 m – height 20 m



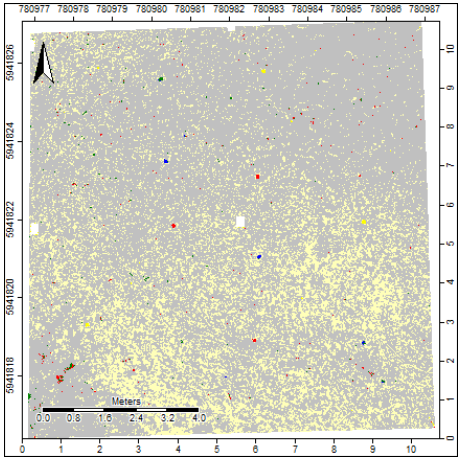
area 10x10 m – height 30 m



area 10x10 m – height 50 m



area 10x10 m – height 80 m



SM3: Anthropogenic litter list adapted from MSFD (2013) with adjustments for lake shore monitoring. Categories added/changed in green. categories deleted in red

new Lake Code	Code MSFD (2013)	Code OSPAR (2011)	Category	Material
L1	G1	1	4/6-pack yokes. six-pack rings	Artificial polymer materials
L2	G2		Bags	Artificial polymer materials
L3	G3	2	Shopping Bags incl. pieces	Artificial polymer materials
L4	G4	3	Small plastic bags. e.g. freezer bags incl. pieces	Artificial polymer materials
L5	G5	112	Plastic bag collective role; what remains from rip-off plastic bags	Artificial polymer materials
L6	G6	4	Bottles	Artificial polymer materials
L7	G7	4	Drink bottles <= 0.5 l	Artificial polymer materials
L8	G8	4	Drink bottles > 0.5 l	Artificial polymer materials
L9	G9	5	Cleaner bottles & containers	Artificial polymer materials
L10	G10	6	Food containers incl. fast food containers	Artificial polymer materials
L11	G11	7	Beach use related cosmetic bottles and containers. e.g. Sunblocks	Artificial polymer materials
L12	G12	7	Other cosmetics bottles & containers	Artificial polymer materials
L13	G13	12	Other bottles & containers (drums)	Artificial polymer materials
L14	G14	8	Engine oil bottles & containers < 50 cm	Artificial polymer materials
L15	G15	9	Engine oil bottles & containers > 50 cm	Artificial polymer materials
L16	G16	10	Jerry cans (square plastic containers with handle)	Artificial polymer materials
L17	G17	11	Injection gun containers	Artificial polymer materials
L18	G18	13	Crates and containers / baskets	Artificial polymer materials
L19	G19	14	Car parts	Artificial polymer materials
L20	G20		Plastic caps and lids	Artificial polymer materials
L21	G21	15	Plastic caps/lids drinks	Artificial polymer materials
L22	G22	15	Plastic caps/lids chemicals. detergents (non-food)	Artificial polymer materials
L23	G23	15	Plastic caps/lids unidentified	Artificial polymer materials
L24	G24	15	Plastic rings from bottle caps/lids	Artificial polymer materials
L25	G25		Tobacco pouches/plastic cigarette box packaging	Artificial polymer materials
L26	G26	16	Cigarette lighters	Artificial polymer materials
L27	G27	64	Cigarette butts and filters	Artificial polymer materials
L28	G28	17	Pens and pen lids	Artificial polymer materials
L29	G29	18	Combs/hair brushes/sunglasses	Artificial polymer materials
L30	G30	19	Crisp packets/sweet wrappers	Artificial polymer materials
L31			Plastic foil for wrapping	Artificial polymer materials
L32	G31	19	Lolly sticks	Artificial polymer materials
L33	G32	20	Toys and party poppers	Artificial polymer materials
L34			Fireworks	Artificial polymer materials
L35	G33	21	Cups and cup lids	Artificial polymer materials
L36	G34	22	Cutlery and trays	Artificial polymer materials
L37	G35	22	Straws and stirrers	Artificial polymer materials
L38			Bread bag fastener	Artificial polymer materials
L39	G36	23	Fertiliser/animal feed bags	Artificial polymer materials
L40	G37	24	Mesh vegetable bags	Artificial polymer materials
L41	G38		Cover/packaging	Artificial polymer materials
L42	G39		Gloves	Artificial polymer materials
L43	G40	25	Gloves (washing up)	Artificial polymer materials
L44	G41	113	Gloves (industrial/professional rubber gloves)	Artificial polymer materials
L45			Sticking plaster	Artificial polymer materials
	~~G42~~	~~26~~	~~Crab/lobster pots and tops~~	~~Artificial polymer materials~~
L46	G43	114	Tags	Artificial polymer materials
	~~G44~~	~~27~~	~~Octopus pots~~	~~Artificial polymer materials~~
L47	G45	28	Mussels nets	Artificial polymer materials
	~~G46~~	~~29~~	~~Oyster trays (round from oyster cultures)~~	~~Artificial polymer materials~~
L48	G47	30	Plastic sheeting from mussel culture (Tahitians)	Artificial polymer materials
L49	G48		Synthetic rope	Artificial polymer materials
L50	G49	31	Rope (diameter more than 1 cm)	Artificial polymer materials

L51	G50	32	String and cord (diameter less than 1 cm)	Artificial polymer materials
L52	G51		Fishing net	Artificial polymer materials
L53	G52		Nets and pieces of net	Artificial polymer materials
L54	G53	115	Nets and pieces of net < 50 cm	Artificial polymer materials
L55	G54	116	Nets and pieces of net > 50 cm	Artificial polymer materials
L56	G55		Fishing line (entangled)	Artificial polymer materials
L57	G56	33	Tangled nets/cord	Artificial polymer materials
L58	G57	34	Fish boxes - plastic	Artificial polymer materials
L59	G58	34	Fish boxes - expanded polystyrene	Artificial polymer materials
L60	G59	35	Fishing line/monofilament (angling)	Artificial polymer materials
L61	G60	36	Light sticks (tubes with fluid) incl. Packaging	Artificial polymer materials
L62	G61		Other fishing related	Artificial polymer materials
L63	G62	37	Floats for fishing nets	Artificial polymer materials
L64	G63	37	Buoys	Artificial polymer materials
L65	G64		Fenders	Artificial polymer materials
L66	G65	38	Buckets	Artificial polymer materials
L67	G66	39	Strapping bands	Artificial polymer materials
L68	G67	40	Sheets. Industrial packaging. plastic sheeting	Artificial polymer materials
L69	G68	41	Fibre glass/fragments	Artificial polymer materials
L70	G69	42	Hard hats/Helmets	Artificial polymer materials
L71	G70	43	Shotgun cartridges	Artificial polymer materials
L72	G71	44	Shoes/sandals	Artificial polymer materials
L73	G72		Traffic cones	Artificial polymer materials
L74	G73	45	Foam sponge	Artificial polymer materials
L75	G74		Foam packaging/insulation/polyurethane	Artificial polymer materials
L76	G75	117	Plastic/polystyrene pieces 0 - 2.5 cm	Artificial polymer materials
L77	G76	46	Plastic/polystyrene pieces 2.5 cm > < 50 cm	Artificial polymer materials
L78	G77	47	Plastic/polystyrene pieces > 50 cm	Artificial polymer materials
L79	G78		Plastic pieces 0 - 2.5 cm	Artificial polymer materials
L80	G79		Plastic pieces 2.5 cm > < 50 cm	Artificial polymer materials
L81	G80		Plastic pieces > 50 cm	Artificial polymer materials
L82	G81		Polystyrene pieces 0 - 2.5 cm	Artificial polymer materials
L83	G82		Polystyrene pieces 2.5 cm > < 50 cm	Artificial polymer materials
L84	G83		Polystyrene pieces > 50 cm	Artificial polymer materials
L85	G84		CD. CD-box	Artificial polymer materials
L86	G85		Salt packaging	Artificial polymer materials
L87	G86		Fin trees (from fins scuba diving)	Artificial polymer materials
L88	G87		Masking tape	Artificial polymer materials
L89	G88		Telephone (incl. parts)	Artificial polymer materials
L90	G89		Plastic construction waste	Artificial polymer materials
L91	G90		Plastic flower pots	Artificial polymer materials
L92	G91		Biomass holder from sewage treatment plants	Artificial polymer materials
L93	G92		Bait containers/packaging	Artificial polymer materials
L94	G93		Cable ties	Artificial polymer materials
L95	G94		Table cloth	Artificial polymer materials
L96	G95	98	Cotton bud sticks	Artificial polymer materials
L97	G96	99	Sanitary towels/panty liners/backing strips	Artificial polymer materials
L98	G97	101	Toilet fresheners	Artificial polymer materials
L99	G98		Diapers/nappies	Artificial polymer materials
L100	G99	104	Syringes/needles	Artificial polymer materials
L101	G100	103	Containers / tubes	Artificial polymer materials
L102	G101	121	Dog faeces bag	Artificial polymer materials
L103	G102		Flip-flops	Artificial polymer materials
L104	G103		Micro: Plastic fragments rounded < 5 mm	Artificial polymer materials
L105	G104		Micro: Plastic fragments subrounded < 5 mm	Artificial polymer materials
L106	G105		Micro: Plastic fragments subangular < 5 mm	Artificial polymer materials
L107	G106		Micro: Plastic fragments angular < 5 mm	Artificial polymer materials
L108	G107		Micro: cylindrical pellets < 5 mm	Artificial polymer materials

L109	G108		Micro: disks pellets < 5 mm	Artificial polymer materials
L110	G109		Micro: flat pellets < 5 mm	Artificial polymer materials
L111	G110		Micro: ovoid pellets < 5 mm	Artificial polymer materials
L112	G111		Micro: spheruloids pellets < 5 mm	Artificial polymer materials
L113	G112		Industrial pellets	Artificial polymer materials
L114	G113		Micro: Filament < 5 mm	Artificial polymer materials
L115	G114		Micro: Films < 5 mm	Artificial polymer materials
L116	G115		Micro: Foamed plastic < 5 mm	Artificial polymer materials
L117	G116		Micro: Granules < 5 mm	Artificial polymer materials
L118	G117		Styrofoam pieces	Artificial polymer materials
L119	G118		Micro: Small industrial spheres (< 5 mm)	Artificial polymer materials
L120	G119		Sheet like user plastic (> 1 mm)	Artificial polymer materials
L121	G120		Threads	Artificial polymer materials
L122	G121		Foamed user plastic (> 1 mm)	Artificial polymer materials
L123	G122		Plastic fragments (> 1 mm)	Artificial polymer materials
L124	G123		Micro: Polyurethane granules < 5 mm	Artificial polymer materials
L125			Paint flakes	Artificial polymer materials
L126	G124	48	Other plastic/polystyrene items (identifiable)	Artificial polymer materials
L127	G125	49	Balloons and balloon sticks	Rubber
L128	G126		Balls	Rubber
L129	G127	50	Rubber boots	Rubber
L130	G128	52	Tires and belts	Rubber
L131	G129		Inner-tubes and rubber sheet	Rubber
L132	G130		Wheels	Rubber
L133	G131		Rubber bands (small. for kitchen/household/post use)	Rubber
L134	G132		Bobbins (fishing)	Rubber
L135	G133	97	Condoms (incl. packaging)	Rubber
L136	G134	53	Other rubber pieces	Rubber
L137	G135		Clothing (clothes. shoes)	Cloth/textile
L138	G136		Shoes	Cloth/textile
L139	G137	54	Clothing/rags (clothing. hats. towels)	Cloth/textile
L140	G138	57	Shoes and sandals (e.g. leather. cloth)	Cloth/textile
L141	G139		Backpacks & bags	Cloth/textile
L142	G140	56	Sacking (hessian)	Cloth/textile
L143	G141	55	Carpet & Furnishing	Cloth/textile
L144	G142		Rope. string and nets	Cloth/textile
L145	G143		Sails. canvas	Cloth/textile
L146	G144	100	Tampons and tampon applicators	Cloth/textile
L147	G145	59	Other textiles (incl. rags)	Cloth/textile
L148	G146		Paper/Cardboard	Paper/Cardboard
L149	G147	60	Paper bags	Paper/Cardboard
L150	G148	61	Cardboard (boxes & fragments)	Paper/Cardboard
L151	G149		Paper packaging	Paper/Cardboard
L152	G150	118	Cartons/Tetrapak Milk	Paper/Cardboard
L153	G151	62	Cartons/Tetrapak (others)	Paper/Cardboard
L154			Bottle banderole	Paper/Cardboard
L155	G152	63	Cigarette packets	Paper/Cardboard
L156	G153	65	Cups. food trays. food wrappers. drink containers	Paper/Cardboard
L157	G154	66	Newspapers & magazines	Paper/Cardboard
L158	G155		(Tubes for) fireworks	Paper/Cardboard
L159			Tissues	Paper/Cardboard
L160	G156		Paper fragments	Paper/Cardboard
L161	G157		Paper	Paper/Cardboard
L162	G158	67	Other paper items	Paper/Cardboard
L163	G159	68	Corks	Processed/worked wood
L164	G160	69	Pallets	Processed/worked wood
L165	G161	69	Processed timber	Processed/worked wood
L166	G162	70	Crates	Processed/worked wood

	~~G163~~	~~71~~	~~Crab/lobster pots~~	~~Processed/worked wood~~
L167	G164	119	Fish boxes	Processed/worked wood
L168	G165	72	Ice-cream sticks. chip forks. chopsticks. toothpicks	Processed/worked wood
L169	G166	73	Paint brushes	Processed/worked wood
L170	G167		Matches & fireworks	Processed/worked wood
L171	G168		Wood boards	Processed/worked wood
L172	G169		Beams/Dunnage	Processed/worked wood
L173	G170		Wood (processed)	Processed/worked wood
L174	G171	74	Other wood < 50 cm	Processed/worked wood
L175	G172	75	Other wood > 50 cm	Processed/worked wood
L176	G173		Other (specify)	Processed/worked wood
L177	G174	76	Aerosol/Spray cans industry	Metal
L178	G175	78	Cans (beverage)	Metal
L179	G176	82	Cans (food)	Metal
L180	G177	81	Foil wrappers. aluminum foil	Metal
L181	G178	77	Bottle caps. lids & pull tabs	Metal
L182	G179	120	Disposable BBQ's	Metal
L183	G180	79	Appliances (refrigerators. washers etc.)	Metal
L184	G181		Tableware (plates. cups & cutlery)	Metal
L185			Fireworks	Metal
L186	G182	80	Fishing related (weights. sinkers. lures. hooks)	Metal
L187	G183		Fish hook remains	Metal
	~~G184~~	~~87~~	~~Lobster/crab pots~~	~~Metal~~
L188	G185		Middle size containers	Metal
L189	G186	83	Industrial scrap	Metal
L190	G187	84	Drums. e.g. oil	Metal
L191	G188		Other cans (< 4 L)	Metal
L192	G189		Gas bottles. drums & buckets (> 4 L)	Metal
L193	G190	86	Paint tins	Metal
L194	G191	88	Wire. wire mesh. barbed wire	Metal
L195	G192		Barrels	Metal
L196	G193		Car parts/batteries	Metal
L197	G194		Cables	Metal
L198	G195		Household Batteries	Metal
L199	G196		Large metallic object	Metal
L200	G197		Other (metal)	Metal
L201	G198	89	Other metal pieces < 50 cm	Metal
L202	G199	90	Other metal pieces > 50 cm	Metal
L203	G200	91	Bottles incl. pieces	Glass/ceramics
L204	G201		Jars incl. pieces	Glass/ceramics
L205	G202	92	Light bulbs	Glass/ceramics
L206	G203		Tableware (plates & cups)	Glass/ceramics
L207	G204	94	Construction material (brick. cement. pipes)	Glass/ceramics
L208	G205	92	Fluorescent light tubes	Glass/ceramics
L209	G206		Glass buoys	Glass/ceramics
	~~G207~~	~~95~~	~~Octopus pots~~	~~Glass/ceramics~~
L210	G208		Glass or ceramic fragments > 2.5 cm	Glass/ceramics
L211	G209		Large glass objects (specify)	Glass/ceramics
L212	G210	96	Other glass items	Glass/ceramics
L213	G211	105	Other medical items (swabs. bandaging. adhesive plaster etc.)	unidentified
L214	G212		Slack/Coal	
L215	G213	108	Paraffin/Wax 0-1 cm	Chemicals
L216	G213	109	Paraffin/Wax 1-10 cm	Chemicals
L217	G213	110	Paraffin/Wax > 10 cm	Chemicals
L218	G214		Oil/Tar	Chemicals
L219	G215		Food waste (galley waste)	Food waste
L220	G216		various rubbish (worked wood. metal parts)	undefined
L221	G217		other (glass. metal. tar) < 5 mm	unidentified

Publication III

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Microplastics in lakeshore and lakebed sediments – External influences and temporal and spatial variabilities of concentrations

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ABSTRACT

Microplastics have been predominantly studied in marine environments compared to freshwater systems. However, the number of studies analyzing microplastic concentrations in water and sediment within lakes and rivers are increasing and are of utmost importance as freshwaters are major pathways for plastics to the oceans. To allow for an adequate risk assessment, detailed knowledge concerning plastic concentrations in different environmental compartments of freshwaters are necessary. Therefore, the major aim of this study was the quantification and analysis of temporal and spatial distribution of microplastics (<5 mm) in freshwater shore and bed sediments at Lake Tollense, Mecklenburg-Western Pomerania, Germany. Likewise, it addresses the hypothesis that lakes may serve as long-term storage basins for microplastics. Concentrations were investigated semi-annually over a two-year period at four sandy bank border segments representing different expositions and levels of anthropogenic influence. In addition, lakebed samples were taken along the longitudinal dimension of Lake Tollense. Mean microplastic abundances were 1,410 ± 822 particles/kg DW for lakeshore sediments and 10,476 ± 4,290 particles/kg DW for lakebed sediments. Fragments were more abundant compared to fibers in both sediment compartments. Spatial and temporal variation was especially recognized for lakeshore sediments whereas microplastic abundances in lakebed sediments did not differ significantly between sampling points and sampling campaigns. This can be related to long-term accumulation at the lakebed. Lower microplastic abundances were found within the intertidal zone at lake beaches where constant wave action reduces accumulation. Increased microplastic abundances were recognized at the beach with least anthropogenic influence but in proximity to a tributary, which may serve as microplastic input pathway into Lake Tollense due to its catchment comprising mainly agricultural areas. Furthermore, spatial variations in microplastic concentrations were related to the abundance of macroplastic items at beaches and correlated with pedologic sediment characteristics, namely the content of organic matter.

