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Microplastics in freshwater sediments: analytical methods, temporal trends, and risk of associated organophosphate esters as exemplar plastics additives

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Abstract

It has been estimated that over 28 million tonnes of plastics end up in water bodies annually. These plastics degrade into microplastics (MPs), which along with microbeads and MPs from other sources such as wastewater treatment plants continue to threaten the aquatic system. At such small sizes, and corresponding larger surface areas per unit mass/volume, MPs exhibit enhanced capacity for absorbing and desorbing toxic chemicals/additives. Therefore, MPs can serve as vectors through which additives as well as other persistent, bio-accumulative, and toxic chemicals can enter the food chain. Additives are a significant component of most plastic products with some identified as hazardous to health and the environment. One group of additives that has continued to attract interest is organophosphate esters (OPEs), which are used both as flame retardants and plasticizers. Some of these OPEs are suspected carcinogens and endocrine disruptors and have been reported to exert serious toxic effects on freshwater biota. Separate studies on the presence and fate in the freshwater environment of these additives and MPs have emerged recently. However, no studies exist that examine the extent to which plastics additives such as OPEs in sediments are sorbed to MPs as opposed to the sediment itself. This has potentially important implications for the bioavailability of such additives and studies to examine this are recommended. This paper reviews critically the current state-of-knowledge on MPs in freshwater sediments, methods for their analysis, as well as their occurrence, temporal trends, and risks to the freshwater aquatic environment. Moreover, to facilitate the study of additives associated with MPs that have been extracted from sediments,

we consider the possible effect of MP isolation methods on the determination of concentrations of associated additives like OPEs.

Keywords: Microplastics, freshwater sediment, organophosphate esters (OPEs), plasticizers, flame retardants, analytical methods.

1. Introduction

The littering effect of human activities on water bodies can be traced back to over a hundred and forty years ago (Verne 1998). Today the concern has mounted as the list of harmful substances threatening the hydrosphere (which sustains nearly half of global primary production and serves as habitat to millions of aquatic species) continues to increase (Dümichen et al. 2015; Field et al. 1998; Ibe and Kullenberg 1995). These substances are associated with materials including medical waste, cigarette filters, fishing nets, and plastics which have recently become the focus of increased environmental/health concern. About 19 to 23 million metric tonnes (11%) of plastic wastes generated globally in 2016 ended up in aquatic ecosystems (Borrelle et al. 2020) and it is estimated that the global mismanaged plastic waste would be approximately 155 to 266 million metric tonnes by 2026 (Everaert et al. 2018; Lebreton and Andrady 2019). That would mean an estimated 17 to 29 million metric tonnes of these mismanaged plastic wastes ending up in aquatic systems. The Great Lakes alone have been reported to receive around 10,000 tonnes of plastics annually (Hoffman and Hittinger 2017), making the problem of plastic pollution in freshwater systems as serious as in marine environments.

Plastics enter the aquatic environment from a wide range of sources. Over time, they degrade through photolytic, mechanical, and biological processes into sizes smaller than 5 mm at which point they are referred to as microplastics (MPs) (Arthur et al. 2009). MPs that originate from the degradation of originally larger plastic items are referred to as secondary MPs, while those released directly into the environment in the micro size range (< 5 mm size) are described as primary MPs (Boucher and Friot 2017). Whether primary or secondary, MPs in aquatic environments will either become suspended in the water column or accumulate within sediments (Woodall et al. 2014). Additionally, due to their smaller sizes and corresponding larger surface areas per unit mass/volume, they may exhibit a greater tendency towards adsorbing and desorbing toxic chemicals/additives (Mason et al. 2016). A wide range of

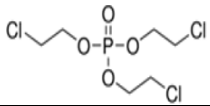
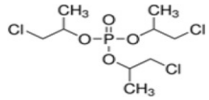
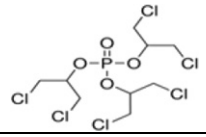
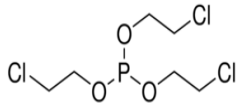
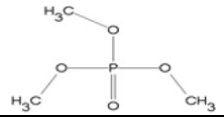
additives such as plasticizers, pigments, fillers/extenders, and flame retardants are often incorporated in plastics during manufacture to give them specific properties. The problem however is that some of these additives-which make up approximately 50% of some plastic products (Hartmann et al. 2019; Nel et al. 2021b) have been found to be hazardous to health and the environment (Lai et al. 2015; Pantelaki and Voutsa 2019).

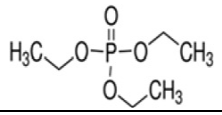
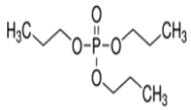
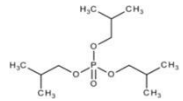
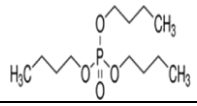
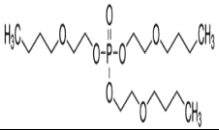
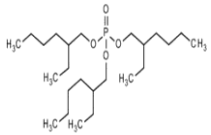
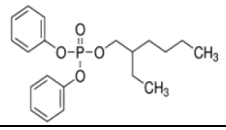
The risk of MPs transporting plastic additives as well as other persistent, bio-accumulative, and toxic (PBT) chemicals into the food chain upon ingestion by aquatic and terrestrial organisms has become a subject of great concern (Guimarães et al. 2021). Assessment of such risks is complex given the risks are due not only to MPs themselves, but to associated chemical additives. Thus, discerning the impacts of MPs from those of additives present in those MPs, as well as those of additives leached from MPs and larger plastic waste into the environment, is an important research goal. Organophosphate esters (OPEs), which are used both as flame retardants and as plasticizers are one of the most important and widely-used plastic additives (Herrera et al. 2003; Wang 2000). Chlorinated OPEs such as tris(2- chloroethyl) phosphate (TCEP), tris(2- chloroisopropyl) phosphate (TCIPP), and tris(1,3- dichloro- 2- propyl) phosphate (TDCIPP) are mostly used as flame retardants, while the non-chlorinated alkyl-, aryl- phosphates are often used as industrial lubricants and as plasticizers (Zeng et al. 2018). OPEs have been associated with serious health and environmental concerns (Table 1). Beyond environmental persistence, the possibility of some- OPEs (mostly the chlorinated alkyl phosphates) (Kawagoshi et al. 2002; Shi et al. 2018) being carcinogenic as well as posing a risk of other such serious health concerns as infertility and neurotoxicity have been reported (Hoffman et al. 2017; Hou et al. 2016; Van der Veen and de Boer 2012, Stapleton et al. 2009; WHO 1993). Decreased semen quality and other fertility problems have been associated with TDCIPP and triphenyl phosphate (TPHP) (Stapleton et al., 2009) while tris(methyl-phenyl) phosphate (TMPP) and triphenyl phosphate (TPHP) have both displayed neurotoxic potential in zebra fish larvae (Shi et al., 2018). TCEP has been classified as a category 2 reproductive toxicant and category 2 carcinogen (EU 2009). Another area of concern is the effect of the alcohols released as a result of hydrolysis of some of these OPEs, as it has been confirmed that butoxyethanol produced by the hydrolysis of tributoxy ethyl phosphate (TBEP), is not just a mutagen but also an endocrine disruptor (Boatman et al. 2004). Overall, the ecotoxic effects of several chemical plastic additives including flame retardants and plasticizers are well-documented in the literature. Therefore, the enhanced release of these hazardous chemical additives to the aquatic environment from MPs due to their large surface area to mass/volume