1. Introduction

As the plastic production rose continuously in the last decade, from 250 million tons in 2009 to 368 million tons in 2019 (PlasticsEurope 2015, 2020), the environmental pollution is likewise gradually increasing. The intensified pollution is caused by the longevity and low decomposition rates of plastic items and particles in the environment (Andrady et al., 2015) as well as by ongoing accidental and intentional plastic input into the environment (Barnes et al., 2009). First presence of plastics in the environment were proven in the early 1970s (Carpenter et al., 1972; Carpenter and Smith 1972). The number of studies investigating plastic contamination in different environmental compartments strongly increased since the early 2000s century (Ivleva et al., 2017). Plastics of all sizes were found in different compartments around the world, ranging from water and ice to sediments to biota as well as atmosphere (Bellasi et al., 2020; Bianco and Passananti 2020; Rochman 2018; Ivleva et al., 2017; Dris et al., 2015; Ivar do Sul and Costa 2014; Wagner et al., 2014).

Plastics are classified according to their size, namely macro-, meso-, micro-, and nanoplastics. However, definitions vary for the individual

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terms (Hartmann et al., 2019). Commonly, microplastics are referred to as particles <5 mm (Arthur et al., 2009). Attention on plastics shifted from large to small ones with time and even nanoplastics (<1 μm; GESAMP 2015) are increasingly studied today (Lehner et al., 2019; Mattsson et al., 2018; Koelmans et al., 2015). Similarly, the number of studies investigating freshwater bodies rose lately (Lambert and Wagner 2018; Rochman 2018; Horton et al., 2017a; Dris et al., 2015; Wagner et al., 2014). However, Blettler et al. (2018) stated that 87% of plastic pollution studies dealt with marine environments compared to 13% of studies analyzing freshwater systems. Marine environments are supposed to be the final sink for plastics (Rochman 2018; Bergmann et al., 2017; Wagner et al., 2014; Woodall et al., 2014; Thompson et al., 2004) whereas terrestrial freshwaters are considered as transport pathways for plastics from land to the ocean (Lebreton et al., 2017; Eerkes-Medrano et al., 2015; Mani et al., 2016; Gasperi et al., 2014; Wagner et al., 2014). Studies investigating the impacts of plastic on organisms were also mainly focusing on marine environments whereas freshwater studies were underrepresented (Blettler and Wantzen 2019). Possible impacts of plastic on marine and freshwater organisms comprise the entanglement within large plastic items, the ingestion of small particles and consequently the uptake of possibly toxic, chemical compounds, added or adsorbed to microplastics (e.g., GESAMP 2015; GEF 2012; Cole et al.,

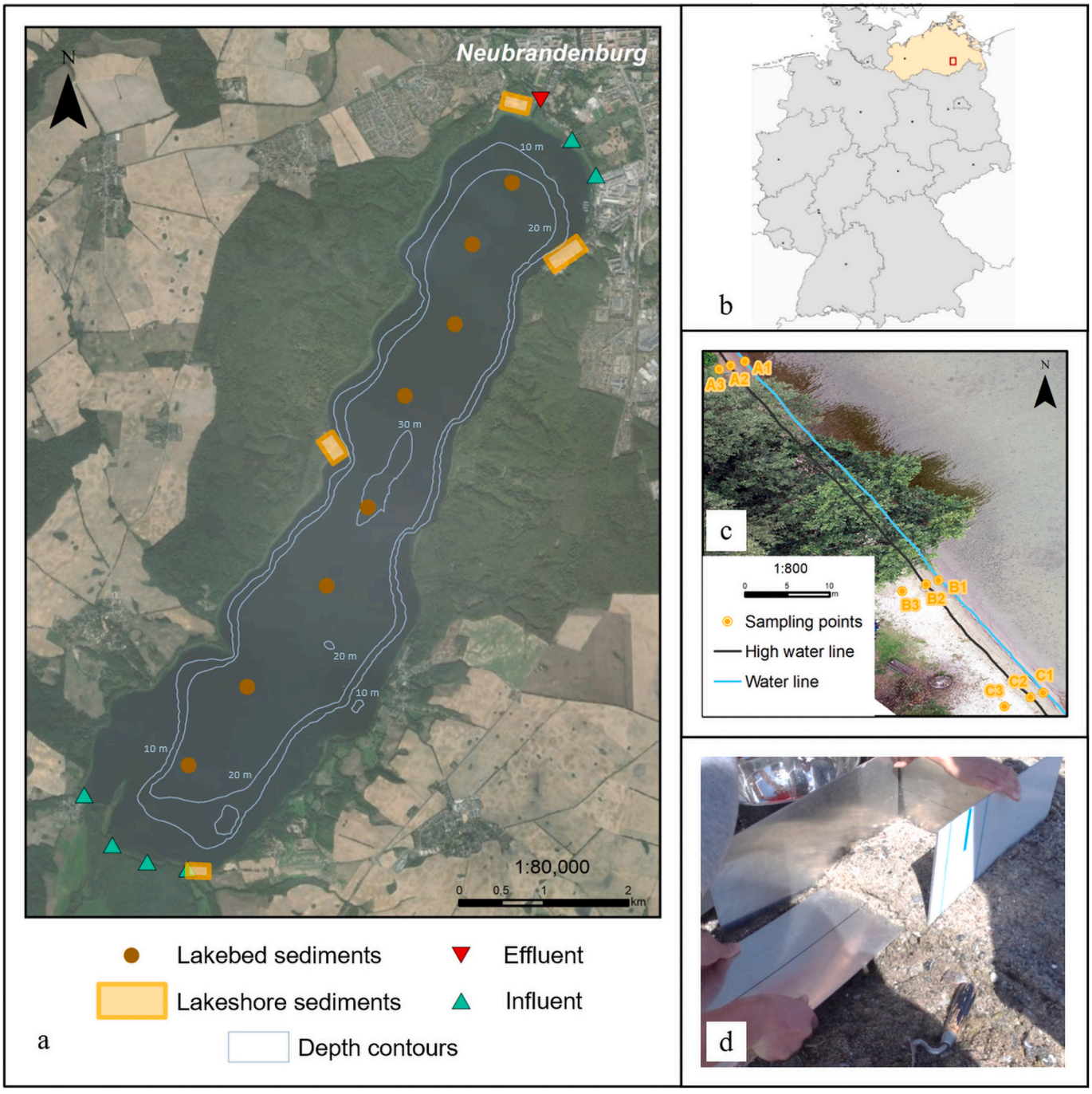

Fig. 1. Overview of the study area and sampling approaches. a) Map showing investigated lakeshores, lakebed sampling points and locations of tributaries at Lake Tollense, (projection: Transverse Mercator; Coordinate system: WGS_1984_UTM_Zone_32N; Satellite image: ArcGIS Imagery ©ESRI; Bathymetry: MLUMV, 2017). b) Location of study area in Germany. c) Sampling points for lakeshore sediments at beaches in three transects and in relation to the water and high water line. d) Sediment sampling at lakeshores in quadrates of 25 × 25 cm.

2011; Teuten et al., 2009; UNEP 2005; Thompson et al., 2004; Laist 1997; Laist 1987). For an adequate risk assessment of plastics within freshwater environments, detailed information on impacts on organisms is necessary as well as knowledge concerning plastic concentrations in different environmental compartments.

Microplastic pollution in freshwater bodies was demonstrated at the water surface (e.g., Lenaker et al., 2019; LfU, 2019; Mani et al., 2016; Eriksen et al., 2013), within the water column (e.g., Lenaker et al., 2021; Tamminga and Fischer 2020; Eo et al., 2019; Dris et al., 2018), in shoreline (e.g., LfU, 2019; Fischer et al., 2016; Faure et al., 2015; Imhof et al., 2013) and bed sediments (e.g., Wilkens et al., 2020; Zobkov et al., 2020; Turner et al., 2019; Castañeda et al., 2014) in different regions of the world. Results show high variability of microplastic concentrations. Unfortunately, the comparison of results is hampered due to differences in applied methods during sampling and laboratory processing (Horton et al., 2017a; Ivleva et al., 2017; Dris et al., 2015). Still, data provided by studies investigating microplastic abundances in freshwaters is highly needed. Especially, as freshwater sediments are expected to be temporary or long-term sinks for microplastics (Li et al., 2020; Bordós et al., 2019; Turner et al., 2019; Vaughan et al., 2017; Imhof et al., 2013).

In this study, microplastic concentrations in sediments of Lake Tollense, Mecklenburg-Western Pomerania, Germany, were analyzed over a two-year period by semi-annual sampling. Lakeshore sediments from four beach segments around the lake were taken as well as lakebed sediments at several points in the center of the lake. The major aim of the study was the quantification of microplastics in freshwater sediments using the example of Lake Tollense and addressing the hypothesis that lake sediments serve as a sink for microplastics. Furthermore, spatial and temporal distribution of microplastics were analyzed as well as the factors influencing the microplastic pollution within lake sediments, including pedologic characteristics such as percentage of organic matter and grain sizes.

2. Materials and methods

2.1. Study area

The study was conducted at Lake Tollense in Mecklenburg-Western Pomerania (MV) in the northeast of Germany (Fig. 1b). Lake Tollense covers an area of 17.9 km^2, has a mean depth of 17.3 m (max. depth 31.2 m) and a catchment area of 515 km^2 which is predominated by agricultural and forest areas (Nixdorf et al., 2004). Several tributaries from sub-catchments with different landcover characteristics contribute to the water and material budget of Lake Tollense. One of the major inflows is the Nonnenbach (0.57 m^3/s) which enters Lake Tollense in the south. The Nonnenbach catchment area covers 219 km^2 (MLUMV, 2017) and human activity in the form of agricultural use is highly present here. The only effluent, the Tollense, leaves the lake in the north (Nixdorf et al., 2004). The city of Neubrandenburg (63,400 inhabitants in 2020, LAiV, 2020) is situated at the lake's northern shore leading to a higher population density and anthropogenic use (e.g., swimming, boating, fishing) for this part of the lake. In contrast, a nature reserve is located at the south-western shore of the lake resulting in a reduced anthropogenic frequentation.

Four sandy bank border segments were selected for sampling lakeshore sediments (Fig. 1a). These differ in their exposition and in the degree of anthropogenic influence. The beach at the northern bank (North beach) as well as the one in the east (East beach) are characterized by proximity to the city of Neubrandenburg and show a high degree of anthropogenic activity. In contrast, the beach at the southern shore (South beach) is located furthest from the city and is within the nature reserve. The bank border segment in the west (West beach) is located on a small headland and within a campground area. A more detailed description of the study area and the sampled beaches can be found in Hengstmann and Fischer (2020).

2.2. Sampling

Sampling was conducted semi-annually in March and September. Four sampling campaigns are included in this study (September 2017, March 2018, September 2018, March 2019). Sampling months were chosen on account of the tourist season at Lake Tollense lasting from May to September and were based on the change in water level (highest in February and lowest in August; Pegelportal MV 2020).

Lakeshore sediment samples (n = 185) were taken at the four beach segments. At least three parallel transects (perpendicular to the water line) were established at each beach. Within these transects, samples were taken in three different distances from the waterline according to Fischer et al. (2016): (i) within the intertidal zone, (ii) at the high water line, (iii) within the elevated area of the beach (Fig. 1c). For beach regeneration, non-natural sand is deposited in the back of North and East beach. This sediment was not sampled but the third position was located closer to the first two sampling points. Numbers of parallels and positions, and consequently number of samples, differed for the first two sampling campaigns since more than three transects were sampled at some beaches and distances to the water line were skipped due to high water level in March 2018 (Sep. '17: n = 62, Mar. '18: n = 51). Based on results of the first sampling campaigns and to overcome analytical challenges, sample numbers were reduced to 36 per sampling (three transects with three sampling points) for sampling campaigns after March 2018.

Lakeshore sediment samples were taken using a metal quadrate of 25 × 25 cm (625 cm^2) and scraping off the first 1–2 cm of sediment with a metal sheet (Fig. 1d). Sediment was transferred into a large stainless-steel bowl for homogenization before filling it into 2 L glass jars for transportation to the laboratory. Furthermore, lakebed sediments were collected during each sampling campaign (n = 32). Eight transects spanning the width of the lake were determined covering the complete length of the lake in nearly equal distances. At the center of these transects, lakebed sediments were taken from a vessel using a van-Veen grab (©KC Denmark, sample area 250 cm^2, sample volume 3.14 L). The material was transferred into a stainless-steel bowl for homogenization. A subsample of 2 L was then filled into a glass jar for transportation to the laboratory.

In addition to lakeshore and lakebed sediment sampling, meteorological and hydrological data was recorded, and aerial images of the beach segments were taken via an unmanned aerial vehicle (DJI Phantom II Vision+ and DJI Phantom IV Pro). Wind data was also obtained from the nearest meteorological station (Trollenhagen, linear distance ca. 12 km) provided by Deutscher Wetterdienst (DWD, 2020).

2.3. Laboratory analysis

Samples were analyzed in a multi-step process in the laboratory. However, protocols marginally differed for lakeshore and lakebed sediments. For lakeshore sediments, samples were dried at 40 °C until weight constancy and 50 ml of dry sediment were used for the microplastic analysis. For lakebed sediments, 100 ml wet sediment were analyzed. In parallel, 100 ml of wet sediment were dried at 40 °C to receive corresponding dry weight. Samples were digested using in a first step hydrogen peroxide (H_2O_2; lakeshore: 30%, 60 ml; lakebed: 10%, 80 ml; for 7 days) and in a second step sodium hypochlorite (NaClO; 6–14% in a 3:1 volume ratio, for 24 h) to eliminate biogenic organic material (Hengstmann et al., 2018; Tamminga et al., 2018). Hydrochloric acid (HCl, 10%, for 2–4 h) was added to eliminate calcareous components and its application was tested prior to implementation (Pfeiffer and Fischer 2020). Between different digestion steps, samples were washed in a 63 μm sieve using deionized water. Subsequently, samples were wet sieved over a sieve cascade comprising mesh sizes of 1,000 μm, 630 μm, 300 μm, 200 μm, and 63 μm. In case of lakeshore sediments, sieved subsamples were density separated deploying an elutriation column, as described in Hengstmann et al. (2018), and were afterwards filtered

onto paper filters (VWR, qualitative filter paper 413, 5–13 μm particle retention). Filters were placed into covered petri dishes for drying. Due to small amounts of remaining material in the specific sieves for lakebed sediments, elutriation was skipped for these samples to avoid unnecessary additional processing steps. Samples were directly filtered instead.

Pedologic analyses were performed on all sediment samples. This included the determination of bulk density (BD) for lakeshore sediments (DIN EN ISO 11272:2017-07, 2017), water content at 105 °C (WC; DIN ISO 11465:1996-12, 1996) and organic matter as loss on ignition (LOI; DIN EN 12879:2001-02, 2001 as well as a grain size analysis (DIN ISO 11277:2002-08, 2002). BD was used to extrapolate microplastic concentrations to 1 L of sediment while WC values were applied to correct sediment dry weight. Therefore, later reported microplastic concentrations per kilogram dry weight (kg DW) refer to sediments dried at 105 °C.

2.4. Microplastic identification

Filters were stained using 1 ml of the lipophilic dye Nile Red solved in Chloroform (1 mg/ml) according to Tamminga et al. (2017). After drying (24 h), they were analyzed in a UV light photobox (for lakeshore samples from September 2017 only) and under a fluorescent microscope (AxioLab A.1, Zeiss, TRITC HC filterset (AHF), 2.5 magnification) with external illuminant (as described in Hengstmann and Fischer 2019). One and multiple images per filter were taken in the UV light photobox (Pentax K-30, exposure time 2″, ISO 100, min. resolution 2 × 2 mm) and under the microscope (Canon EOS 80D, exposure time 1″, ISO 500, resolution 1 × 1 μm), respectively. Images from the UV light photobox were analyzed in Photoshop (©Adobe, Version CS5) and microplastic particles were counted and measured, differentiating fragments and fibers. Images from fluorescence microscopy were analyzed in a semi-automated process described below.

September 2017 lakeshore samples were primarily photographed in the UV light photobox and the counting process differed slightly. Only particles matching the size-class of subsamples resulting from wet-sieving were counted due to lower resolution of UV light photobox images. For the subsequent sampling campaigns, all particles ≥63 μm were counted in the microscope images as fractioning via sieving might not be free from error, especially for small particles (Lenaker et al., 2021; Enders et al., 2020; Filella 2015). Therefore, microplastic concentrations for lakeshore samples from September 2017 were calculational adapted to fit the scale of following sampling concentrations. Detailed information on calculations is provided in the Supplementary Material (SM; Text SM1).

An automated software tool was used for the detection of microplastics in microscope images for sampling campaigns after September 2017, to accelerate the evaluation and ensure consistency between measurements. For this purpose, a dedicated Python (Van Rossum and Drake 2009) code based on the open-source libraries openCV (Bradski 2000) and Scikit-Learn (Pedregosa et al., 2011) was written. The automated process was divided into segmentation, classification, and evaluation of particle properties. The segmentation step, i.e. identifying contours of potentially relevant particles (fragments or fibers), was performed by first converting each original RGB microscope image to a greyscale version. Each pixel in such a greyscale image is represented by an integer between 0 (black) and 255 (white). Afterwards, a threshold value of 60 was used to convert each image to a bi-level black and white image. This approach was highly effective, because Nile Red stained particles appeared with a grey style value well above 60 in all samples. Consequently, the thresholding does not miss any stained particles. The contours of all remaining particles could then be identified by openCVs contour finder. Only those above a minimal size of 63 μm (determined by the smallest mesh size during wet sieving) were kept. For the classification step, a set of features was used to classify particles by their type (plastic vs. non-plastic) and shape (fragment vs. fiber). To achieve this, a set of training data with roughly 1,300 particles, was created and each particle was classified manually. A classifier was then trained based on two color features: (i) average H-value (using HSV color space) of the pixels on the contour and (ii) average H-value of all enclosed pixels. A high classification accuracy could be achieved, independent of the type of classifier. A second classifier based on geometrical features such as the particle area, perimeter, and object moments, was trained to distinguish between fragments and fibers. The segmentation and classification of each plastic candidate was manually checked to ensure the results were comparable to previous manual classifications. For the evaluation step, the code determined the length and width of the minimal enclosing rectangle around the contour to calculate the length and width of a microplastic fragment. In case of fibers, the approximation of the width was based on the area, a, and perimeter, p. Because the perimeter, p, is very close to twice the length, l, the width can be estimated by $a/l = 2a/p$. From the perimeter and width, the length was directly calculated.

Major advantages of this semi-automated approach are the unbiased classification of particles, a more efficient analysis of microscopic images, and that geometrical properties of the selected contours can be determined automatically with high precision. In practice, the automated approach worked very well for microplastic fragments, but was less effective for fibers. This was reflected by cases of incorrect segmentation, especially when fibers were not completely stained by the Nile Red or when particles overlapped. Future improvements could be made by moving from a static-towards a more flexible deep-learning-based approach, possibly leading to a fully automated setup.

2.5. Spectroscopic analysis

Subsets of identified particles were analyzed spectroscopically to gain information on their chemical composition. Selected particles were transferred onto object slides by tweezers for μ-Raman spectroscopy (DXR2xi Raman Imaging Microscope, Thermo Fisher Scientific). Particles were analyzed using a 532 nm laser and magnifications of 10× or 50×. A laser power of 5–10 mW was applied in combination with an exposure time of up to 10 Hz. At least 500 spectra were incorporated. The resulting spectrum was compared to multiple spectrum libraries. Best fits were chosen on account of the match (>70%) and a visible comparison by the analyst. In total, a subset of 571 particles from lakeshore and lakebed sediments were tested corresponding to 2% of all particles identified on microscopic images. Figure SM1 shows examples of different polymers identified by the fluorescence microscope and μ-Raman spectroscope.

2.6. Quality assurance and quality control (QA/QC)

Strict anti contamination conditions were kept during laboratory analysis, i.e., wearing cotton lab coats and nitrile gloves, regular cleaning of working surfaces, using glass or stainless-steel equipment whenever possible, rinsing equipment with deionized water or acetone before use and covering samples between the processing steps. Additionally, several blank samples (one per three sediment samples) were processed in parallel to assess the remaining contamination by laboratory material and via air. Blank samples started with empty, rinsed glass beakers and passed all laboratory and identification steps. Statistical outliers within the blank sample dataset were excluded from the subsequent evaluation of contamination levels. Mean numbers of microplastics found in blank samples were subtracted from microplastic counts of analyzed sediment samples by size class for each individual sampling. Furthermore, two lakeshore sediment samples were analyzed in three parallels for their microplastic concentration to assess the variability within one sample.

2.7. Statistical analysis

Statistical analyses were performed using R statistics (R Core Team

2020, Version 3.6.3) in an R Studio environment (RStudio Team 2019, Version 1.2.5033). The Shapiro-Wilk test was applied to test for normal distribution of microplastic concentrations and pedologic values as $n \leq 50$ for most sample sets. For data showing a non-normal distribution, differences between groups were tested using the Kruskal-Wallis test with post-hoc Dunn test. For normally distributed data, one-way ANOVA was applied to test for differences between groups with post-hoc Tukey test. The significance level p was set to 0.05. Relations of microplastic concentrations with wind, pedologic parameters, grain size fractions and anthropogenic litter abundances were tested using the Spearman correlation index.

Visualization of results was performed using the R-library ggplot2 (Wickham 2016). Additionally, ArcGIS (©ESRI, Version 10.3) was used for map visualizations.

3. Results

3.1. Quality assurance and quality control (QA/QC)

The evaluation of QA/QC measures resulted in a mean and standard deviation (SD) of 28 ± 16 microplastic fragments and 8 ± 4 fibers per sample for blank samples paralleled with lakeshore sediments. The contamination for lakebed sediment blanks was 54 ± 33 fragments and 9 ± 4 fibers per sample. Many individual laboratory processing steps might cause contamination in samples. Increased microplastic numbers in blank samples for lakebed sediment analysis may result from more extensive rinsing between digestion steps due to very fine material. For both blank sample sets, contamination increased with decreasing size of particles. Analysis via μ-Raman-spectroscopy revealed that 83% of analyzed fragments (n = 90) were made of polyethylene (PE), probably resulting from dash bottles made of low-density polyethylene (LDPE) and used for each washing step. Fibers (n = 41) were mainly polyethylene terephthalate (PET) fibers (95%) showing that airborne contamination is still apparent even though anti contamination measures were provided.

For the assessment of variability in lakeshore samples, replicates were analyzed. The analysis of three replicates resulted in an average microplastic concentration of 16 ± 25 particles per replicate for the first lakeshore sediment sample. The mean relative percent difference (RDP) was 22%. For the second triplet sample, a mean concentration of 138 ± 33 particles per replicate was found. In this case, the mean RDP was 34%. The two samples chosen for the analysis of replicates strongly differ in their overall microplastic concentration. However, the variation between samples, represented by standard variation and RDP, is similar. The variation between replicates may be explained by insufficient homogenization of samples before extracting subsamples for microplastic analysis, heterogeneity of the sediment itself and/or small variabilities during laboratory processing.

3.2. Pedologic characteristic of sediments

Mean (±SD) water content for lakeshore samples was 0.08 ± 0.05% (median (MD) = 0.07; interquartile range (IQR) = 0.06) whereas lakebed sediments were characterized by higher water content with 2.22 ± 0.56% (MD = 2.00; IQR = 0.88). Similarly, analysis of organic matter as LOI resulted in a lower mean of 0.27 ± 0.28% (MD = 0.16; IQR = 0.23) for lakeshore sediments compared to lakebed sediments (8.97 ± 0.56%; MD = 9.24; IQR = 0.99). Significantly higher percentages of organic matter were found in samples from South and West beach. The intertidal zone showed significantly lower organic matter content compared to the other two littoral zones. For lakebed sediments, no significant differences in water content or organic matter content were registered.

Grain size analysis and classification according to DIN EN ISO 14688-1:2020-11 (2020) showed that sand (0.063–<2 mm) is the predominant grain size in lakeshore sediment samples with grain sizes <0.063 mm accounting for only 0–2.97%. Within the grain size fraction of sand, medium sand (0.2–<0.63 mm) was dominating with 80.88 ± 16.33% (MD = 85.24; IQR = 18.78) (Fig. SM2). Conversely, sand was least abundant in lakebed sediments making up 0.87–2.92%. Here, silt (0.002–<0.063 mm) was the dominating grain size fraction with a mean of 76.46 ± 3.11% (MD = 77.06; IQR = 4.31) (Fig. SM2). West and South beach were characterized by significantly higher shares of silt/clay and medium sand fractions compared to East beach, and East beach in reverse showed significantly higher abundances of fine sand. Furthermore, coarse sand was significantly more abundant at the intertidal zone. Significant differences were found for lakebed sediments concerning the comparison between four lake regions (North, Central-North, Central-South, South) when combining two sampling points. The sand fraction was significantly more abundant in the South compared to the Central-North, whereas clay was found at higher levels in the North compared to the South. By contrast, silt was significantly less abundant in the North.

3.3. Microplastics in lakeshore sediments

Microplastics were detected in all lakeshore samples at Lake Tollense. The total mean (±SD) of microplastic abundance was 1,410 ± 822 particles/kg DW (MD = 1,328; IQR = 905). Fragments were more abundant than fibers with 1,033 ± 706 particles/kg DW (MD = 878; IQR = 860) and 376 ± 246 particles/kg DW (MD = 324; IQR = 297), respectively. Microplastic concentrations were furthermore expressed as particles per area, referring to a sampling depth of 1–2 cm, for comparability reasons. A mean value of 36,943 ± 20,697 particles/m^2 was detected (MD = 33,620; IQR = 21,875). When expressed as particles per volume, the mean microplastic concentration was 2,243 ± 1,219 particles/L (MD = 2,122; IQR = 1,356). Detailed data for all sampling campaigns is available in the SM (Tab. SM1).

3.3.1. Sizes distribution

Analyzing the size of microplastics, the sampling in September 2017 was excluded since no information on individual microplastic size was available due to methodical differences concerning identification. In general, an increase in microplastic particles with decreasing size was detected. However, fragments and fibers behaved differently. Increasing abundances with decreasing size was further visible for fragments, whereas fibers were evenly distributed across the whole size range (Fig. 2). The mean size of fragments was 184 ± 149 μm (MD = 146; IQR = 118). For fibers, mean length was higher with 1,010 ± 916 μm (MD = 711; IQR = 1,005). At East beach, significantly larger fragments and fibers were present. Furthermore, fragments were significantly larger in the intertidal zone compared to the high water line and the elevated area.

3.3.2. Temporal and spatial variability

Microplastic abundance differed temporally and spatially. The sampling in March 2018 showed the highest particle concentration in lakeshore sediments, whereas the lowest abundance was found in September 2018 (Table 1). Similarly, microplastic fragment abundances were significantly lower in September compared to abundances in March. In contrast, fiber abundance was significantly higher in September 2017 compared to the other sampling campaigns. March 2019 showed significantly lower fiber concentrations.

Highest microplastic concentration was found at South beach with 2,072 ± 977 particles/kg DW (MD = 1,906; IQR = 1,057), for particles in total but also for fragments and fibers individually. East beach showed the least contamination by microplastics with a mean of 940 ± 597 particles/kg DW (MD = 838; IQR = 965). The same pattern was visible for fragments whereas fibers were least abundant at North beach. Significant differences were found between South beach and all other beaches for particles, fragments, and fibers as well as between East and West beach for particles and fragments.

Spatial differences were also detected between different sampling

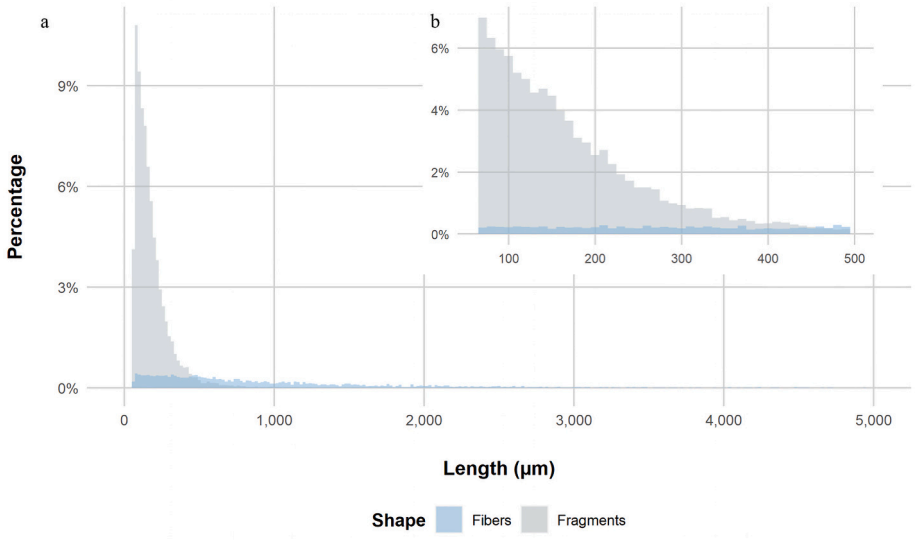

Fig. 2. Size distribution for microplastic fragments and fibers in lakeshore sediments. a) Overall size distribution between 0 and 5,000 µm (bin width = 20 µm) and b) detailed section between 63 and 500 µm (bin width = 10 µm).

Table 1
Microplastic abundance per kg DW for individual sampling campaigns according to their shape. Abundances are presented as mean ± standard deviation and median with interquartile range. Superscript letters indicate significant differences between sampling campaigns (p = 0.05).

Sampling	Particles	Fragments	Fibers
Sep. '17	1,297 ± 375^{ab}	802 ± 240^{a}	495 ± 202^{a}
	1,247; 426	759; 276	461; 235
Mar. '18	1,672 ± 850^{a}	1,362 ± 745^{b}	311 ± 151^{b}
	1,714; 1,014	1,403; 841	300; 208
Sep. '18	1,118 ± 926^{b}	704 ± 723^{a}	414 ± 255^{b}
	616; 1,266	358; 772	351; 403
Mar. '19	1,524 ± 1,091^{a}	1,297 ± 875^{b}	227 ± 307^{c}
	1,470; 1,417	1,252; 1,260	163; 147

points within the littoral zone (Fig. 3). Significantly lower microplastics were detected in the intertidal zone (1,159 ± 705 particles/kg DW; MD = 1,091; IQR = 881), whereas the sampling points at the high water line and within the elevated area showed higher microplastic concentrations with 1,527 ± 771 particles/kg DW (MD = 1,470; IQR = 999) and 1,594 ± 958 particles/kg DW (MD = 1,419; IQR = 858), respectively. The

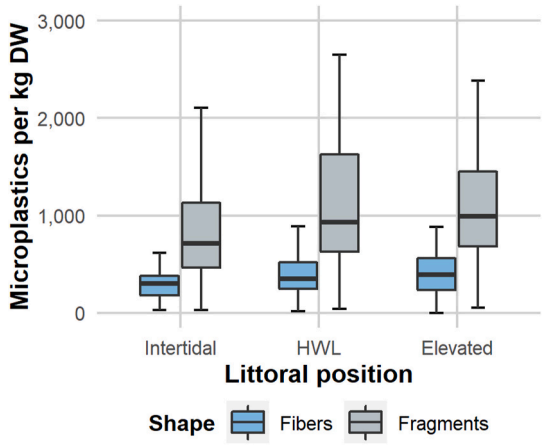

Fig. 3. Fragment and fiber abundances per kg DW in lakeshore sediments regarding the three different littoral sampling points. HWL = high water line. Statistical outliers are not displayed.

same is true for fragments and fibers individually.

3.3.3. Polymer composition

Analysis via µ-Raman spectroscopy was conducted for 447 particles from Lake Tollense shore sediments. The synthetic origin was confirmed for 92.8% of these particles consisting of polymers (89.6%), pigments (2.8%) and plasticizers (0.4%). The remaining particles were of biogenic (5.4%) and mineral origin (1.8%). For fragments, PE was the predominant polymer with 73% followed by polyamide (PA; 10%), polypropylene (PP; 8%) and PET (6%). Other polymers represented 1% or less each (Fig. 4a). Chemical composition of fibers was strongly dominated by PET with 92% of all fibers analyzed. PA and PP still accounted for 3% and 2%, respectively (Fig. 4a).

3.4. Microplastic in lakebed sediments

Microplastic particles were furthermore detected in all lakebed samples from Lake Tollense. Mean microplastic concentration was 10,476 ± 4,290 particles/kg DW (MD = 9,879; IQR = 6,809). Again, fragments (82%) were more abundant than fibers (18%). Detailed data for all sampling campaigns is available in the SM (Tab. SM2). Table 2 summarizes important variables for lakebed sediments compared to lakeshore sediments concerning sediment characteristics and microplastic pollution. Microplastic concentrations were not expressed as particles per area or particles per volume for lakebed sediments due to larger sampling depths via van-Veen grab sampling, unknown volumes of the complete sample due to subsampling, and because no information on the bulk density of sediment was gathered for lakebed sediments.

3.4.1. Size distribution

Size distribution across the analyzed size range differed between fragments and fibers. Fibers were evenly distributed over all size classes. Fragments increased with decreasing size up to a size of around 135 µm. Below this size limit, microplastic concentrations slightly decreased (Fig. 5). The described behavior was true for all sampling campaigns individually, as well as for single sampling points. Mean length for fragments within lakebed sediments was 192 ± 152 µm (MD = 151; IQR = 117) whereas mean length for fibers was higher with 873 ± 783 µm (MD = 610; IQR = 777).