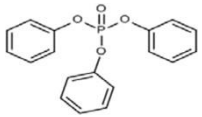
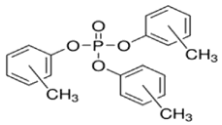
ratios and subsequent toxic impacts on aquatic organisms is highly concerning. Most recent reviews have focused on sources/abundance, environmental impact, fate, and transport of MPs (Choong et al. 2020; Chowdhury et al. 2020; Koutnik et al. 2021), however, not many have looked at plastic additives and to the best of our knowledge, none have considered the possible impact of MP isolation methods on measurements of plastic additives like OPEs.

It is against this backdrop that this paper provides a critical review of the current state-of-knowledge on MPs in freshwater sediment including MPs occurrence, temporal trends, and potential risk to the aquatic environment. Methods for the analysis of MPs in freshwater sediment are reviewed, including consideration of the impact of such methods on measurements of additives like OPEs in MPs isolated from sediments. Finally, research gaps are highlighted and recommendations for future research are presented.

Table 1: Names, abbreviations, Structures, CAS Numbers, molecular formula, application environmental/health hazard of some OPEs (Sigma-Aldrich, 2021, Wei et al., 2015, Van der Veen and de Boer, 2012, Reemtsma et al., 2008, Hawley and Lewis Sr, 2001, Cadogan and Howick, 2000).

COMPOUND	ABBR.	STRUCTURE	CAS NO.	MOLECULAR FORMULA	APPLICATIONS	ENVIRONMENTAL/HEALTH HAZARD
Tris (chloroethyl) phosphate	TCEP		115-96-8	$C_6H_{12}Cl_3O_4P$	Flame Retardant, Plasticizer, Lacquer Paint, Glue, Industrial processes	Carcinogenic, highly toxic, environmentally persistent, and cytotoxic in high concentration
Tris (2-chloroisopropyl) phosphate	TClPP		13674-84-5	$C_9H_{18}Cl_3O_4P$	Flame Retardant, Plasticizer	Suspected carcinogen, and cytotoxic in high concentration
Tris (1,3-dichloro-2-propyl) phosphate	TDClPP		13674-87-8	$C_9H_{15}Cl_6O_4P$	Flame Retardant, Plasticizer, Lacquer Paint, Glue	Carcinogenic, Developmental, and reproductive toxicity, endocrine disruption
Tris(2-chloro-ethyl) phosphite	CLP1		140-08-9	$C_6H_{12}Cl_3O_3P$	Flame Retardant, Stabilizer and Hydraulic fluids, and Industrial processes	Suspected carcinogen, Reproductive toxicity
Trimethyl phosphate	TMP		512-56-1	$C_3H_9O_4P$	Industrial processes	Genotoxic, possible reproductive toxicity

Triethyl phosphate	TEP		78-40-0	$C_6H_{15}O_4P$	Plasticizer, Catalyst, ethylating agent, Raw material for insecticides	Possible developmental and reproductive toxicity
Tripropyl phosphate	TPP		513-08-06	$C_9H_{21}O_4P$	Flame Retardant, Plasticizer, Hydraulic fluids, Lacquer paint, glue, Cosmetic products and industrial processes	
Tris (isobutyl) phosphate	TIBP		126-71-6	C12H27O4P	Plasticizers, Hydraulic fluid, Floor finish, wax, lacquer paint, glue, Ant-foam agent and industrial processes	
Tri-n-butyl phosphate	TNBP		126-73-8	C12H27O4P	Flame retardants, Hydraulic fluid, Plasticizer,	Neurotoxic, suspected carcinogen
Tris (2-butoxyethyl) phosphate	TBOEP		78-51-3	C18H39O7P	Flame Retardant, Plasticizer, floor finish, wax, lacquer paint, glue, anti-foam agent	Suspected carcinogen, developmental toxicity
Tris (2-ethylhexyl) phosphate	TEHP		78-42-2	C24H51O4P	Flame Retardant, Plasticizer and fungus resistant	Not considered to be carcinogenic in humans. Reproductive and developmental toxicity needs to be investigated.
2-Ethylhexyl diphenyl phosphate	EHDPP		1241-94-7	C20H27O4P	Flame retardants, Plasticizer, certain food packaging applications	

Tris (phenyl) phosphate	TPHP		115-86-6	C18H15O4P	Flame retardants and Plasticizers	Reproductive and developmental toxicity, Neurotoxicity, genotoxicity, endocrine effect. Bioaccumulation and very toxic to aquatic life.
Tris (methylphenyl) phosphate (or Tricresyl phosphate)	TMPP		1330-78-5	$C_{21}H_{21}O_4P$	Plasticizer, flame-retardant, Industrial processes, non-flammable fluid in hydraulic systems and as stabilizers	Neurotoxic, acute toxicity, Hazardous to the aquatic environment,

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1.1. Risk of MPs and associated additives to the freshwater environment

Despite the focus on the marine environment, MPs have been reported in freshwater systems such as lakes and rivers (Eriksen et al. 2013, Hoellein et al. 2014; Zbyszewski et al. 2014) and the likelihood that the abundance of these MPs in such compartments will continue to increase has been reported by several researchers (Eriksen et al. 2013; Geyer et al. 2017). This increasing trend in MP pollution is accompanied by several risks to the freshwater environment associated with both the MPs and their chemical additives.