3.4.2. Temporal and spatial variability

No significant differences between the sampling campaigns were

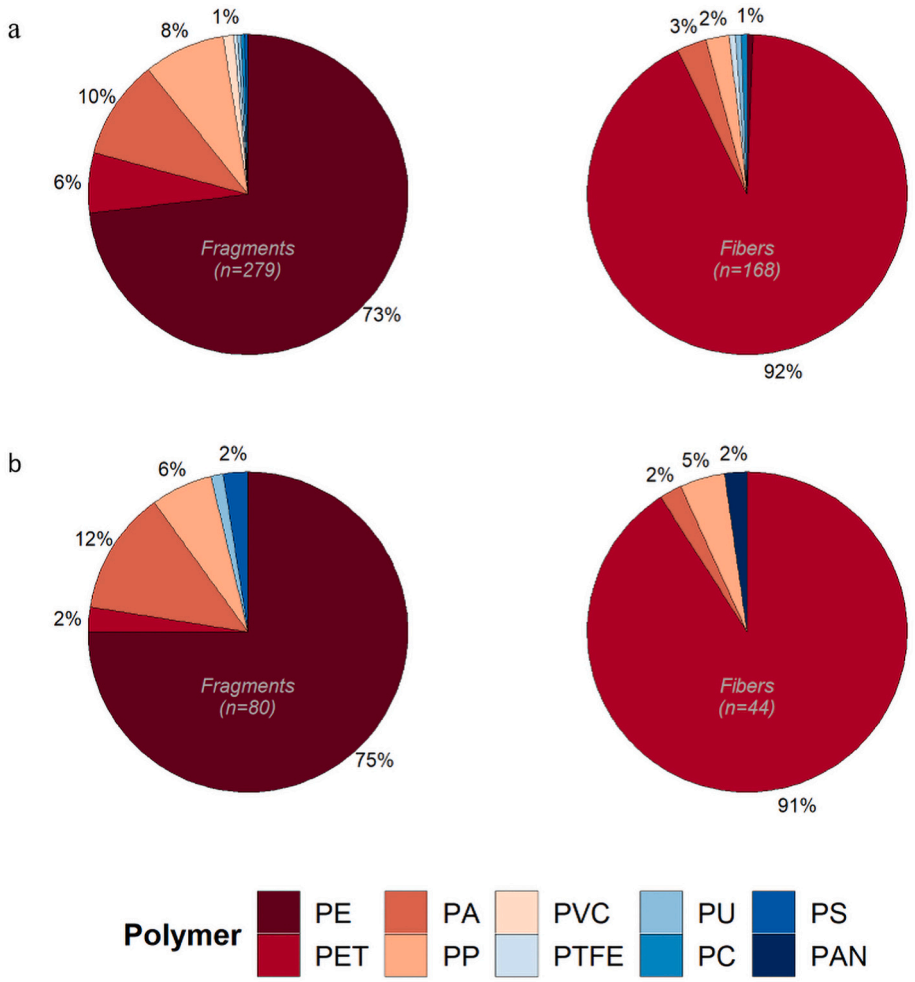

Fig. 4. Percentage of different polymers analyzed via μ-Raman spectroscopy for a) lakeshore sediments and b) lakebed sediments according to particle shape. The number of analyzed particles is given in parentheses (n = x). PE = polyethylene, PET = polyethylene terephthalate, PA = polyamide, PP = polypropylene, PVC = polyvinyl chloride, PTFE = polytetrafluoroethylene, PU = polyurethane, PC = polycarbonate, PS = polystyrene, PAN = polyacrylonitrile.

Table 2
Comparison of results for major variables between lakeshore and lakebed sediments. Values are presented as mean ± standard deviation except for the percentage of fragments and fibers and the polymer composition.

	Lakeshore sediments	Lakebed sediments
Water content (%)	0.08 ± 0.05	2.22 ± 0.57
Organic content (%)	0.27 ± 0.28	8.97 ± 0.56
Silt and clay content (%)	0.61 ± 0.49	92.02 ± 3.61
Microplastic conc. (particles/kg DW)	1,410 ± 822	10,476 ± 4,290
Fragments and fibers (%)	74/26	82/18
Size fragments (μm)	184 ± 149	192 ± 152
Size fibers (μm)	1,010 ± 916	873 ± 783
Polymer composition	PE > PET > PA > PP	PE > PET > PA > PP

detected. However, tendencies in the temporal distribution were visible. Microplastic particle and fragment concentrations were highest for September 2017 whereas lowest abundances were found for March 2018. Fiber concentrations were higher in March 2018 and September 2018. Due to low sample numbers per sampling point (n = 4) significance tests concerning spatial differences are not representative. Sampling point SLU_3 tended to show the highest abundances for microplastic particles and fragments, whereas fibers were most abundant at the two northern sampling points SLU_7 and SLU_8 (Fig. 6). Significant differences were found for fibers when two sampling points were combined into a regional classification of the lake (North, Central-North, Central-South, South). In this case, fibers showed significantly higher concentrations in the northern part of the lake compared to the Central-North and South.

3.4.3. Polymer composition

In total, 124 particles from lakebed sediment samples were analyzed via μ-Raman spectroscopy. Of these, 97.6% were confirmed as synthetic polymers and 1.6% were classified as artificial pigments. Biogenic particles accounted for 0.8% of all particles analyzed. PE was the predominant polymer for fragments with 75%. PA and PP accounted for 12% and 6%, respectively, whereas PET, polyurethane (PU) and polystyrene (PS) represented 2% or less (Fig. 4b). In contrast, fibers were dominated by PET with 91% (Fig. 4b). Further polymers identified were PP (5%), PA (2%) and polyacrylonitrile (PAN; 2%).

3.5. External influence on microplastic concentrations

A low significant correlation was detected between microplastic fiber concentrations and water content of lakeshore sediments. Significant correlations were further found between microplastics and organic content (Tab. SM3). A positive, significant but moderate correlation between LOI values and fiber concentrations was found for lakeshore sediments ($r = 0.37$, $p < 0.001$). Consequently, fiber abundance is increasing with increasing organic content of the sample. This relation was most pronounced for the sampling in September 2018 and least

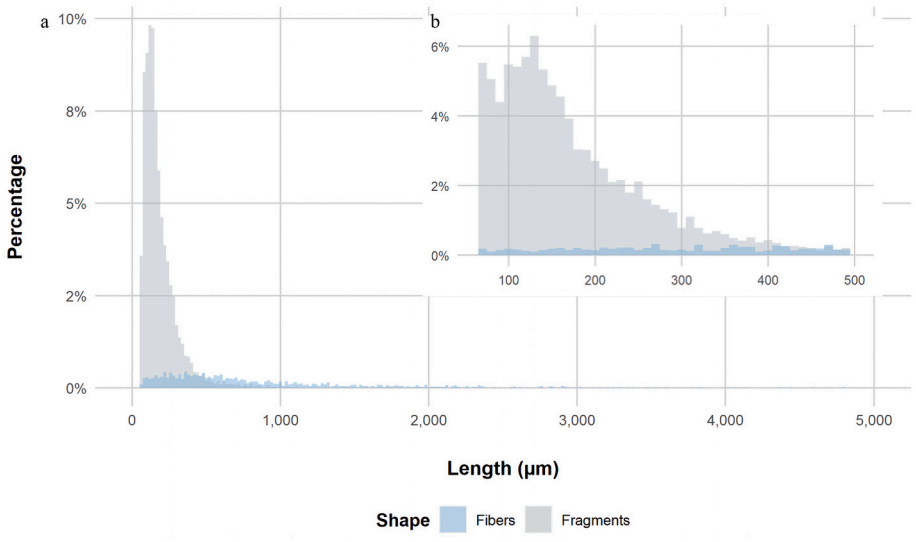

Fig. 5. Size distribution for microplastic fragments and fibers in lakebed sediments. a) Overall size distribution between 0 and 5,000 µm (bin width = 20 µm) and b) detailed section between 63 and 500 µm (bin width = 10 µm).

pronounced for September 2017. Fragments significantly and strongly correlated with LOI values only in September 2018 ($r = 0.56$) and March 2019 ($r = 0.54$). For lakebed sediments, no significant correlation between fragment or fiber abundances and water content or LOI values were verified including all samples. The individual sampling campaigns showed diverging results probably due to small sample numbers ($n = 8$). When combining lakeshore and lakebed sediments, a strong ($r = 0.61$) and significant correlation was verified between fiber abundances and organic content as well as a moderate ($r = 0.43$) and significant correlation between fragments and organic content ($p < 0.001$). Spearman correlation coefficients showed a low but significant correlation between microplastic fragments and particles and the content of clay and silt grain size fraction for lakeshore sediments. However, this trend could not be verified for lakebed sediments. Similarly, correlation coefficients did not reveal a clear trend concerning relations with sand grain size fractions (Tab. SM3).

Microplastic abundances in lakeshore sediments were further correlated to wind data. Correlations were more pronounced for the relation between fiber abundance and wind direction compared to fragment abundance. However, correlations were not significant. Wind velocities from the DWD dataset showed a positive, moderate and significant correlation with microplastic particle concentration (Tab. SM3). This refers to increasing microplastic abundances with increasing wind velocity. Combining relative wind direction and relative wind velocity in one variable, the correlation to fragment abundance was moderately. Again, correlations were not significant, and fiber abundance did not show a relation to the combined wind variable at all.

Lakeshore segments at Lake Tollense were not only investigated regarding their microplastic concentrations, but anthropogenic litter monitoring was also conducted (results see Hengstmann and Fischer 2020). Values from both investigations were related to each other. Microplastic particle and fragment abundance were strongly, negatively correlated to the number of macroplastic items at the beach segments at a significance level of $p < 0.001$ (Fig. 7). Contrastingly, fiber abundances showed a moderate, not significant relation. This correlation indicates that microplastic particle and fragment abundance decrease with increasing macroplastic abundance. The general negative relation was not confirmed for South beach when considering beaches separately.

4. Discussion

4.1. Microplastics in lake sediments

Microplastic abundances in freshwater environments have been studied around the world in the past decades. However, a comparison between the individual studies is hampered due to differences in methodical approaches as no standard protocol for microplastic identification in sediments exists so far (Koelmans et al., 2019; Prata et al., 2019; Dris et al., 2018; Horton et al., 2017a; Ivleva et al., 2017; Van Cauwenberghe et al., 2015). Therefore, results of this study can only be compared to other freshwater studies with reservations. An overview of freshwater sediment studies is provided in Table 3, including a short description of applied methods. Microplastic abundances in Lake Tollense shore and bed sediments seem to be predominantly higher compared to other studies in Asia, Europe, and North America. At first sight, results are exclusively comparable to the microplastic pollution found in lakeshore sediments of four lakes in Bavaria, Germany, with 99–129,375 particles/m^2 (LfU, 2019).

Diverging methods applied in different studies might influence the concentrations, though. Most importantly, analyzed size ranges differ between studies. At Lake Tollense, particles as small as 63 µm and up to 5,000 µm in length were analyzed. This broad size range of particles partially differs from size ranges reported by other studies. According to Koelmans et al. (2020), microplastic concentrations can be transferred to size classes divergent from the ones analyzed. Following the approaches provided by Kooi and Koelmans (2019) and Koelmans et al. (2020), Lake Tollense concentrations were transferred to other size ranges to compare abundances to former lake studies. According to these transfer calculations, microplastic abundances at Lake Tollense were more comparable to values of other studies. For example, transferring microplastic concentrations for lakebed sediments to the size range 174–5,000 µm as analyzed by Zobkov et al. (2020), abundances for Lake Tollense are in the same order of magnitude (mean 2,770 particles/kg DW) as the results presented for Lake Onego (mean 2,189 particles/kg DW). Similarly, the difference in microplastic concentrations between the most polluted lake in Switzerland, Lake Brienz, with 2,500 ± 3,000 particles/m^2 (Faure et al., 2015) and Lake Tollense is much smaller when transferring the data to the same size range (6,388 particles/m^2). Transferred mean and median values for several size ranges for Lake Tollense microplastic concentrations are presented in the SM (Tab. SM4).

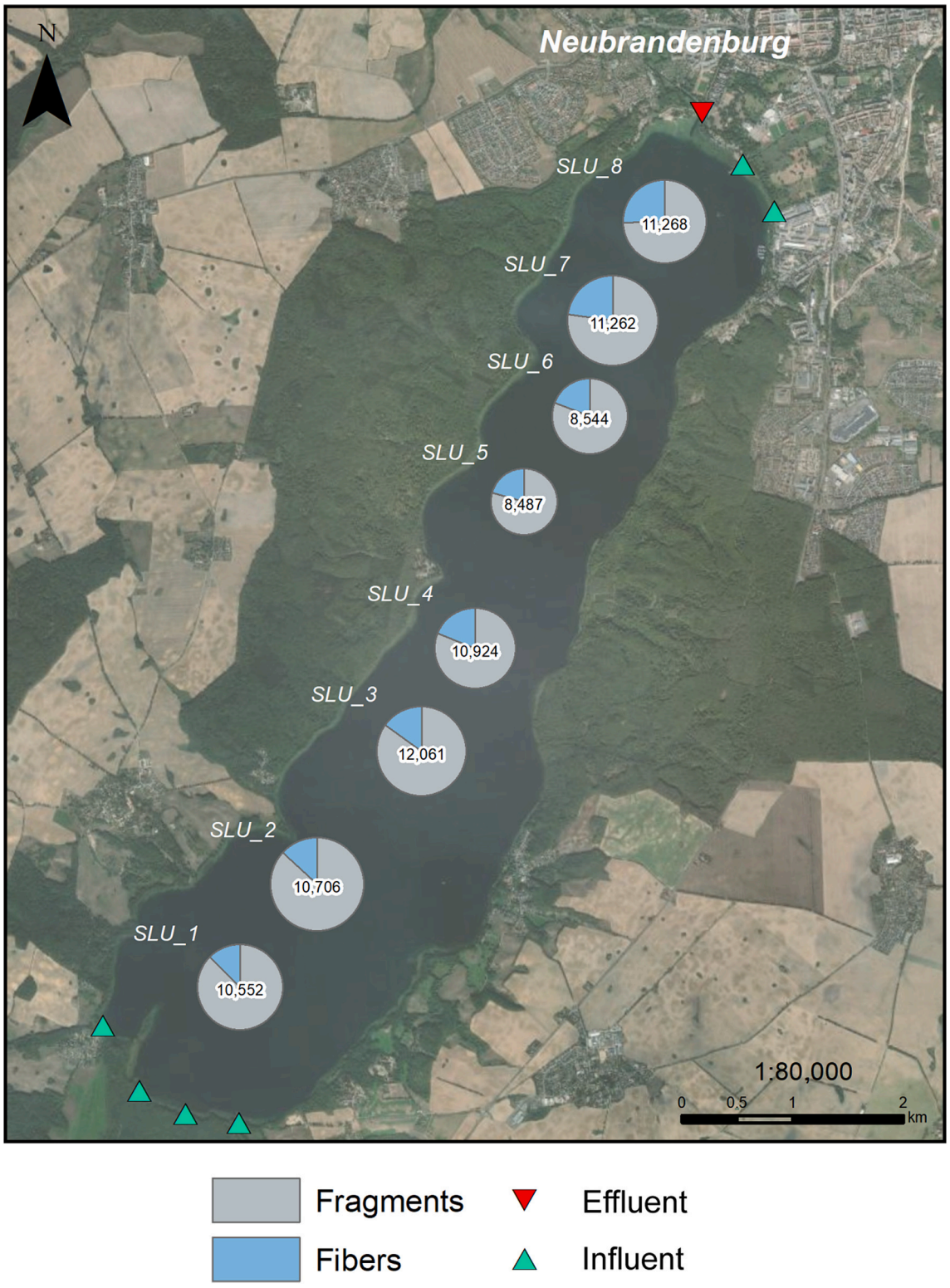

Fig. 6. Microplastic abundance per kg DW in lakebed sediments and respective shares of fragments and fibers. Pie charts visualize relative mean concentrations and shape percentages whereas numbers refer to mean abundances at the sampling location. Triangles point to the in- and outflow of tributaries. Projection: Transverse Mercator; Coordinate system; WGS_1984_UTM_Zone_32N; Satellite image: ArcGIS. Imagery.

Further methodical differences, as the choice of density separation approaches or the microplastic identification process, may have an impact on concentration differences between studies. For example, studies using sodium chloride (NaCl) for density separation might not extract polymers with densities >1.2 g/cm^3 (Ivleva et al., 2017; Löder and Gerdts 2015; Van Cauwenberghe et al., 2015). Therefore, they might underestimate numbers compared to the elutriation column used in this study. Studies only relying on the visual inspection of particles may not report actual concentrations as visual sorting is inaccurate especially for small particles (Silva et al., 2018; Ivleva et al., 2017; Lenz

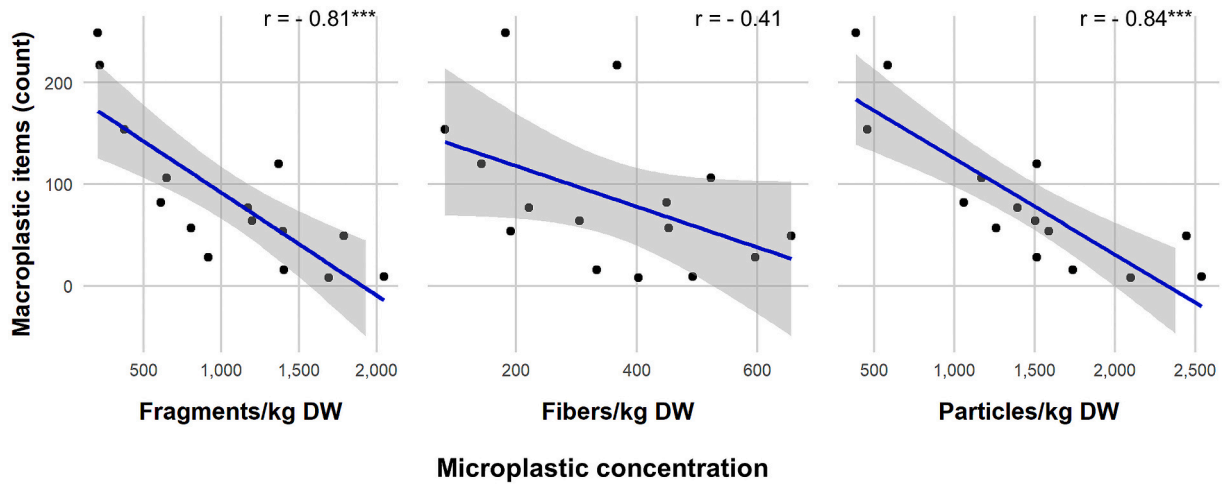

Fig. 7. Relation between microplastic concentrations and the number of plastic litter items at Lake Tollense beaches. Scatterplots refer to fragment, fiber and particle abundances, respectively. Blue line represents linear regression line with confidence interval (0.95) shaded in grey. Spearman correlation coefficient r is presented in combination with significance level (*** refers to $p < 0.001$). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

et al., 2015; Löder and Gerdts 2015). In this study, visual inspection was enhanced by using a staining approach via Nile Red and fluorescence microscopy as well as by analyzing a subset of particles by μ-Raman spectroscopy.