1.2 Risks associated with MPs.

The environmental persistence of MPs is a subject of great concern as fears exist that they can reside in the environment for hundreds of years once released (Zbyszewski and Corcoran 2011), and moreover because of the potential toxic effects of these MPs on biota. The neurotoxic potential of MPs has been reported following a study involving zebrafish juveniles (Guimarães et al. 2021). MPs have also been reported to impact adversely on plants as decreased root cell viability and growth was observed in a freshwater floating plant (duckweed *Lemna minor*) following exposure to microbeads (Kalčíková et al. 2017). Adsorption of MP particles to the roots of a vascular plant (*Spirodela polyrhiza*) has also been reported (Dovidat et al. 2020). Coupled with the fact that these plants form an essential part of the ecosystem, this raises concern over rising MPs concentrations in the freshwater environment.

Ingestion of MPs by aquatic organisms has also been reported covering 39 freshwater species (4 fish species and 35 invertebrates) (Scherer et al. 2018). A separate study by Hurley et al (2017) showed that sediments play a major role in driving this intake of MPs as *Tubifex* worms ingested a mean concentration of 129 ± 65.4 MP particles/g tissue following their exposure to sediments with concentrations ranging from 56 to 2543 MP particles/kg (Hurley et al. 2017). Although egestion has been reported as rapid and effective, MPs tend to remain in guts of fishes and sometimes translocate to internal organs (Parker et al. 2021). The presence of MPs in the guts, stomach, and liver of freshwater organisms such as snails, crustaceans, worms (Imhof et al. 2013), zooplankton (Cole et al. 2013), and fish (Sanchez et al. 2014, McGoran et al. 2017, Silva-Cavalcanti et al. 2017) have also been reported. These ingested MPs not only serve as vectors by which organic and inorganic pollutants are transported into living organisms, (Bakir et al. 2012; Koelmans et al. 2013) but they can clog the digestive tract of organisms such as in the Norway lobster *Nephrops norvegicus* (Murray and Cowie 2011) and zebra mussel *Dreissena polymorpha* (Magni et al. 2018) and eventually lead to death (Barnes et al. 2009).

Reduced feeding in zooplankton (Cole et al. 2013) and lugworms (Besseling et al. 2013) as well as decreased fecundity for copepods (e.g. *Tigriopus japonicus*) (Lee et al. 2013) has been reported following ingestion of MPs. There have also been reports of plastics in dried fish tissues from four commonly consumed dried fish species (Karami et al. 2017). These and other similar studies show that although the effect of MPs on human health is still unclear, dietary exposure to MPs may be substantial.

1.3 Risks from chemical plastic additives associated with MPs.

Some toxic effects of MPs have been attributed to associated additives and sorbed pollutants, with previous studies highlighting the enhanced toxicity of pollutants when they coexist with MPs (Deng et al. 2018; Ogata et al. 2009; Teuten et al. 2009). Rochman et al, (2013) observed less signs of stress in fish fed virgin polyethylene fragments than those fed marine polyethylene fragments (Rochman et al. 2013b); while a separate study has reported embryotoxicity in purple sea urchin (*Paracentrotus lividus*) following exposure to MP additives leached from micronized plastic toys (Oliviero et al. 2019). Furthermore, increased bioaccumulation of polychlorinated biphenyls (PCBs) in zooplankton because of MP ingestion and the possibility of biomagnification as these MPs and associated additives move up the food chain has been reported (Mattsson et al. 2017; Teuten et al. 2009). Rochman et al. (2013) also reported bioaccumulation of chemical pollutants (polycyclic aromatic hydrocarbons (PAHs), PCBs and polybrominated diphenyl ethers (PBDEs) following ingestion of polyethylene MPs by exposed fish species that also suffered liver toxicity due to such exposure (Rochman et al. 2013b). A recent study by Garcia-Garin et al. (2020), detected OPEs in muscle samples of the edible Bogue – *Boops boops* (Garcia-Garin et al. 2020). Although this study did not correlate the detection of OPEs to MP ingestion, previous studies have recommended *Boops boops* for studies on MP pollution because of the frequent detection of particles in their guts (Bray et al. 2019). It is however interesting to note that despite the potential adverse biological effects of OPEs outlined in section 1, there remain to date no studies that have examined the risks to freshwater biota from OPEs associated with sediment MPs. Research to examine such risks from OPEs and other additives is urgently required.

2. Occurrence of MPs in freshwater sediment

In order to investigate research trends in publications studying the occurrence/concentrations of MPs in freshwater sediments between 2000 and 2020, a search was conducted in the *ISI web of knowledge* database using different combinations of the keywords "*microplastic",*

"freshwater" and "sediment". A total of 245 relevant publications were found of which 20% (49) have been published between 1st of January and 28th June 2021. 31% (77) were published in 2020, 24% (58) were published in 2019, 13% (33) in 2018 and only 11% (28) between 2017 and 2013 (Figure 1). This illustrates increasing interest in the study of MPs in freshwater ecosystems.

A notable geographical trend is that over 78% of these studies were conducted in only 7 countries (Figure 2). This shows a serious lack of data on freshwater microplastics for most parts of the world. Only 12 studies were undertaken in Africa (5 in south Africa) even though Africa's 205,670 km^2 lake area accounts for 14.1% of the world's lake area (Herdendorf, 1982).

Between 2015 and June 202, there have been significant improvements in the understanding and measurement of MPs in freshwater sediment (Table 2). The use of microscopy for the identification of MPs was recommended by researchers in 2015 (Masura et al. 2015; Song et al. 2015) and has subsequently become the most common method used. Vaughan et al., (2017) used microscopy to determine the abundance and distribution of MPs in what was described as "the first assessment of microplastics concentration in the sediments of either a small or an urban lake and the first for any lake in the UK". The study reported relatively low concentrations (25–30 particles per 100 g d/w) of MPs in Edgbaston Pool, Birmingham, UK but acknowledged the fact that more recent methods of identification and characterization (e.g. Fourier-transform infrared spectroscopy (FTIR) and Raman spectroscopy) were not available (Vaughan et al. 2017). Compared to other studies of freshwater lakes across the globe (i.e. Fischer et al. 2016; Su et al. 2016), Yuan et al. (2019a) described Poyang Lake in China as having higher levels (54–506 items/kg dw) and biological risks of MPs. Research has also shown that a combination of one or more methods can enhance the quality of results, as one method can compensate for the disadvantages of another (Hidalgo-Ruz et al. 2012). It is clear from the foregoing that 2017 to 2018 marked an upturn of interest in the study of MPs in freshwater sediments. Therefore, an overview of studies between January 2016 and December 2020 provides a more recent and focused perspective of the current trends in the study of MPs in freshwater sediments (Table 2). Furthermore, our focus on recent publications provides insights into the current challenge arising from the lack of harmonization in reporting formats and sampling designs that has been identified as a major impediment to elucidating temporal trends in environmental pollution with MPs (Browne et al. 2015; Hanvey et al. 2017; Mai et al. 2018).

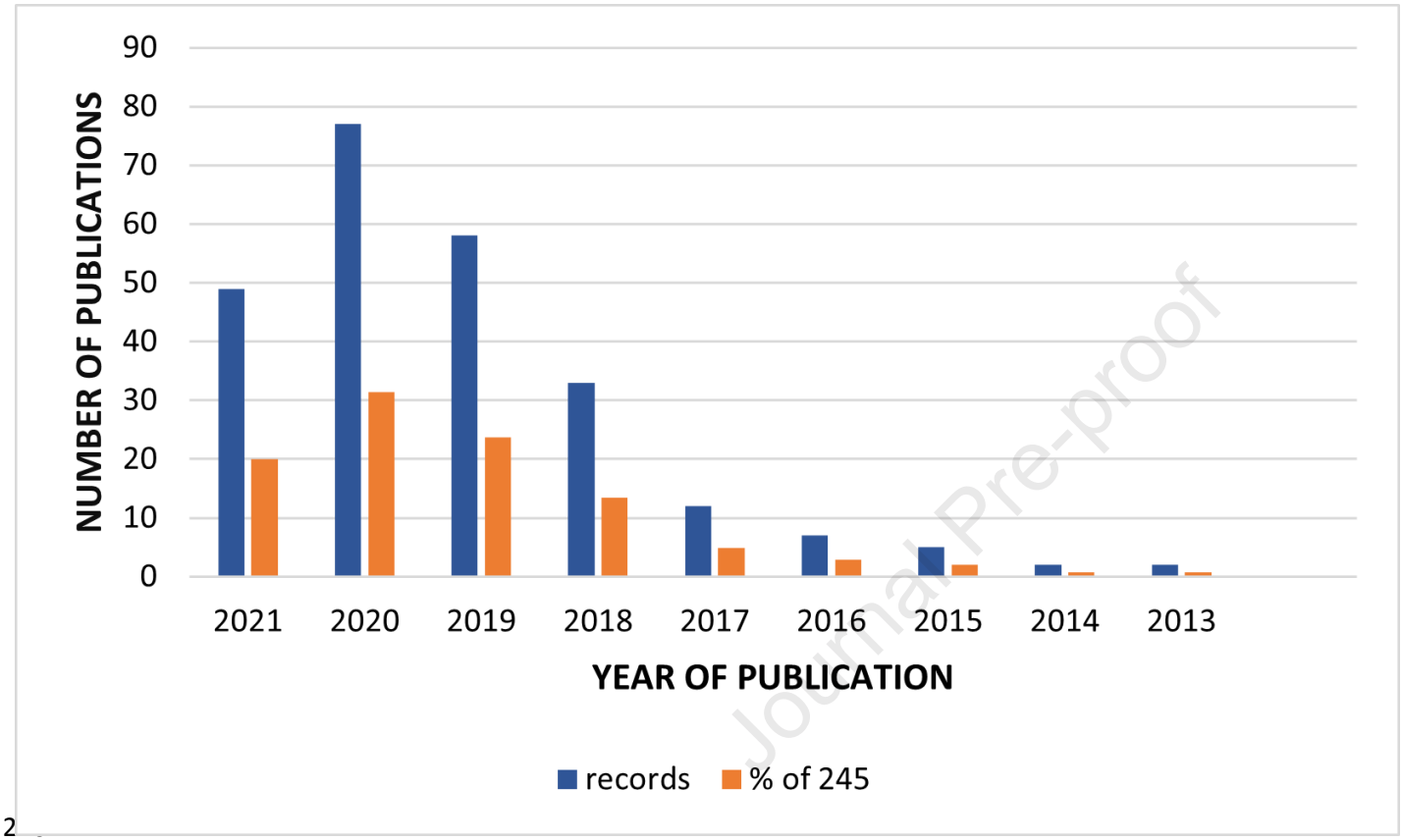

Figure 1: Number of publications per year on MPs in freshwater sediments between January 2000 and June 2021 (data obtained from ISI web of knowledge)