High microplastic concentrations in a deep and dimictic lake as Lake Tollense might be explained by several factors. Lake sediments act as a temporary and long-term storage for microplastics (Li et al., 2020; Bordós et al., 2019; Turner et al., 2019; Vaughan et al., 2017; Imhof et al., 2013) and function as a smaller but similar reservoir when compared to ocean basins where microplastic particles accumulate. Lake Tollense is comparably deep (max. 31.2 m) providing very calm and steady conditions at the lakebed. Therefore, the retention of particles in lakebed sediments might be increased. Especially compared to rivers, which are very dynamic and show high microplastic abundances in sediments as well (e.g., Lenaker et al., 2019; Ballent et al., 2016; Klein et al., 2015; Castañeda et al., 2014), lakes promote sedimentation. Therefore, accumulation and concentration of microplastic in lake sediments is enhanced (Zobkov et al., 2020). As verified by the study results, accumulation of microplastics is not only present for lakebed sediments but microplastic particles further accumulate at beaches at the high water line and in elevated areas compared to the water line (see section 4.5).

Lake Tollense is furthermore characterized by a large catchment area. A high percentage of agricultural area (56.4%; Nixdorf et al., 2004), the proximity to the city of Neubrandenburg and the touristic activity within the catchment may function as extensive sources for microplastics. Microplastics are, among others, transported into the lake via surface runoff from agricultural and inhabited areas and by tributaries leading to elevated pollution of Lake Tollense sediments by microplastics. Former studies also identified agricultural areas as a source for microplastics (Lenaker et al., 2021; Rochman 2018; Horton et al., 2017b; Wagner et al., 2014) and human presence or activity in general are major origins for plastics in freshwater environments (Baldwin et al. 2016, 2020; Bellasi et al., 2020; Helm et al., 2020; Dris et al., 2018; Horton et al., 2017a; Faure et al., 2015). Furthermore, direct littering at the shores of Lake Tollense, especially during the tourist season from May to September, can increase microplastic pollution in sediments when large plastic items fragment *in situ* (Dris et al., 2018; Andrady 2011; Barnes et al., 2009).

4.2. Size distribution of microplastics

The general increasement of microplastic fragment abundance with decreasing size is in accordance with former microplastic studies in lake sediments (Lenaker et al., 2021; Baldwin et al., 2020; Hu et al., 2020; Kooi and Koelmans 2019; LfU, 2019; Yuan et al., 2019; Fischer et al., 2016). The described pattern is related to the degradation of larger fragments by environmental factors such as UV irradiation as well as abrasion by wind and wave activity, enhanced in beach environments, which lead to the formation of more and simultaneously smaller fragments (Helm 2020; Andrady 2011; Barnes et al., 2009). As fibers are less concentrated and more equally distributed across the complete size range in sediments of Lake Tollense, factors controlling the degradation may have lower or different effects on this type of microplastics. For example, Sørensen et al. (2021) showed that UV induced surface morphology changes rather occurred for PA fibers instead of fragmentation. Furthermore, fibers are more prone to transport via air (Bullard et al., 2021; Dris et al., 2016; Torre et al., 2016). Accumulation on beach surfaces is thereby reduced whereas resuspension by wind is increased.

For lakebed sediments, the exponential increase with decreasing size stopped at around 135 μm. Similarly, Imhof et al. (2016) found a maximum in particle sizes at ca. 130 μm for microplastics in lake sediments. Microplastic fragments of this size may increasingly be mistaken for food particles and ingested by organisms during settling to the lakebed. The ingestion of particles by organisms is related to the size of particles (O'Connor et al., 2020; Ivleva et al., 2017; Scherer et al., 2017; Setälä et al., 2014; Cole et al., 2013). For example, *Gammarus pulex*, common in freshwater communities, only ingested PS particles between 16 and 165 μm in a laboratory experiment even though larger particles were available (Redondo-Hasselerharm et al., 2018). Furthermore, small particles might stay in the water column inducing the maximum at circa 135 μm in size. Especially small polymer particles with densities lower than water are too small and lightweight to sink to the lakebed. Particle sinking velocities are lower for particles of small sizes (Kaiser et al., 2019; Khatmullina and Isachenko 2017; Kowalski et al., 2016). The formation of bacterial or algal biofilms on plastic particle surfaces, so-called biofouling, can start and enhance the sinking process for small and low-density polymers (Kaiser et al., 2017; Chubarenko et al., 2016; Lagarde et al., 2016; Lobelle and Cunliffe 2011). However, biofilms can also be reduced again during sinking and particles stay within the water column or start to rise again (Kooi et al., 2017; Andrady 2011; Ye and Andrady 1991).

Methodological factors can play a role in the detection of very small particles as well. Aged, more altered particles are expected in lakebed sediments due to long-term accumulation of microplastics. The detection of smallest and aged particles by identification methods such as Nile

Table 3
Comparison of microplastic concentrations per kg DW or per m^2 in lakeshore and -bed sediment including information on sampling and laboratory procedures. If not stated, mean microplastic (MP) concentration is given. n.r.= not reported. SPT = sodium polytungstate.

Study	Country	Study area	Lake characteristics (area/ depth/catchment)	Sample number	Sampling method (ø or dimension/depth)	Preparation	Density separation	Identification	Size range	Abundances
					Lakeshore sediments					
present study	**Germany**	**Lake Tollense**	**17.9 km^2/mean 17.6 m/ 515 km^2**	**185**	**Quadrats (25 × 25 cm/1-2 cm)**	**sieved, digestion**	**elutriation**	**Nile Red, subset Raman (n = 447)**	**63 - 5,000 µm**	**1,410 ± 822 MP/kg DW 36,943 ± 20,697 MP/m^2**
LfU (2019)	Germany	Bavaria (4 lakes)	n.r./n.r./n.r.	22	sediment tubes (10 cm/5 cm)	digestion	MPSS/$ZnCl_2$ (1.65 g/cm^3)	microscope, FTIR	20 - 5,000 µm	median 17,068 MP/m^2 (99–129,375)
Faure et al. *(2015)*	Switzerland	6 lakes	68–581 km^2/n.r./ 1,127–10,856 km^2	33	quadrats (30 × 30 cm/ 5 cm)	sieved, digestion	NaCl (1.2 g/cm^3)	microscope, subset FTIR spectroscopy	300 - 5,000 µm	1,300 ± 2,000 MP/m^2
Imhof et al. *(2018)*	Italy	Lake Garda	n.r./n.r./n.r.	15	sediment tubes (10 cm/5 cm)	digestion	MPSS/$ZnCl_2$ (1.65 g/cm^3)	Raman	1 - 5,000 µm	3,508 ± 8,855 MP/m^2
Dean et al. *(2018)*	Canada	Lake Erie	n.r./mean 19 m/n.r./n.r.	12	split spoon corer (-/30 cm)	sieved	SPT (1.5 g/cm^3)	microscope, subset Raman	63 - 5,000 µm	50 - 146 MP/kg DW
Ballent et al. *(2016)*	Canada	Lake Ontario	n.r./n.r./> 64,000 km^2	10	split spoon corer (-/30 cm)	sieved	SPT (1.5 g/cm^3)	microscope, subset Raman	63 - 5,600 µm	140 MP/kg DW
Xiong et al. *(2018)*	China	Quinghai Lake	4,500 km^2/mean 20 m/ 30,000 km^2	7	quadrats (20 × 20 cm/ 0–2 cm)	sieved	$CHKO_2$ (1.5 g/cm^3)	visual, microscope, subset Raman	112 - 5,000 µm	67 - 1,292 MP/m^2
Zhang et al. *(2016)*	China	Tibet Plateau (4 lakes)	n.r./n.r./n.r.	42	Quadrats (20 × 20 cm/ 2 cm)	sieved	$CHKO_2$ (1.5 g/cm^3)	visual, microscope, subset Raman	n.r. - 5,000 µm	max. 563 ± 1,219 MP/m^2
					Lakebed sediments					
present study	**Germany**	**Lake Tollense**	**17.9 km^2/mean 17.6 m/ 515 km^2**	**32**	**van-Veen grab (~16 × 16cm/-)**	**sieved, digestion**	**elutriation**	**Nile Red, subset Raman (n = 124)**	**63–5,000 µm**	**10,476 ± 4,290 MP/kg DW**
Turner et al. *(2019)*	UK	Hampstead No. 1 Pond	0.015 km^2/max. 3 m/0.7 km^2	1	piston corer (6cm/212 cm)	sieved	SPT (2.1 g/cm^3)	microscope, subset Raman	>500 µm	max. 539 MP/kg DW
Zobkov et al. *(2020)*	Russia	Lake Onego	9,720 km^2/mean 17.8 m/ 53,100 km^2	11	Peterson, Box corer grab (-/5 cm)	sieved, digestion	$CHKO_2$ (1.5 g/cm^3)	microscope, subset Raman	174 - 5,000 mm	2,189 ± 1,164 MP/kg DW
Ballent et al. *(2016)*	Canada	Lake Ontario	n.r./n.r./> 64,000 km^2	33	corer, grab (6.5, 20 cm/6–15 cm)	sieved	SPT (1.5 g/cm^3)	microscope, subset Raman	63 - 5,600 µm	980 MP/kg DW (max. > 27,000 MP/kg)
Baldwin et al. *(2020)*	USA	Lake Mead & Mohave	759 km^2/n.r./n.r.	9	Ponar grab (~15 × 15cm/3 cm)	sieved, digestion	Li_2WO_4 (1.6 g/cm^3)	microscope	355 - 5,600 µm	88 - 1,010 MP/kg DW
Lenaker et al. *(2021)*	USA	Lake Erie	n.r./n.r./n.r.	20	multicorer (10 cm/2 cm)	sieved, digestion	$ZnCl_2$ (1.6 g/cm^3)	microscope, subset FTIR (5.5%)	125 - >1,000 µm	>355 µm: 431 MP/kg DW 125–355 µm: 631 MP/kg DW
Jian et al. *(2020)*	China	Lake Poyang & tributaries	3,500 km^2/n.r./n.r.	42	Petersen dredge (50 × 50 cm/5 cm)	digestion	NaCl (1.2 g/cm^3)	microscope, subset FTIR	300 - 5,000 µm	41 ± 10 to 1,936 ± 121 MP/ kg DW
Bharat et al. (2021)	India	Veeranam Lake	19.4 km^2/mean 3.4 m/n.r.	28	van-Veen Grab (−/−)	digestion	$ZnCl_2$ (1.6 g/cm^3)	microscope, FTIR	300 - >2,000 µm	94 - 604 MP/kg

Red staining and spectroscopy can be more difficult, e.g., due to insufficient resolution of methods and due to altering of the surface and degradation of particles (Maes et al., 2017; Shim et al., 2017; Löder and Gerdts 2015).

4.3. Polymer composition

Fragments were predominantly composed of PE for both lakeshore and lakebed sediments, followed by PP and PA. Plastic production and plastic use is also predominated by PE and PP items (PlasticsEurope 2020) explaining high proportions of these polymers found in freshwater samples (e.g., Yang et al., 2021; Koelmans et al., 2019; LfU, 2019; Faure et al., 2015). PE and PP are low-density polymers, which rather float in freshwater bodies as their density is lower than water. These polymers may be transported within the water phase and washed onto beaches by currents and waves (Zhang et al., 2016). Additional fillers, e.g., minerals, and biofilms forming on the surface of microplastic particles as well as the formation of aggregates increase densities and particles may start to sink to the lakebed (Leiser et al., 2020; Bond et al., 2018; Chubarenko et al., 2016; Lobelle and Cunliffe 2011; Ye and Andrady 1991).

For fibers, PET was most common in Lake Tollense sediments. Textiles made of synthetic fibers are considered a major source for fibers in environmental samples in general and PET fibers in particular (Turner et al., 2019; Dris et al., 2018; Napper and Thompson 2016; Browne et al., 2011). Polymers, such as PA, PS, PET and PVC, have densities higher than water, easily tend to sink in freshwater bodies and accumulate in sediments (Lenaker et al., 2021; Koelmans et al., 2019; Schwarz et al., 2019; Browne et al., 2007). They are nearly equally found in lakeshore and lakebed sediments at Lake Tollense. PA was identified for fibers and fragments in both sediment compartments and often occurs as nylon fibers in textiles (Scopetani et al., 2019; Yuan et al., 2019). Furthermore, it is utilized for fishing lines, ropes, and packaging (Schwarz et al., 2019; McKeen 2017; GESAMP 2015; Andrady 2011) that might fragment and accumulate in Lake Tollense sediments.

4.4. Temporal variation of microplastic abundance

No meaningful temporal variations were detected for microplastic abundances in lakebed sediments at Lake Tollense. This result seems reasonable since microplastic accumulation within lakebed sediments is related to long time periods. Contrastingly, microplastic concentrations in lakeshore sediments differed significantly between sampling seasons. Higher abundances of fragments in March compared to September can be explained by variations in water levels. In March, water levels are higher resulting in smaller beach areas and consequently smaller microplastic accumulation zones. Furthermore, runoff events result in higher microplastic abundances in rivers and streams (Baldwin et al., 2016; Corcoran et al., 2015). Higher runoff within the catchment area in March, leading to higher water levels in the lake (Pegelportal MV 2020), may transport an increased number of microplastic fragments into Lake Tollense which are subsequently accumulated in lakeshore sediments. Another environmental factor influencing the transport of microplastics onto lakeshores can be wind itself and wind driven currents and waves (Bellasi et al., 2020; Helm 2020; Dris et al., 2018; Imhof et al. 2013, 2018; Vaughan et al., 2017; Zhang et al., 2016). Correlation coefficients showed that microplastic abundances increase with increasing wind velocities at Lake Tollense beaches. Mean wind velocities on sampling days were highest in March 2018 (DWD, 2020) when highest abundances for microplastic fragments were recorded. Least fragment concentrations were found in September 2018 when wind velocities were lowest. Consequently, wind and wind driven currents also play an important role for microplastic distribution at Lake Tollense beaches.

Fiber abundances were higher in lakeshore sediments in September; thus, other factors are more important. Microplastic fibers in freshwater environments mainly originate from textiles and fishing activities (Turner et al., 2019; Dris et al., 2018; Napper and Thompson 2016; GESAMP, 2015). Therefore, the input of fibers is enhanced in the tourist season from May to September when beaches are frequently visited for recreational purposes and fibers may be released from synthetic clothes. Additionally, fibers are more easily transported by wind and water and are consequently more dispersed within the study area (Bullard et al., 2021; Baldwin et al., 2020; Dris et al., 2016; Torre et al., 2016). Higher wind velocities were measured in March reducing the accumulation of fibers in sediments but rather transporting them elsewhere.

4.5. Spatial variation of microplastic abundances

Spatial variation in microplastic abundances was detected for lakeshore sediments. Former studies showed that microplastic concentrations in lakeshore sediments tend to be higher in areas with higher population density and anthropogenic activity (e.g., Lenaker et al., 2021; Baldwin et al., 2020; LfU, 2019; Faure et al., 2015). South beach at Lake Tollense shows highest microplastic pollution, however, it is located in a nature reserve and is characterized by the least direct human activity of investigated beaches. As for microplastics, highest anthropogenic litter abundances were found at South beach. The reduction of anthropogenic litter items at beaches with higher anthropogenic activity were related to regular beach cleanings (Hengstmann and Fischer 2020). As described before, macroplastic items tend to fragment over time forming several microplastic particles. Consequently, higher amounts of large plastic items at South beach will eventually lead to higher microplastic abundances. Correlation coefficients between macro- and microplastic concentrations showed a strong negative relation, except for South beach. The influence of temporal variation (March vs. September, see section 4.4) as well as long-term observations are of importance for the consideration of general decrease of microplastics with increasing macroplastic amounts presented in this study. The result further underlines that microplastic abundances do not exclusively depend on macroplastic abundances at lakeshores. South beach is in proximity to the inflow of the Nonnenbach, a major influent (0.57 m^3/s) of Lake Tollense (Fig. 1). Riverine inputs may impact and increase microplastic abundances as already stated for other lakes worldwide (Bharath et al., 2021; Lenaker et al., 2021; Hu et al., 2020; Zobkov et al., 2020; Vaughan et al., 2017; Ballent et al., 2016; Zhang et al., 2016; Corcoran et al., 2015; Faure et al., 2015). Rural runoff originating from agricultural areas within the catchment is combined in the Nonnenbach and can transport high loads of microplastics into Lake Tollense which are subsequently accumulated at South beach. Rural runoff is considered a major non-point source for microplastics in freshwaters (Dris et al., 2018) due to the use of sewage sludge and organic fertilizer containing microplastics (Bellasi et al., 2020; Weithmann et al., 2018; Wagner et al., 2014) or fragmenting plastic foils used as soil cover (Dris et al., 2018; Hussain and Hamid 2004).

North and East beach show low concentrations of microplastics despite their proximity to the city of Neubrandenburg and high anthropogenic use in the form of recreation, bathing and fishing. As stated before, the removement of macroplastic items by regular beach grooming may result in less fragmentation and consequently lower number of microplastics. Additionally, resuspension and erosion of material might play a key role at these beaches which are exposed in a southern and southwestern direction, respectively. A long-term dominant wind direction of southwest (DWD, 2020) increases wind and wave activity at North and East beach. North beach is characterized by a very flat beach area with low vegetation cover in the back of the beach. Therefore, microplastic particles, especially fibers, deposited on the sediment surface are easily carried away by wind (Bullard et al., 2021; Rezaei et al., 2019). Erosion patterns due to lateral runoff and change in water levels were recognized at East beach. Not only sediment is eroded, but also microplastic particles can be easily washed offshore.