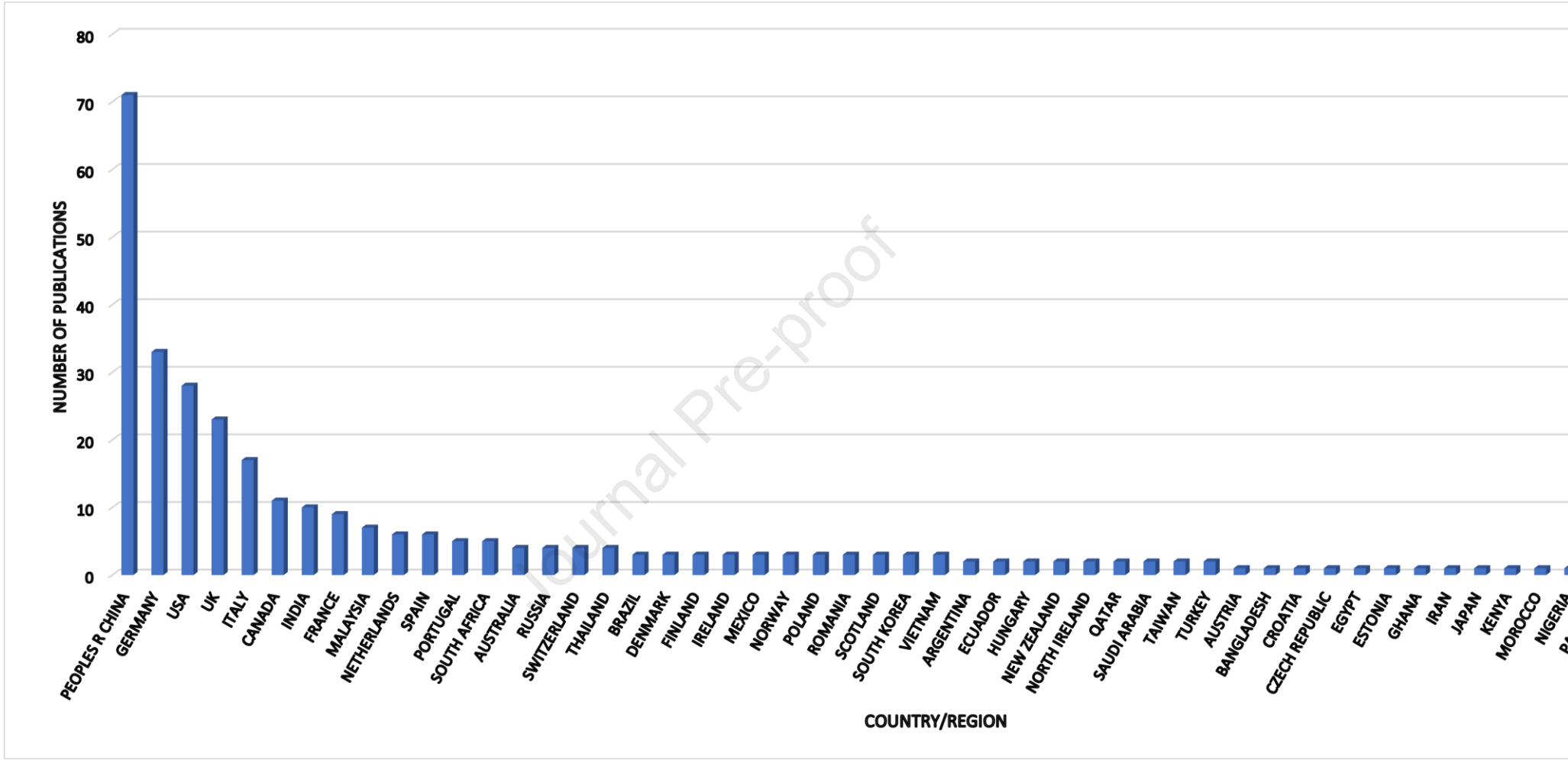

Figure 2: Geographical distribution of publications on microplastics in freshwater lake sediments between January 2000 and June 2021

(Data obtained from ISI web of knowledge)

227 **Table 2:** Overview of studies of microplastics in freshwater environments between 2016 and 2020.

Country/region	Size range	Concentration of MPs	Trend identified	Gaps identified	Reference
Europe (Germany)	20–5000 µm	Median concentration of 7.57×10^5 p m^{-3}	High MP pollution comparable to the high levels reported in some Asian countries.	Difficulty of data comparisons because of the diversity of analysis and methods used in monitoring studies	Scherer et al. 2020
Europe (Hungary)	≥100 µm	Range from 0.46 to 1.62 particles/kg, and a mean value of 0.81 ± 0.37 particles/kg.	Higher concentrations of the identified polymer types were seen at pond inlet compared to outlet and the concentration of sediment MP in this study were much lower than other international results	Intensive investigation of sediment and water samples is required, and such studies should ensure that all suspected particles are exactly identified using suitable methods such as FTIR as this is missing in most current studies.	(Bordós et al. 2019)
Europe (United Kingdom-London)	1 µm to 500 µm	Approximately 539 particles per kilogram of dried sediment	Relatively low number of MPs were found in older sediments.	Improvements in contaminant reduction measures are highly recommended for future stratigraphic work. Use of more effective microplastic detection methods along with future paleolimnological work to allow a more diverse quantification of historical flux of microplastic wastes in terrestrial, freshwater, and marine environments.	Turner et al. 2019

Asia (China)	0.03-0.1 mm	821 ± 100 MP/kg to 1936 ± 121 MP/kg	Compared to an earlier study on the same study location (Yuan et al., 2019), A higher concentration of MPs (approximately 4–30 times higher) was reported.	further long-term and systematic studies of the environmental fate of smaller size MPs (<0.1 mm) in freshwater systems.	Jian et al. 2020
Asia (Urban surface waters of Wuhan, China)	50 – 5000 µm.	1660 ± 639 to 8925 ± 1591 number of particles per cubic meter of water (n/m^3)	High heterogeneity was discovered in the concentration of MPs across the study areas. A decrease in the concentration of MPs with increasing distance from the urban center was discovered. The plastic levels recorded for the rivers were less than for the lakes, likely because of enhanced loss of MPs from rivers as a result of stronger hydrodynamics. Fibrous (52.9 – 95.6%), colored (50.4 – 86.9%) and small (MPs less than 2 mm accounting for more	Comprehensive monitoring of the abundance of microplastics in inland freshwaters is recommended. Further research on accurate determination of MP sources and pollution control strategies is needed. A need for studies of the effect of color on the rate of ingestion of MPs is highlighted	Wang et al. 2017

			than 80%) MPs were the most dominant.		
Asia (Lake Ulansuhai in China)	Six size categories: < 0.5 mm, 0.5 – 1 mm, 1–2 mm, 2–3 mm, 3–4 mm and 4–5 mm	1760 ± 710 to 10,120 ± 4090 number of particles per cubic meter of water (n/m^3)	Spatial distribution of MPs was such that higher levels of MP were recorded at the entrance of a farmland drainage canal with a downward trend towards the end of the drainage canal of the lake	Further studies of the toxic hazard MPs pose to plants and aquatic animals is recommended	Wang et al. 2019
Asia (Poyang Lake China)	<0.5 mm	54 – 506 number of particles per kilogram dry weight (dw)		More accurate and effective studies should be carried out on the impacts and fate of microplastics in freshwater ecosystems. Also, the potential impacts of MP consumption by human beings should be studied	Yuan et al. 2019
Asia (Changsha, China Surface sediments)	<1mm	Ranged from 270.17 ± 48.23 items/kg to 866.59 ± 37.96 items/kg (dw)		More research on understanding the environmental behavior of microplastics in urban waters	Wen et al. 2018

Asia (West Dongting Lake and South Dongting Lake, both in China)	macroplastics (>25 mm) and mesoplastics (5–25 mm)	From 617 to 2217 items/m^3 and 717 to 2317 items/m^3 in surface water of West Dongting Lake and South Dongting Lake, respectively while the lakeshore sediment for both study locations ranged from 320 to 480 items/m^3 and 200–1150 items/m^3, respectively.		Further work is encouraged on the effect of hydrodynamic conditions on the distribution of microplastics	Jiang et al. 2018
North America (Lake Ontario Canada)	‹ 2 mm	Approximately 760 particles per Kg dry sediment.	It is likely that fibers are transported through suspension for greater distances than fragments. Some of the previously identified methodological problems in microplastic research have been resolved. However, standardization of operational protocol will still require more research.	Thorough analysis of microplastic morphology and composition may be used to shed more light on the role played by the type of microplastic on the depth to which they are transported in an aquatic environment. Future studies aimed at understanding how microplastic abundance in sediment vary with distance from outfall are recommended. This should be done using studies directly adjacent to storm waters,	Ballent et al. 2016

				wastewater treatment plants and sewer outfalls. Future monitoring of microplastic in the sediments of Lake Ontario is also recommended	
North America (Lake Michigan, USA)	<1 mm	An average plastic abundance of ~17,000 particles/km^2 lake surface area	The relative abundance of fragments shows that secondary sources of microplastics are more significant to this study area than primary sources. The order of abundance of plastic types (polyethylene- mostly high-density being the most abundant followed by polypropylene) is consistent with global trends in the mass production of plastics.		Mason et al. 2016
North America (St. Lawrence River-Canada)	Mostly <400 µm	Concentration of MPs range from 65 to 7562 plastics per kg d/w (mean concentration is 832 (±150 SE) plastics per kg d/w.	Factors such as sediment characteristics, environmental filters and point sources are related to the variation in the distribution, abundance and diversity of microplastics across a large river.	There is the need for a standardized protocol in MP analysis and future studies should look at the mass balance of microplastic loads as well as their fate during transport to the ocean as this will provide answers on whether river sediments are permanent or transient sinks for microplastic pollution	Crew et al. 2020

Africa (Johannesburg South Africa)	53 to 4000 µm	MP abundances range from 4 particles kg^{-1} dw to 1347.5 particles kg^{-1} dw and mean of 166.8 particles kg^{-1} dry weight	Distribution of MPs is influenced by such factors as water depth, flow and obstructions in rivers such as large weirs.	Increased focus on understanding microplastic pollution in South Africa and further studies aimed at understanding the behavior of microplastics in the environment.	Dahms et al. 2020
Africa (Lake Victoria in Uganda)	300-5000 µm	The abundance of MPs ranged from 0 – 108 particles/kg (dw)	Landing beaches were identified as major plastic pollution hotspots as MP concentration for landing beaches far exceed results for recreational areas.		Egessa et al. 2020
Africa (Nigeria)	20 – 5000 µm	MP abundances range from 347 to 4031 items·kg−1 (In dry season) and 507–7593 items·kg^{-1} (In rainy season)	The most common plastic materials found were Polyethylene terephthalate (PET) and Plasticised polyvinyl chloride (PVC P). The least abundant were Polyamide (PA) and Polystyrene (PS).		Oni et al. 2020
Japan Thailand Malaysia South Africa	315 µm–5 mm (Although most were between 315 µm – 1 mm)	Japan: 1900 pieces/kg dry sediment Thailand: 100 pieces/kg dry sediment	For all study locations, there has been an increasing trend in the abundance of MPs from the 1950s to the 2000s.	A more detailed analysis of microplastic pollution with time using more of the core is recommended. To enable a better understanding of the controlling factors behind the difference in	Matsuguma et al. 2017

			Sediment is an important sink for MPs as more MPs were found in sediments than the surface water of Tokyo Bay. There is a consistency between the trend of microplastic abundance in sediments and the global trend of plastic production.	polymer composition between the different study areas, Analysis of more samples is recommended.	

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3. Analysis of MPs in sediment

The analysis of MPs in sediments requires strict adherence to standard procedures. This is particularly important considering the possibility of post-sampling contaminations (Woodall et al. 2015). Based on published methodology, such impacts were addressed according to the following sub-headings: (1) sampling, (2) extraction, (3) identification / quantification, and (4) quality assurance/quality control (Figure 3).