The intertidal zone, which is in constant contact with water, showed significantly lower microplastic abundances since microplastic particles

are deposited and again resuspended by waves. Due to continuous motion in this zone, accumulation of particles is reduced which is further underlined by greater values for microplastic fragment length. In contrast, the high water line is a natural zone of accumulation where biogenic organic and plastic debris is deposited at high water levels. Microplastics are accumulated in this area until subsequent high water events. Similarly, the elevated area of beaches serves as an accumulation zone for longer timeframes. Increased microplastic abundances at the drift- and high water line or in elevated areas of lake beaches were reported by former studies as well (Dean et al., 2018; Imhof et al., 2018; Fischer et al., 2016). The difference in microplastic abundances between littoral sampling points verified in this study underlines the importance of sampling position within a beach area (Dris et al. 2015, 2018; Imhof et al., 2018). Studies exclusively sampling accumulation areas as the drift line (e.g., LfU, 2019; Faure et al., 2015; Imhof et al., 2013) likely overestimate microplastic abundances at lake shores.

As microplastics accumulated in lakebed sediments over long time periods, it seems reasonable that spatial difference is not strongly pronounced. Significant higher concentrations of fibers in the northern part of the lake are presumably resulting from an increased anthropogenic influence close to the city of Neubrandenburg in general and, in particular, increased fishing activities in this part of the lake. Abrasion of textiles and fishing materials constitute a major source for microplastic fibers in freshwater environments (see section 4.4). Considering fragments, tendentiously higher abundances were recognized in the central-south of the lake. Microplastics transported into Lake Tollense by the Nonnenbach might need calm accumulation conditions as well as time to settle to the lakebed (Vaughan et al., 2017). Therefore, microplastics that are not washed ashore at South beach are transported further into the lake by the fading flow of the tributary (Baldwin et al., 2020). Additionally, a general water movement towards the northern part of the lake is apparent in Lake Tollense since the only effluent is leaving the lake here. Therefore, particles are transported into deeper and northern areas of the lake, more distant from the inflow of the Nonnenbach. Enhanced settling conditions from south to north are underlined by increasing clay content of lakebed sediments in this direction.

4.6. Influence of pedologic characteristics

As LOI values correlated with fragment and fiber abundances for lakeshore sediments alone and in combination with lakebed sediments, the microplastic concentration is influenced by the percentage of organic matter in sediments. For both sediment compartments, this relation was verified by former studies (Haave et al., 2019; Turner et al., 2019; Ballent et al., 2016; Fischer et al., 2016; Strand et al., 2013) while other studies could not confirm this behavior (Hengstmann et al., 2018; Renzi et al., 2018; Vermaire et al., 2017). No correlation was verified between the two variables for lakebed sediments of Lake Tollense alone. However, the latter may be a result of small sample numbers and similar content of organic matter within lakebed samples (SD = 0.56%, mean = 8.69%). The relation between microplastics and organic matter in lakeshore sediments is underlined when considering the spatial variation of both variables. Organic content was significantly higher at South and West beach compared to North and East beach which is in accordance with microplastic results. Significantly lower organic content was found in the intertidal zone consistent with lower microplastic abundances. Consequently, accumulation areas for organic matter also serve as accumulation areas for microplastics.

A similar relation is expected for small grain sizes (silt/clay) and microplastics since calm sedimentation zones are necessary for the accumulation of fine sediment particles as well. Correlations between silt and/or clay and microplastic abundances were already shown by other marine and freshwater studies (Zobkov et al., 2020; Enders et al., 2019; Haave et al., 2019; Fischer et al., 2016; Strand et al., 2013). However, this hypothesis could not be verified for lake sediments at Lake Tollense. Similarly, it was not confirmed by other studies (Renzi et al., 2018; Vermaire et al., 2017; Alomar et al., 2016; Browne et al., 2010). Again, the similarity between samples as well as small fractions of clay within the sediments in general might curtain a relation. Further research, investigating sediments of varying grain sizes at Lake Tollense is needed to gain more information on the influence of grain size distribution on microplastic abundances.

5. Conclusion

Ubiquitous and high abundances of microplastics in shore and bed sediments analyzed over a two-year period were found at Lake Tollense. The study highlighted the retention capabilities of lake sediments and with this the storage characteristics lakes provide for microplastic particles on transport pathways from land-based sources to marine environments, functioning as miniature oceans. These results underline the importance of investigating lake sediments concerning their plastic pollution levels. Human activity was verified as a source for microplastics emphasizing the need to implement further, more strict measures regarding anthropogenic plastic use in diverse fields. Besides human activity, external factors, such as wind and sediment characteristics were verified as potential influences on the microplastic abundance and distribution in lake sediments. Organic matter accumulation zones coincide with microplastic accumulation zones. The lake's hydrology (e.g., varying water levels) and the inflow of tributaries further impact microplastic occurrence and distribution. Tributaries import microplastics into the lake which can be subsequently washed ashore on adjacent shorelines. Therefore, samples taken at shores close to inflowing tributaries are more polluted by microplastics, especially when tributaries comprise large catchment areas with high utilization rates. Even though research on the plastic pollution of freshwaters has increased lately and this study further contributes to the data availability concerning microplastic concentrations in lake sediments, knowledge gaps still exist. These must be closed by further studies on the behavior of microplastics within lakes, their transport to and from freshwater bodies, and their impacts on organisms living close to or in these aquatic environments.

Authors contribution

Elena Hengstmann: Conceptualization, Investigation, Methodology, Formal analysis, Writing – original draft, Visualization. Esther Weil: Software, Methodology. Paul Christian Wallbott: Software, Methodology. Matthias Tamminga: Conceptualization, Investigation, Writing – review & editing. Elke Kerstin Fischer: Conceptualization, Resources, Writing – review & editing, Supervision.

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Declaration of competing interest

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

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Appendix A. Supplementary data

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Supplementary Material

Microplastics in lakeshore and lakebed sediments – External influences and temporal and spatial variabilities of concentrations

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Text SM1: Correction of September 2017 microplastic concentrations.

Microplastic concentrations for lakeshore samples from September 2017 were scaled to be comparable to following sampling concentrations since the identification and counting process differed for these samples. In a first step, regression data according to Hengstmann and Fischer (2019) for the comparison between UV light photobox and microscope images was used to account for resolution differences between the two methods. The second step accounted for the difference in counting. Only particles matching the size-class of subsamples resulting from wet-sieving were counted in UV light photobox images whereas all particles ≥ 63 µm were counted in the microscope images. A size class dependent comparison between total microplastics and the subset of microplastics fitting the size class of subsamples resulting from wet sieving was evaluated based on lakeshore data from sampling campaigns March 2018 to March 2019. Regression equations were again considered and utilized to extrapolate September 2017 results. Combining both steps, September 2017 lakeshore data was transferred and microplastic concentrations were in a similar range as results of subsequent sampling campaigns.

Figure SM1: Stained microplastic particles identified by the fluorescence microscope and associated µ-Raman spectroscopy image and spectra. Black scale bars represent 500 µm.

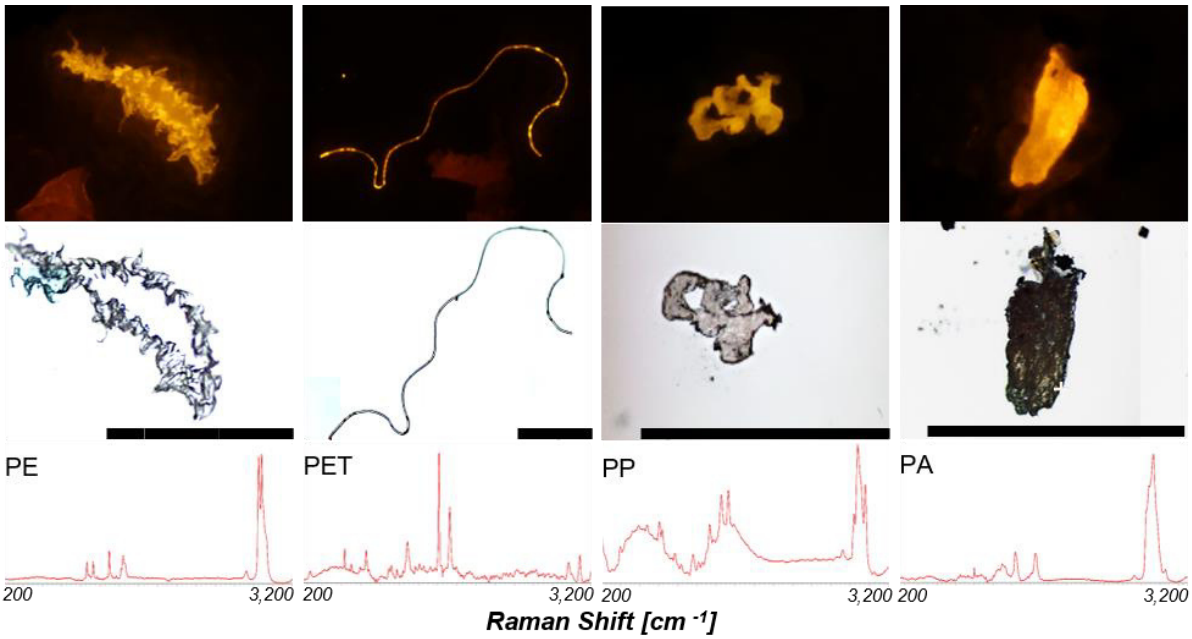
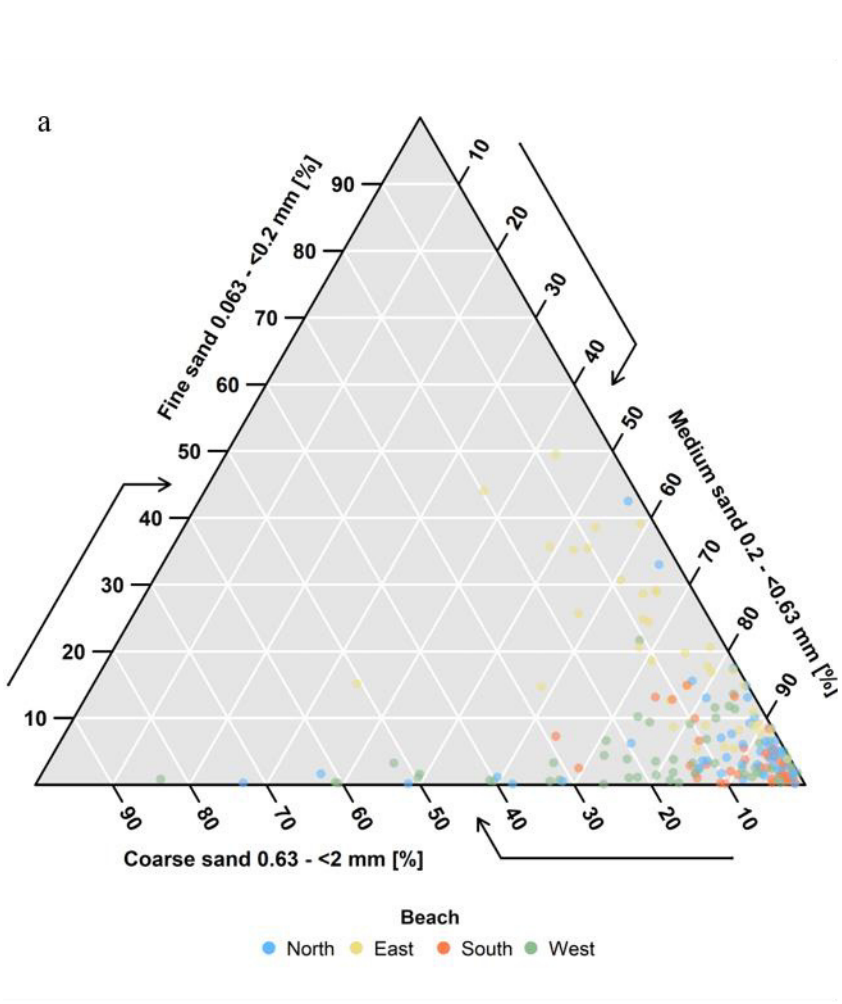


Figure SM2: a) Sand size fractions for lakeshore sediments and b) grain size fractions for lakebed sediments at Lake Tollense.



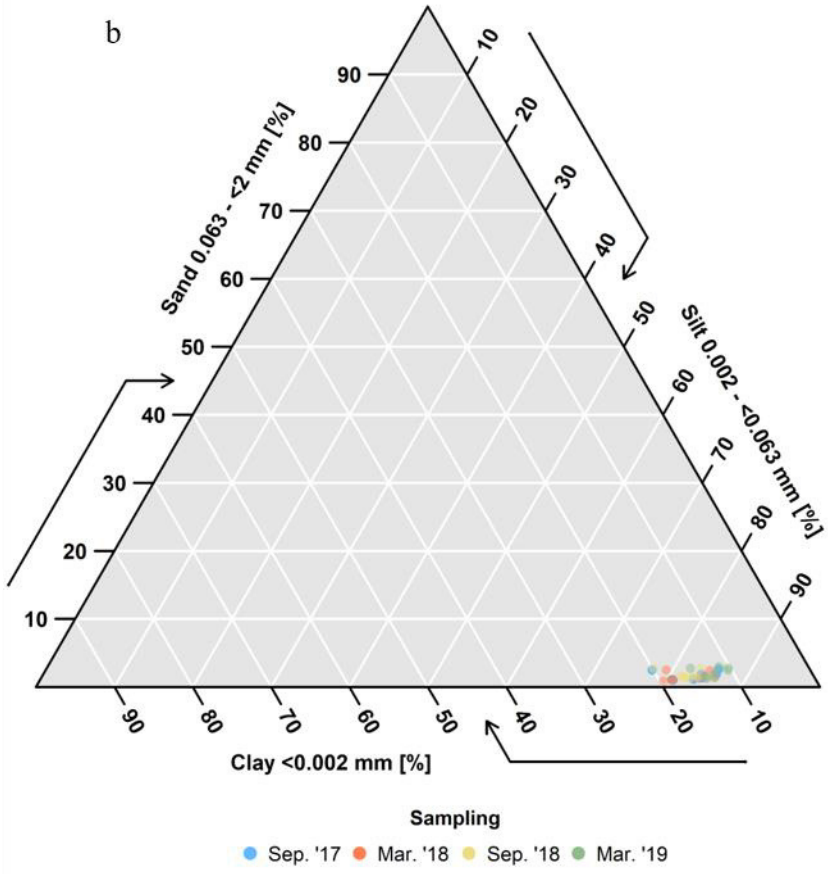


Table SM1: Microplastic (MP) abundances in lakeshore sediments at Lake Tollense presented in different units and including pedologic information. (hwl = high water line)