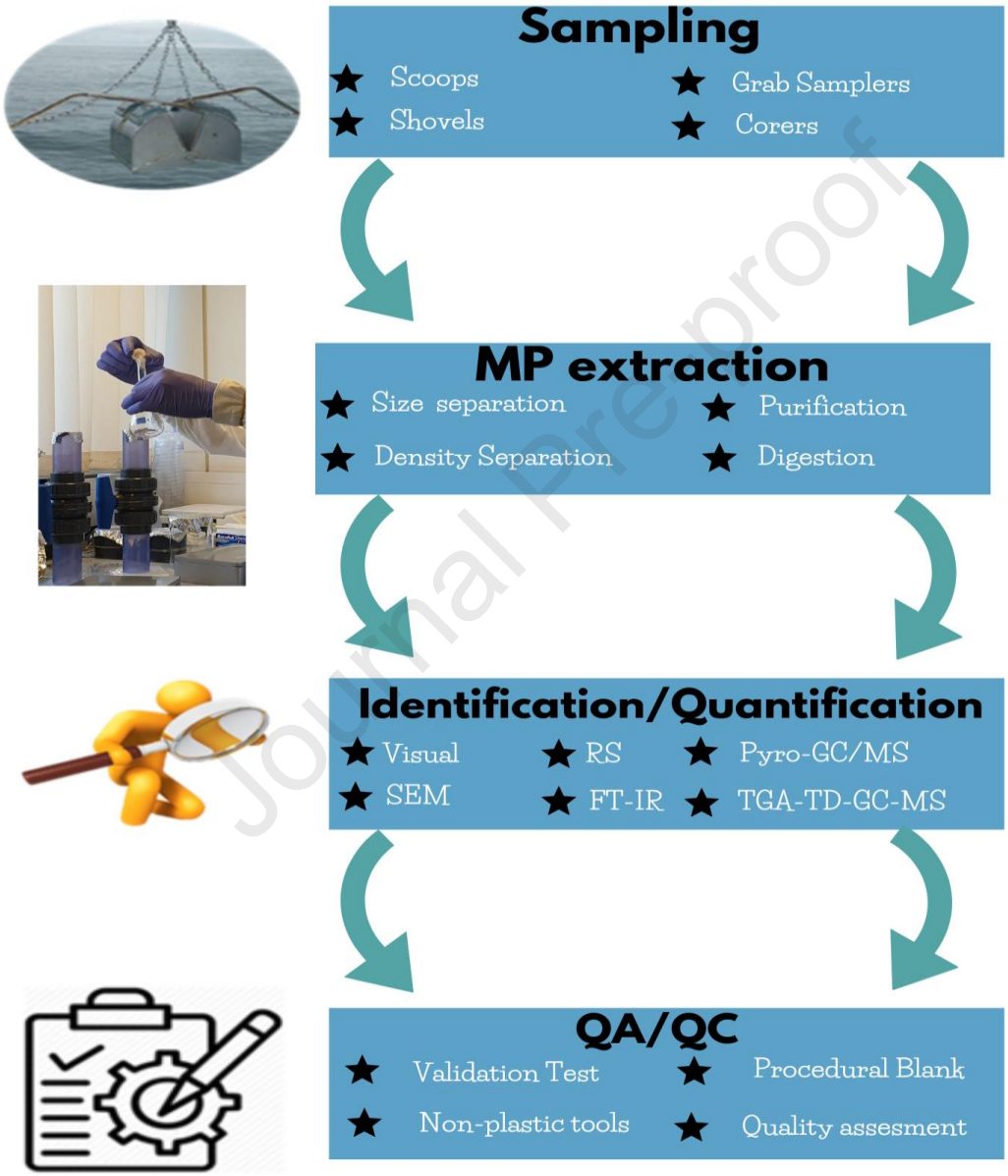

Figure 3: Flow chart of the procedures followed for analysis of MPs in sediments (Pyro-GC/MS*: Pyrolysis Gas Chromatography/Mass Spectrometry*, TGA-TD-GC–MS: *Thermogravimetric analysis-thermal desorption-gas chromatography–mass spectroscopy,* FTIR: *Fourier transform infrared,* SEM: *scanning electron microscope, RS: Raman spectroscopy*)

3.1 Sampling

Several tools and approaches have been employed to sample sediment most of which are dictated by the objectives of the research and the peculiarities of the study location. However, previous reviews on the methods of measuring MPs in sediments (Hanvey et al. 2017; Mai et al. 2018) identified a lack of standardized procedures in sediment sampling, leading to the reporting of data in a variety of units. These units include MP pieces per m^2 (do Sul and Costa 2014; Wessel et al. 2016), pieces per mL or L (Browne et al. 2011; Woodall et al. 2015), or per weight (g or kg) of sediment (Alomar et al. 2016; Klein et al. 2018; Ng and Obbard 2006). This has made it difficult to compare the results of different studies quantifying MP contamination across the world and is further complicated by the density estimation required for conversion between such units that can lead to errors (Van Cauwenberghe et al. 2015).

In order to get a more focused view of this subject as it relates to the most recent studies, the keywords "Sediment sampling for microplastics in freshwater" were used to search the entire database of the ISI Web of Science for relevant studies between 2018 and 2020. The result was then narrowed down to seventeen (17) publications from different regions of the world using such criteria as year of publication and relevance to the topic of interest. For sampling below surface sediment, eight of the studies used Van Veen or other grab samplers (Baldwin et al. 2020; Crew et al. 2020; Eo et al. 2019; Ramírez-Álvarez et al. 2020; Rodrigues et al. 2018; Scherer et al. 2020; Shruti et al. 2019; Xu et al. 2020) and one used a box corer (Pagter et al. 2020). For surface sediment sampling, metal hand scoop (Schessl et al. 2019), stainless-steel shovel (Jiang et al. 2018; Peng et al. 2018), metal pail shovel (Deng et al. 2020; Ramírez-Álvarez et al. 2020), stainless steel split spoon or Ekman dredge (Eo et al. 2019; Wilkens et al. 2020), garden spade (Mani et al. 2019; Peller et al. 2019) and stainless-steel spatula (Toumi et al. 2019) were used. This further confirms the lack of standardization between sampling methods while showing that the box corer seems the most common approach to depth sampling. It is clear though, that the choice of equipment and sampling procedure is often governed largely by the nature of sediments to be sampled and the accessibility of the sampling site. A further consideration is that secondary contamination by MPs can be avoided by use of non-plastic sampling tools (Jiang et al. 2018; Wen et al. 2018).

3.2. Extraction

Once sediment samples are collected and transported to a laboratory for analysis, the next objective is to separate the MPs from a complex matrix of other organic and inorganic materials. A standard method for doing this is yet to be established (Horton et al. 2017) but most recent studies seem to employ a combination of size and density separation techniques (Vaughan et al. 2017). The use of fluidization/elutriation (another density-based technique) has also been reported in some recent publications (Claessens et al. 2011; Kedzierski et al. 2016; Nuelle et al. 2014; Zhu 2015).

Most of the reviewed studies employed varying mesh sizes including: 8 µm to 47 mm (Zhang et al. 2019), and 90 µm to 4.75 mm (Pan et al. 2019); it is however recommended that the choice of mesh size should be such that it allows the collection of a wide size fraction, while minimizing clogging of mesh holes (Mai et al. 2018).