Sample information				*MP per kg DW*			*MP per m²*			*MP per L*			*Pedologic characteristics*			*Grain sizes*			
Sampling	***Date***	***Beach***	***Littoral Position***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Bulk density (g/cm³)***	***Water content (%)***	***Organic matter (%)***	***Coarse sand (%)***	***Medium sand (%)***	***Fine sand (%)***	***Silt & clay (%)***
Sep. '17	11.09.17	South	intertidal	852	297	1,150	21,573	7,446	29,019	1,454	492	1,947	1.75	0.03	0.11	10.21	88.98	0.15	0.66
			hwl	466	349	815	12,632	7,878	20,510	787	573	1,360	1.65	0.04	0.17	2.25	97.16	0.40	0.19
			elevated	1,040	561	1,601	29,171	15,458	44,628	1,681	934	2,615	1.68	0.03	0.12	1.10	97.75	1.00	0.14
			intertidal	577	320	897	17,109	9,507	26,616	967	532	1,498	1.75	0.03	0.10	5.91	89.96	3.70	0.42
			hwl	990	747	1,737	29,109	22,023	51,132	1,597	1,213	2,810	1.61	0.03	0.12	4.51	92.79	2.65	0.05
			elevated	1,007	725	1,732	29,129	20,816	49,945	1,602	1,162	2,764	1.58	0.04	0.13	2.55	92.86	4.21	0.38
			intertidal	1,127	304	1,431	31,348	8,206	39,555	1,941	501	2,442	1.80	0.04	0.10	11.32	85.72	3.00	0.00
			hwl	741	352	1,094	20,532	9,441	29,973	1,252	594	1,846	1.77	0.03	0.15	0.60	96.97	2.20	0.22
			elevated	601	1,084	1,685	16,367	28,636	45,003	976	1,710	2,686	1.56	0.04	0.16	3.86	94.99	0.85	0.30
			intertidal	642	615	1,257	17,094	15,899	32,993	1,057	1,029	2,086	1.69	0.04	0.10	9.31	80.11	9.81	0.76
			hwl	815	763	1,578	20,787	18,346	39,133	1,320	1,205	2,525	1.56	0.04	0.12	1.70	92.69	5.16	0.45
			intertidal	760	575	1,335	20,532	15,038	35,570	1,353	1,040	2,393	1.93	0.05	0.14	28.60	63.72	7.21	0.46
			hwl	1,397	471	1,868	25,275	9,012	34,287	1,909	670	2,579	1.28	0.20	1.58	1.32	94.48	5.40	0.00
			elevated	1,786	1,190	2,976	49,201	32,933	82,135	2,647	1,830	4,478	1.50	0.09	0.39	10.40	82.85	6.58	0.17
	12.09.17	East	intertidal	715	729	1,445	20,566	21,822	42,388	1,182	1,216	2,399	1.70	0.05	0.10	7.61	86.43	5.61	0.35
			hwl	614	513	1,127	15,749	12,051	27,800	1,009	835	1,844	1.60	0.07	0.16	11.17	75.57	12.48	0.78
			intertidal	742	311	1,053	21,295	8,477	29,772	1,241	512	1,753	1.78	0.06	0.13	3.76	78.10	17.63	0.51
			hwl	832	646	1,478	21,998	16,496	38,494	1,346	1,033	2,379	1.59	0.07	0.13	7.82	76.95	14.63	0.60
			intertidal	466	263	729	12,804	6,906	19,711	788	424	1,212	1.73	0.06	0.09	8.51	65.95	24.59	0.95
			hwl	921	544	1,465	23,964	13,739	37,703	1,514	897	2,410	1.68	0.08	0.17	8.02	66.62	24.06	1.29
			elevated	698	721	1,419	17,431	15,992	33,423	1,157	1,205	2,362	1.68	0.11	0.27	10.34	69.07	18.12	2.47
			intertidal	674	390	1,064	18,974	10,785	29,759	1,111	630	1,741	1.63	0.07	0.11	7.81	52.85	38.02	1.32
			hwl	490	502	993	13,457	13,025	26,482	825	788	1,614	1.52	0.08	0.11	8.47	59.96	30.26	1.32
			intertidal	513	848	1,362	13,980	21,280	35,260	871	1,421	2,292	1.68	0.07	0.11	16.48	57.45	25.49	0.58
			hwl	446	282	728	12,295	7,147	19,442	758	451	1,208	1.60	0.07	0.16	4.61	64.15	28.27	2.97
	13.09.17	West	intertidal	644	450	1,094	18,274	12,817	31,091	1,039	709	1,748	1.54	0.07	0.24	40.33	58.28	0.65	0.74
			hwl	760	270	1,030	21,599	7,303	28,901	1,237	430	1,667	1.59	0.07	0.47	17.04	81.54	0.65	0.76
			intertidal	969	546	1,515	27,874	15,512	43,386	1,562	901	2,463	1.64	0.05	0.20	2.76	90.53	6.17	0.54
			hwl	997	461	1,458	25,594	11,603	37,197	1,565	722	2,286	1.55	0.13	0.75	16.19	81.98	1.82	0.02
			intertidal	779	500	1,280	18,617	10,975	29,593	1,294	828	2,122	1.69	0.09	0.16	0.25	95.44	4.01	0.30
			hwl	1,017	978	1,995	26,218	24,714	50,932	1,665	1,599	3,264	1.65	0.12	0.55	30.86	65.39	3.27	0.48
			elevated	681	568	1,249	15,292	10,174	25,467	1,044	805	1,849	1.30	0.37	1.95	51.09	44.38	3.17	1.36
			intertidal	744	361	1,104	19,839	9,305	29,144	1,236	586	1,822	1.69	0.09	0.30	23.74	71.28	4.37	0.61
			hwl	794	452	1,246	19,373	10,310	29,683	1,236	675	1,911	1.41	0.14	0.68	9.28	82.97	6.70	1.05
			elevated	976	421	1,397	23,161	9,849	33,010	1,515	651	2,167	1.49	0.08	0.24	18.56	79.80	1.40	0.23
			intertidal	734	314	1,048	21,736	8,833	30,569	1,170	502	1,672	1.54	0.05	0.14	0.20	94.13	4.96	0.71

Table SM1 continued

Sample information				*MP per kg DW*			*MP per m²*			*MP per L*			*Pedologic characteristics*			*Grain sizes*			
Sampling	***Date***	***Beach***	***Littoral Position***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Bulk density (g/cm³)***	***Water content (%)***	***Organic matter (%)***	***Coarse sand (%)***	***Medium sand (%)***	***Fine sand (%)***	***Silt & clay (%)***
Sep. '17	13.09.17	West	hwl	878	648	1,526	22,829	16,310	39,140	1,384	977	2,360	1.47	0.11	0.46	10.01	85.98	3.17	0.84
			depression	1,232	550	1,782	31,675	13,906	45,581	1,648	737	2,384	1.18	0.11	0.40	17.54	78.35	3.47	0.64
			hwl	818	324	1,141	20,009	7,859	27,868	1,144	460	1,604	1.08	0.12	0.46	40.13	59.24	0.55	0.07
			elevated	706	373	1,079	18,013	9,037	27,050	1,140	584	1,724	1.51	0.06	0.15	21.14	74.91	3.81	0.14
	14.09.17		intertidal	585	299	884	15,452	7,481	22,933	970	484	1,454	1.67	0.05	0.15	1.35	91.88	6.51	0.25
			hwl	678	302	980	16,621	7,208	23,829	1,083	468	1,552	1.45	0.06	0.20	2.36	95.40	1.55	0.69
			elevated	917	394	1,310	20,056	8,534	28,589	1,223	520	1,744	1.01	0.09	0.35	32.59	66.09	0.50	0.82
			intertidal	631	460	1,091	16,586	11,142	27,728	923	573	1,496	0.94	0.06	0.16	10.27	79.93	9.07	0.73
			hwl	612	306	918	17,763	8,639	26,401	1,007	486	1,493	1.60	0.06	0.20	2.46	83.31	13.48	0.75
			hwl	767	499	1,266	20,366	13,129	33,495	1,205	762	1,967	1.48	0.09	0.32	5.17	91.58	2.46	0.79
			elevated	808	498	1,306	20,297	11,827	32,124	1,285	775	2,060	1.52	0.07	0.26	4.72	92.95	1.71	0.63
	15.09.17	North	intertidal	921	271	1,192	24,192	7,005	31,198	1,484	437	1,921	1.63	0.04	0.10	1.85	91.02	6.46	0.67
			hwl	548	381	929	14,494	9,212	23,706	848	495	1,343	0.92	0.04	0.12	0.75	93.74	5.01	0.50
			elevated	649	351	1,000	17,454	9,106	26,560	983	503	1,486	1.16	0.05	0.16	10.87	85.18	3.56	0.39
			intertidal	571	343	914	16,643	10,067	26,710	960	557	1,517	1.65	0.04	0.12	1.45	93.10	5.01	0.44
			hwl	931	681	1,612	21,054	14,068	35,121	1,513	1,159	2,672	1.67	0.04	0.14	0.15	90.91	8.41	0.52
			elevated	668	321	989	16,347	7,518	23,865	1,077	510	1,586	1.54	0.03	0.12	0.50	95.40	3.61	0.49
			intertidal	715	327	1,041	19,603	8,676	28,279	1,200	548	1,748	1.76	0.04	0.14	2.35	94.47	2.15	1.02
			hwl	647	555	1,203	17,883	15,380	33,263	1,064	917	1,981	1.64	0.06	0.09	0.60	92.24	6.51	0.65
			elevated	876	452	1,328	24,232	12,200	36,433	1,305	657	1,962	1.33	0.05	0.17	7.32	84.88	6.97	0.84
	17.09.17		intertidal	539	284	823	14,752	7,369	22,121	921	469	1,390	1.79	0.04	0.13	5.66	85.95	7.51	0.88
			hwl	1,085	487	1,571	30,960	13,875	44,835	1,598	732	2,330	1.42	0.04	0.14	0.20	84.60	14.73	0.47
			elevated	1,189	560	1,749	28,399	13,049	41,448	1,797	844	2,641	1.44	0.06	0.22	2.51	87.19	9.13	1.18
			intertidal	759	359	1,118	22,804	10,537	33,341	1,233	573	1,805	1.59	0.04	0.13	7.56	90.85	0.80	0.78
			hwl	761	473	1,234	20,343	12,118	32,461	1,263	786	2,049	1.70	0.04	0.09	1.45	85.87	9.76	2.92
			elevated	1,195	779	1,974	27,308	16,936	44,244	1,684	1,091	2,775	1.30	0.09	0.52	0.91	85.92	13.03	0.14

Table SM1 continued

Sample information				*MP per kg DW*			*MP per m²*			*MP per L*			*Pedologic characteristics*			*Grain sizes*			
Sampling	***Date***	***Beach***	***Littoral Position***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Bulk density (g/cm³)***	***Water content (%)***	***Organic matter (%)***	***Coarse sand (%)***	***Medium sand (%)***	***Fine sand (%)***	***Silt & clay (%)***
Mar. '18	15.03.18	South	intertidal	1,415	491	1,906	43,076	14,953	58,029	43,076	14,953	58,029	1.70	0.07	0.16	2.05	96.52	0.95	0.47
			hwl	3,624	673	4,297	103,215	19,174	122,389	103,215	19,174	122,389	1.43	0.14	0.44	4.07	95.09	0.30	0.54
			intertidal	2,522	436	2,958	39,739	6,864	46,603	39,739	6,864	46,603	1.66	0.08	0.26	10.84	88.62	0.20	0.34
			hwl	1,703	613	2,316	28,253	10,176	38,430	28,253	10,176	38,430	1.83	0.20	0.64	1.56	94.50	3.63	0.31
			intertidal	2,104	448	2,552	55,915	11,902	67,817	55,915	11,902	67,817	1.54	0.07	0.13	1.20	97.38	0.90	0.51
			hwl	1,956	300	2,256	65,888	10,103	75,991	65,888	10,103	75,991	1.86	0.07	0.15	2.00	96.45	1.30	0.24
			elevated	1,434	275	1,709	36,315	6,954	43,268	36,315	6,954	43,268	1.60	0.08	0.29	2.81	95.65	1.10	0.44
			intertidal	1,162	293	1,455	28,586	7,212	35,799	28,586	7,212	35,799	1.58	0.07	0.12	13.47	83.60	2.86	0.07
			intertidal	244	181	425	9,407	6,953	16,360	9,407	6,953	16,360	1.57	0.07	0.10	7.96	90.49	1.55	0.00
			hwl	895	298	1,193	29,304	9,738	39,043	29,304	9,738	39,043	1.37	0.03	0.11	2.00	95.78	1.65	0.56
			elevated	1,564	427	1,991	48,558	13,259	61,817	48,558	13,259	61,817	1.68	0.06	0.12	0.60	95.32	3.41	0.67
	16.03.18	West	intertidal	1,452	300	1,752	35,432	7,328	42,761	35,432	7,328	42,761	1.75	0.06	0.16	5.21	92.69	1.80	0.29
			hwl	1,713	559	2,272	42,160	13,747	55,906	42,160	13,747	55,906	1.84	0.06	0.20	22.30	70.17	6.52	1.01
			intertidal	1,548	339	1,887	38,882	8,509	47,392	38,882	8,509	47,392	1.60	0.08	0.16	0.45	96.26	2.71	0.58
			hwl	1,769	637	2,406	57,849	20,834	78,683	57,849	20,834	78,683	1.91	0.12	0.26	10.59	67.17	21.47	0.77
			intertidal	1,403	311	1,714	44,440	9,857	54,297	44,440	9,857	54,297	1.73	0.07	0.15	3.01	96.35	0.30	0.34
			hwl	1,435	320	1,755	26,229	5,848	32,077	26,229	5,848	32,077	1.63	0.14	0.19	7.02	91.03	1.50	0.44
			elevated	2,609	402	3,012	71,939	11,095	83,034	71,939	11,095	83,034	1.57	0.22	0.60	21.07	77.31	1.11	0.51
			intertidal	971	360	1,331	29,119	10,784	39,903	29,119	10,784	39,903	1.72	0.14	0.12	0.45	95.88	3.71	0.00
			hwl	2,071	349	2,420	49,161	8,290	57,451	49,161	8,290	57,451	1.70	0.16	0.18	12.59	84.51	2.26	0.64
			elevated	2,385	379	2,763	63,300	10,050	73,350	63,300	10,050	73,350	1.57	0.11	0.21	6.92	90.34	2.51	0.23
			intertidal	226	30	256	7,270	974	8,244	7,270	974	8,244	1.62	0.11	0.15	22.20	76.28	1.00	0.52
			hwl	1,279	367	1,646	32,421	9,291	41,712	32,421	9,291	41,712	1.67	0.07	0.13	6.31	92.86	1.15	0.00
			elevated	1,461	512	1,973	49,977	17,501	67,478	49,977	17,501	67,478	1.72	0.05	0.11	2.05	96.70	0.75	0.50
			intertidal	272	165	437	8,641	5,257	13,898	8,641	5,257	13,898	1.67	0.06	0.15	16.13	83.02	0.25	0.60
			intertidal	559	104	663	16,866	3,132	19,998	16,866	3,132	19,998	1.76	0.08	0.15	25.95	73.35	0.10	0.59
			intertidal	1,319	201	1,520	28,643	4,361	33,004	28,643	4,361	33,004	1.48	0.08	0.16	12.93	83.73	3.21	0.14
	17.03.18	North	intertidal	551	145	696	14,771	3,895	18,666	14,771	3,895	18,666	1.74	0.12	0.12	72.20	26.66	0.25	0.89
			hwl	1,349	385	1,734	35,173	10,038	45,212	35,173	10,038	45,212	1.58	0.15	0.12	39.25	59.24	1.10	0.41
			elevated	1,140	248	1,388	30,867	6,701	37,568	30,867	6,701	37,568	1.66	0.06	0.10	4.66	92.27	2.95	0.12
			intertidal	2,256	380	2,636	70,847	11,938	82,785	70,847	11,938	82,785	1.73	0.08	0.14	61.77	36.12	1.60	0.51
			hwl	1,356	119	1,474	37,525	3,284	40,809	37,525	3,284	40,809	1.72	0.10	0.20	51.19	48.18	0.15	0.48
			elevated	1,489	238	1,727	33,424	5,339	38,763	33,424	5,339	38,763	1.50	0.12	0.10	1.75	95.44	2.50	0.30
			intertidal	905	149	1,054	28,280	4,651	32,930	28,280	4,651	32,930	1.72	0.05	0.10	19.22	73.39	6.13	1.26
			hwl	1,312	104	1,416	35,744	2,841	38,585	35,744	2,841	38,585	1.65	0.08	0.16	10.02	88.12	1.65	0.20
			elevated	769	109	879	19,493	2,772	22,265	19,493	2,772	22,265	1.56	0.27	0.12	3.26	91.40	4.97	0.37

Table SM1 continued

Sample information				*MP per kg DW*			*MP per m²*			*MP per L*			*Pedologic characteristics*			*Grain sizes*			
Sampling	***Date***	***Beach***	***Littoral Position***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Bulk density (g/cm³)***	***Water content (%)***	***Organic matter (%)***	***Coarse sand (%)***	***Medium sand (%)***	***Fine sand (%)***	***Silt & clay (%)***
Mar. '18	17.03.18	North	intertidal	164	183	346	4,375	4,880	9,255	4,375	4,880	9,255	1.75	0.21	0.08	1.90	92.24	5.66	0.19
			hwl	1,117	322	1,438	35,118	10,116	45,233	35,118	10,116	45,233	1.73	0.07	0.13	0.60	95.57	3.71	0.12
			elevated	3,051	558	3,609	64,200	11,747	75,947	64,200	11,747	75,947	1.61	0.13	0.38	0.70	95.13	3.77	0.40
			intertidal	407	62	470	14,345	2,194	16,538	14,345	2,194	16,538	1.78	0.18	0.09	6.62	90.26	3.06	0.07
			hwl	727	136	863	22,971	4,294	27,265	22,971	4,294	27,265	1.77	0.11	0.14	0.70	94.36	4.71	0.23
			elevated	974	189	1,164	22,928	4,457	27,385	22,928	4,457	27,385	1.57	0.22	0.50	1.16	96.06	2.82	0.00
	18.03.18	East	intertidal	1,942	176	2,117	66,044	5,975	72,019	66,044	5,975	72,019	1.85	0.11	0.09	6.51	87.50	5.36	0.63
			hwl	1,849	252	2,100	59,746	8,131	67,877	59,746	8,131	67,877	1.72	0.10	0.12	1.40	89.03	9.02	0.54
			elevated	1,094	233	1,328	35,093	7,482	42,575	35,093	7,482	42,575	1.67	0.10	0.11	1.25	87.31	10.77	0.67
			intertidal	1,420	330	1,750	46,803	10,887	57,690	46,803	10,887	57,690	1.76	0.25	0.07	3.66	78.92	16.80	0.62
			hwl	1,555	407	1,962	33,618	8,806	42,424	33,618	8,806	42,424	1.65	0.11	0.09	1.85	90.10	7.71	0.34
			elevated	1,846	282	2,128	47,177	7,199	54,376	47,177	7,199	54,376	1.69	0.14	0.10	1.95	76.10	20.29	1.65
			intertidal	492	407	898	18,475	15,276	33,751	18,475	15,276	33,751	1.83	0.13	0.15	11.33	82.68	5.31	0.68
			intertidal	28	283	311	951	9,507	10,458	951	9,507	10,458	1.73	0.10	0.20	11.08	67.40	20.41	1.10
			intertidal	555	376	931	20,619	13,967	34,585	20,619	13,967	34,585	1.83	0.09	0.19	26.62	57.86	14.54	0.97

Table SM1 continued

Sample information				*MP per kg DW*			*MP per m²*			*MP per L*			*Pedologic characteristics*			*Grain sizes*			
Sampling	***Date***	***Beach***	***Littoral Position***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***	***Bulk density (g/cm³)***	***Water content (%)***	***Organic matter (%)***	***Coarse sand (%)***	***Medium sand (%)***	***Fine sand (%)***	***Silt & clay (%)***
Sep. '18	04.09.18	South	intertidal	2,457	977	3,434	59,306	23,592	82,898	59,306	23,592	82,898	1.38	0.09	0.66	13.45	85.24	0.96	0.35
			hwl	2,447	888	3,335	78,119	28,340	106,459	78,119	28,340	106,459	1.58	0.14	1.04	5.16	89.10	5.51	0.23
			elevated	1,460	661	2,121	46,160	20,909	67,069	46,160	20,909	67,069	1.61	0.09	0.46	1.31	95.53	3.07	0.10
			intertidal	2,025	443	2,467	52,892	11,570	64,462	52,892	11,570	64,462	1.61	0.08	0.49	10.81	76.15	12.72	0.33
			hwl	1,989	640	2,629	55,554	17,889	73,443	55,554	17,889	73,443	1.59	0.07	0.34	0.45	90.77	8.33	0.44
			elevated	1,275	596	1,870	23,097	10,792	33,889	23,097	10,792	33,889	1.48	0.20	1.15	2.53	84.39	13.23	0.00
			intertidal	1,563	599	2,162	49,922	19,132	69,054	49,922	19,132	69,054	1.75	0.08	0.57	1.86	93.65	4.13	0.36
			hwl	1,866	639	2,505	46,980	16,079	63,059	46,980	16,079	63,059	1.69	0.06	0.34	1.15	98.50	0.20	0.15
			elevated	1,023	464	1,487	19,143	8,674	27,817	19,143	8,674	27,817	1.63	0.11	0.74	8.67	89.36	2.02	0.00
	05.09.18	West	intertidal	795	888	1,683	19,094	21,327	40,422	19,094	21,327	40,422	1.64	0.10	0.43	6.53	82.79	9.90	0.77
			hwl	945	731	1,675	18,960	14,659	33,620	18,960	14,659	33,620	1.76	0.10	0.60	5.84	81.87	11.43	0.86
			elevated	908	881	1,789	23,818	23,121	46,939	23,818	23,121	46,939	1.67	0.13	0.61	3.42	84.74	11.28	0.56
			intertidal	258	196	453	6,945	5,268	12,213	6,945	5,268	12,213	1.66	0.15	0.50	49.65	49.19	1.06	0.10
			hwl	313	249	562	7,567	6,014	13,581	7,567	6,014	13,581	1.65	0.11	0.57	3.93	83.77	11.69	0.62
			elevated	540	293	833	13,429	7,298	20,727	13,429	7,298	20,727	1.67	0.09	0.42	15.13	80.81	3.77	0.29
			intertidal	634	321	955	16,372	8,293	24,665	16,372	8,293	24,665	1.66	0.11	0.48	15.38	74.81	9.35	0.46
			hwl	757	189	946	21,706	5,406	27,112	21,706	5,406	27,112	1.61	0.09	0.49	16.55	73.02	10.16	0.27
			elevated	337	298	634	10,192	9,019	19,211	10,192	9,019	19,211	1.68	0.10	0.41	31.51	67.59	0.70	0.19
	06.09.18	North	intertidal	337	61	399	8,657	1,570	10,227	8,657	1,570	10,227	1.81	0.06	0.31	5.02	88.23	5.97	0.78
			hwl	276	238	514	7,606	6,566	14,172	7,606	6,566	14,172	1.73	0.07	0.40	2.46	64.22	32.79	0.54
			elevated	209	389	597	5,638	10,507	16,144	5,638	10,507	16,144	1.50	0.09	0.41	1.71	55.40	42.14	0.75
			intertidal	380	170	550	9,540	4,285	13,826	9,540	4,285	13,826	1.79	0.05	0.34	11.44	84.48	3.56	0.52
			hwl	383	167	550	9,621	4,192	13,813	9,621	4,192	13,813	1.71	0.08	0.36	0.55	81.41	17.32	0.72
			elevated	107	52	160	2,856	1,384	4,240	2,856	1,384	4,240	1.61	0.13	0.33	6.78	76.35	15.27	1.59
			intertidal	32	53	84	818	1,347	2,165	818	1,347	2,165	1.84	0.07	0.29	8.07	87.31	4.06	0.55
			hwl	44	349	393	1,383	10,951	12,334	1,383	10,951	12,334	1.65	0.08	0.40	2.71	94.35	2.76	0.17
			elevated	56	161	217	1,751	5,002	6,753	1,751	5,002	6,753	1.73	0.07	0.29	6.22	79.79	12.85	1.14
	07.09.18	East	intertidal	29	185	213	751	4,832	5,583	751	4,832	5,583	1.86	0.08	0.31	6.63	64.09	28.33	0.96
			hwl	232	182	413	5,646	4,427	10,073	5,646	4,427	10,073	1.68	0.08	0.36	1.86	58.53	38.75	0.86
			elevated	209	569	778	5,122	13,955	19,077	5,122	13,955	19,077	1.73	0.10	0.38	4.87	65.79	28.63	0.72
			intertidal	61	353	414	1,705	9,917	11,622	1,705	9,917	11,622	1.85	0.14	0.47	52.45	35.54	15.65	0.00
			hwl	198	361	559	4,531	8,248	12,779	4,531	8,248	12,779	1.94	0.11	0.48	15.19	48.43	35.16	1.22
			elevated	839	580	1,419	24,116	16,669	40,784	24,116	16,669	40,784	1.66	0.15	0.40	10.31	53.24	34.89	1.57
			intertidal	136	278	415	3,090	6,294	9,384	3,090	6,294	9,384	1.93	0.13	0.33	19.35	35.63	43.17	1.85
			hwl	175	339	514	4,050	7,840	11,890	4,050	7,840	11,890	1.81	0.12	0.35	7.53	42.48	48.91	1.08
			elevated	58	460	518	1,712	13,562	15,273	1,712	13,562	15,273	1.68	0.16	0.47	12.27	51.70	34.70	1.33

Table SM1 continued

Sample information				*MP per kg DW*			*MP per m²*			*MP per L*			*Pedologic characteristics*			*Grain sizes*			
Sampling	*Date*	*Beach*	*Littoral Position*	*Fragments*	*Fibers*	*Particles*	*Fragments*	*Fibers*	*Particles*	*Fragments*	*Fibers*	*Particles*	*Bulk density (g/cm³)*	*Water content (%)*	*Organic matter (%)*	*Coarse sand (%)*	*Medium sand (%)*	*Fine sand (%)*	*Silt & clay (%)*
Mar. '19	19.03.19	South	intertidal	1,927	248	2,175	57,521	7,399	64,920	57,521	7,399	64,920	1.71	0.02	0.07	1.85	96.79	1.05	0.30
			hwl	1,973	122	2,094	53,286	3,293	56,579	53,286	3,293	56,579	1.42	0.03	0.19	1.60	97.36	0.65	0.38
			elevated	2,614	262	2,877	44,812	4,495	49,307	44,812	4,495	49,307	1.33	0.05	0.34	2.71	92.05	4.62	0.62
			intertidal	1,532	265	1,797	35,154	6,075	41,229	35,154	6,075	41,229	1.67	0.02	0.07	13.27	84.57	1.70	0.46
			hwl	2,409	254	2,663	49,692	5,242	54,933	49,692	5,242	54,933	1.21	0.05	0.37	3.66	93.65	2.51	0.17
			elevated	4,010	1,656	5,666	60,054	24,791	84,845	60,054	24,791	84,845	1.12	0.32	2.06	12.55	72.32	12.81	2.32
			intertidal	320	105	424	8,736	2,856	11,591	8,736	2,856	11,591	1.67	0.03	0.12	28.04	69.15	2.45	0.36
			hwl	1,895	377	2,272	43,537	8,669	52,206	43,537	8,669	52,206	1.14	0.03	0.18	1.60	96.75	1.20	0.44
			elevated	1,744	1,146	2,890	40,921	26,893	67,814	40,921	26,893	67,814	1.36	0.14	0.95	7.78	75.83	14.61	1.78
	20.03.19	West	intertidal	1,109	174	1,283	29,312	4,587	33,899	29,312	4,587	33,899	1.63	0.04	0.15	60.53	38.65	0.35	0.46
			hwl	2,652	185	2,837	73,628	5,149	78,778	73,628	5,149	78,778	1.65	0.02	0.14	0.50	96.75	2.45	0.30
			elevated	1,543	145	1,687	45,748	4,297	50,045	45,748	4,297	50,045	1.61	0.02	0.12	0.05	97.60	1.75	0.60
			intertidal	399	235	634	9,690	5,701	15,390	9,690	5,701	15,390	1.69	0.05	0.22	82.48	15.74	0.80	0.98
			hwl	1,180	267	1,447	29,794	6,745	36,539	29,794	6,745	36,539	1.66	0.07	0.54	13.28	84.21	1.66	0.85
			elevated	795	294	1,089	20,119	7,446	27,565	20,119	7,446	27,565	1.59	0.03	0.17	3.01	95.19	0.80	1.00
			intertidal	1,009	85	1,095	28,460	2,407	30,866	28,460	2,407	30,866	1.62	0.04	0.16	48.95	48.85	1.55	0.65
			hwl	2,152	236	2,388	50,929	5,596	56,526	50,929	5,596	56,526	1.34	0.05	0.35	60.29	39.11	0.20	0.40
			elevated	1,735	98	1,833	55,105	3,124	58,229	55,105	3,124	58,229	1.05	0.03	0.21	3.11	93.98	2.35	0.56
	21.03.19	North	intertidal	1,325	216	1,541	38,495	6,290	44,785	38,495	6,290	44,785	1.66	0.03	0.16	31.01	67.88	0.50	0.61
			hwl	1,771	183	1,954	51,181	5,298	56,479	51,181	5,298	56,479	1.61	0.03	0.14	2.51	93.00	3.86	0.64
			elevated	1,375	174	1,549	41,590	5,271	46,861	41,590	5,271	46,861	1.25	0.02	0.09	1.25	97.95	0.15	0.65
			intertidal	1,130	215	1,344	33,556	6,378	39,935	33,556	6,378	39,935	1.74	0.03	0.17	12.38	84.31	2.46	0.86
			hwl	1,801	111	1,912	38,056	2,340	40,396	38,056	2,340	40,396	1.65	0.03	0.19	1.85	93.56	4.16	0.43
			elevated	973	76	1,049	26,374	2,066	28,440	26,374	2,066	28,440	1.56	0.02	0.08	0.45	97.15	1.90	0.50
			intertidal	600	79	679	17,080	2,252	19,332	17,080	2,252	19,332	1.63	0.03	0.12	37.74	61.71	0.10	0.45
			hwl	1,402	91	1,493	40,015	2,603	42,618	40,015	2,603	42,618	1.67	0.03	0.13	4.55	93.50	1.40	0.54
			elevated	1,943	138	2,081	53,898	3,835	57,733	53,898	3,835	57,733	1.18	0.02	0.07	0.90	97.34	1.40	0.36
	24.03.19	East	intertidal	105	40	145	2,772	1,045	3,818	2,772	1,045	3,818	1.70	0.03	0.09	12.77	78.25	8.56	0.43
			hwl	463	73	536	10,291	1,626	11,917	10,291	1,626	11,917	1.74	0.04	0.10	4.46	86.77	8.16	0.61
			elevated	95	2	98	2,614	62	2,676	2,614	62	2,676	1.63	0.06	0.06	0.40	95.27	3.80	0.53
			intertidal	89	53	141	2,964	1,771	4,735	2,964	1,771	4,735	1.80	0.04	0.07	5.66	74.32	19.68	0.34
			hwl	255	19	274	7,606	581	8,186	7,606	581	8,186	1.72	0.03	0.08	2.35	86.60	8.61	2.44
			elevated	193	100	294	6,210	3,212	9,422	6,210	3,212	9,422	1.62	0.03	0.06	0.25	90.84	8.15	0.76
			intertidal	482	92	574	15,662	2,998	18,661	15,662	2,998	18,661	1.65	0.04	0.09	8.41	82.06	8.81	0.72
			hwl	592	152	744	15,470	3,982	19,452	15,470	3,982	19,452	1.66	0.03	0.09	0.50	84.20	14.77	0.53
			elevated	1,090	210	1,300	27,824	5,349	33,173	27,824	5,349	33,173	1.65	0.04	0.07	0.75	81.84	17.07	0.34

Table SM2: Microplastic (MP) abundances in lakebed sediments at Lake Tollense including pedologic information.

Sample information				*MP per kg DW*			*Pedologic characteristics*		*Grain sizes*		
Sampling	***Date***	***Sampling point***	***Region***	***Fragments***	***Fibers***	***Particles***	***Water content (%)***	***Organic matter (%)***	***Sand (%)***	***Silt (%)***	***Clay (%)***
Sep. '17	16.09.17	SLU_1	south	13,851	2,984	16,835	1.73	8.34	1.89	79.27	12.23
		SLU_2		12,685	1,831	14,516	1.85	7.99	2.55	79.87	11.62
		SLU_3	central-south	12,759	1,395	14,154	2.02	7.88	1.26	77.03	14.47
		SLU_4		15,118	2,386	17,504	1.88	8.40	1.88	77.36	14.14
		SLU_5	central-north	6,809	1,536	8,345	1.68	8.13	1.09	75.92	15.58
		SLU_6		5,230	1,261	6,491	1.64	8.24	1.07	73.21	18.29
		SLU_7	north	8,177	1,527	9,705	1.75	7.99	0.99	73.28	18.22
		SLU_8		6,905	2,251	9,156	1.47	8.45	2.46	71.36	20.15
Mar. '18	21.03.18	SLU_1	south	2,468	851	3,319	1.85	8.51	1.41	77.43	12.66
		SLU_2		1,598	1,271	2,869	1.99	8.32	2.40	82.70	14.89
		SLU_3	central-south	7,859	1,759	9,619	2.80	8.56	1.23	81.77	16.99
		SLU_4		10,839	2,530	13,369	2.70	9.30	1.52	82.48	16.00
	23.03.18	SLU_5	central-north	14,028	3,225	17,253	2.69	9.30	1.02	78.72	20.26
		SLU_6		6,842	2,120	8,961	2.64	9.44	0.87	77.52	21.61
		SLU_7	north	4,319	2,128	6,447	3.46	9.44	1.04	78.41	20.55
		SLU_8		6,959	3,136	10,094	2.21	9.57	2.45	77.08	20.47
Sep. '18	10.09.18	SLU_1	central-north	4,282	1,378	5,660	3	9.54	1.69	74.5	15.31
		SLU_2		7,774	899	8,673	2.88	9.51	1.42	73.60	16.48
		SLU_3	north	10,873	3,641	14,515	2.93	9.58	1.11	74.27	16.13
		SLU_4		11,437	3,484	14,921	2.42	9.41	2.71	68.92	19.87
	11.09.18	SLU_5	south	11,820	1,897	13,717	2.78	9.14	2.71	75.04	13.75
		SLU_6		9,615	2,376	11,990	2.59	9.42	1.45	75.22	14.83
		SLU_7	central-south	5,813	1,893	7,706	3.11	9.07	1.68	72.91	16.90
		SLU_8		5,136	1,284	6,420	2.91	9.42	1.39	73.44	16.67
Mar. '19	25.03.19	SLU_1	south	7,480	859	8,339	1.37	9.39	2.75	78.46	10.28
		SLU_2		12,001	1,448	13,449	1.81	9.18	2.43	78.62	10.45
		SLU_3	central-south	14,858	1,909	16,767	2.05	8.82	1.98	77.17	12.35
		SLU_4		5,035	1,370	6,405	1.85	9.44	2.92	77.20	11.39
	26.03.19	SLU_5	central-north	2,080	611	2,691	1.87	9.42	1.38	77.39	12.73
		SLU_6		7,439	2,614	10,053	1.87	9.33	1.36	76.28	13.85
		SLU_7	north	10,906	3,476	14,383	1.90	9.17	1.19	76.52	13.80
		SLU_8		8,758	2,144	10,902	1.48	9.32	2.74	73.66	15.10

Table SM3: Spearman correlation coefficients for microplastic concentrations related to sediment characteristics and wind data (DWD 2020). Significance levels are presented by number of *: * p < 0.05, ** p < 0.01, *** p < 0.001.

		Microplastic concentrations					
		Lakeshore sediments			**Lakebed sediments**		
		Fragments	***Fibers***	***Particles***	***Fragments***	***Fibers***	***Particles***
Sediment characteristics	***Water content***	0.02	0.29***	0.08	-0.05	0.19	-0.01
	Organic content	0.09	0.37***	0.17*	-0.27	0.16	-0.20
	Clay content	-0.27***	0.00	-0.23**	-0.22	0.29	-0.10
	Silt content				0.12	-0.06	0.08
	Fine sand content	-0.35***	0.38***	-0.17*	0.13	-0.04	0.09
	Medium sand content	0.17*	0.00	0.00			
	Coarse sand content	-0.25***	0.33***	-0.15*			
Wind	***Direction***	-0.12	-0.33	-0.14	-	-	-
	Velocity	0.50	-0.08	0.52*	-	-	-

Table SM4: Microplastic concentrations transferred to other size ranges according to Koelmans et al. (2019) and Kooi and Koelmans (2020).

	Lakeshore sediments (MP/kg DW)		***Lakeshore sediments (MP/m²)***		***Lakebed sediments (MP/kg DW)***	
Size range	***Mean***	***Median***	***Mean***	***Median***	***Mean***	***Median***
1 - 5,000 µm	135,520	127,639	3,550,729	3,231,343	2,294,910	2,164,336
50 - 5,000 µm	1,821	1,716	47,724	43,431	14,159	13,353
63 - 5,000 µm	*1,410*	*1,328*	*36,943*	*33,620*	*10,476*	*9,879*
112 - 5,000 µm	743	700	19,476	17,725	4,939	4,658
174 - 5,000 µm	453	427	11,880	10,811	2,770	2,613
300 - 5,000 µm	244	230	6,388	5,814	1,346	1,270
500 - 5,000 µm	134	126	3,512	3,196	676	637
355 - 5,600 µm	202	190	5,293	4,817	1,080	1,018

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Versicherung an Eidesstatt *(Declaration on oath)*

Hiermit erkläre ich an Eides statt, dass ich die vorliegende Dissertationsschrift selbst verfasst und keine anderen als die angegebenen Quellen und Hilfsmittel benutzt habe.

I hereby declare upon oath that I have written the present dissertation independently and have not used further resources and aids than those stated.

Hamburg, den 01.12.2021