The second approach is density separation, this approach takes advantage of the low density of most plastic materials (0.9 – 1.55 g/cm^3). In a review of 48 studies from 18 different countries, Hanvey et al. (2017) reported the application of density separation in 84% of studies. This is partly because recovery rates as high as 100% have been previously reported (Mai et al. 2018). The basic idea behind this approach is allowing the MPs to float in a high-density solution thereby permitting the MPs to be filtered out. Four main steps have been identified, specifically: (1) introduction of an aqueous solution of known density, (2) manual or mechanical shaking/stirring for a specific period, (3) allowing time for settling and equilibration, and (4) filtration (Hanvey et al. 2017). The aim of shaking or stirring after introducing the solution is to ensure that the MPs are properly separated from other sediment particles. This can either be done manually, (Wang et al. 2019; Zhang et al. 2019), or mechanically using a vortex mixer (Woodall et al., 2014) or mechanical shaker (Ng and Obbard, 2006). The time allowed for equilibration and settling after the shaking can be as short as five minutes (Corcoran et al. 2015), or up to 12 hours (overnight) (Stolte et al. 2015). There are a variety of high-density solutions that can be used for the first step and a recovery rate of 80 – 100% has been reported for NaCl solution (Fries et al. 2013), 96 – 100% for $ZnCl_2$ solution (Imhof et al. 2012) and 94 – 98% for NaI solution (Claessens et al., 2013). Turner et al., 2019 used sodium polytungstate (SPT) solution to achieve a density as high as 2.2 g/cm^3, suitable for the extraction of high-density plastics such as polytetrafluoroethylene (2.08 - 2.17 g/cm^3) and polyvinylidene fluoride or polyvinylidene difluoride (1.79 g/cm^3) (Turner et al., 2019a).

The use of water for density separation reported a low recovery rate of less than 70% (Quinn et al. 2017).

As an alternative to the density-based techniques, an oil extraction protocol has been reported (Crichton et al. 2017). This cost-effective alternative had a reported recovery rate of 92.7% ± 4.3% for fibers and 99% ± 1.4% for particles. In recent times, the use of techniques capable of providing recovery rates as high as 100% has also been reported. These include the use of pressurized fluid extraction (which has provided recovery rates between 101 and 111%) (Fuller and Gautam 2016) and air-induced overflow (91 – 99%) (Nuelle et al. 2014)

Following the removal of all or most of the inorganic materials such as silicates, the next step is the filtration or wet sieving of the mixture. This is meant to separate suspended low density MPs from other components of the solution. This separation is principally achieved by sieving or vacuum filtration and for most studies, the use of stainless-steel sieves or glass fiber filters is the norm (Mai et al., 2018). Just as for dry sieving, the choice of mesh size should be such as allows the collection of a wide range of size fractions and minimizes clogging of mesh holes.

Digestion of organic matter may improve MP detection accuracy and remove interference during polymer identification (Prata et al. 2019) with several methods used by previous researchers (Table 2). In a review of over 60 publications, Prata et al (2019) reported that 60% did not carry out any form of purification, 35% used H_2O_2 and 15% used Fenton's reagent (a solution of H_2O_2 with ferrous iron as a catalyst). Two other reviews Mai et al. (2018) and Hanvey et al. (2017b) concluded a solution of 30% H_2O_2 to be the method of choice for this process. This is because it is very effective in dissolving higher molecular weight organic matter, while leaving the physical properties of the MPs intact (although it has been speculated that changes in the color of MPs are possible) (Mai et al., 2018).

Table 3: Methods for separating sediment MPs from organic and inorganic matter**.**

Method	**Description**	**Disadvantage**	**Reference**
Acid digestion	Use of acids such as nitric acid (55% HNO_3) and hydrochloric acid (37% HCl) to degrade organic matter	Risk of digesting some low resistance MPs such as nylon and polyethylene terephthalate at high acid concentration and temperature	(Qiu et al. 2016)

Alkali digestion	Use of alkali such as potassium hydroxide (10% KOH) and sodium hydroxide (20 -50% NaOH) for the digestion process	Risk of damage or discoloration of MPs	(Mai et al., 2018), (Qiu et al. 2016)
Oxidizing agents	Use of oxidizing agents such as hydrogen peroxide (35% H_2O_2). Sometimes combined with NaOH and HCl for better performance.	Shrinkage of certain plastics has been observed.	(Prata et al., 2019)
Enzymatic digestion	This involves the use of enzymes such as proteinase K (500 mg/mL) for organic matter digestion.	No known risk to the integrity of MPs associated with this method yet, but its application is not as straightforward as chemical digestion methods	(Karlsson et al. 2017)
Other Methods	Use of microwaves and ultrasonication	High risk of damage to the MPs.	(Karlsson et al., 2017), (Yiying et al. 2009)

3.3. Identification/quantification

The identification and quantification of MPs often begins with their isolation from non-plastic particles based on physical properties that are unique to plastics. The process of identification based on physical properties such as morphology and color are called visual inspection or visual sorting. This approach - conducted using a microscope - has proven rather ineffective, as reports exist of over or underestimation (Nel et al. 2021a; Cheung et al. 2016; Lenz et al. 2015; Löder et al. 2015). For example, in one reported instance, 32% of particles and 25% of fibers were wrongly identified using visual inspection (Lenz et al., 2015). Another study, reported that 20% of particles identified as MPs by visual sorting, were later identified as aluminum silicate after viewing with a scanning electronic microscope (Eriksen et al. 2013). Other more effective approaches have been reported. These include: Raman spectroscopy

(Käppler et al. 2016), FTIR (Peng et al. 2017), scanning electron microscopy (Eriksen et al. 2013), pyrolysis-gas chromatography-mass spectrometry (PyroGC-MS) (Fischer and Scholz-Böttcher, 2017) and thermal desorption gas chromatography-mass spectrometry (TD-GC/MS) (Altmann et al. 2019).

3.3.1 Raman Spectroscopy

Raman spectroscopy is non-destructive and can be used for both identification and quantification of MPs (Hanvey et al. 2017). It has been used successfully to identify and quantify MPs down to as small as 1 µm (Pan et al., 2019, Lenz et al., 2015). Other advantages of this method include the ability to generate information on the chemical composition of the MPs and other associated organic and inorganic matter (Klein et al. 2018).

3.3.2 Fourier Transform Infrared Spectroscopy (FTIR)

According to Hanvey et al. (2017b), 23 out of 43 studies reviewed used FTIR. The use of FTIR has also been reported in other forms such as micro-FTIR (Vianello et al. 2013) which is basically a combination of optical microscopy and FTIR and attenuated total reflectance (ATR) FTIR (Cheung et al. 2016). The advantages of this method include its high throughput efficiency and non-destructive nature. It is however limited to measuring particles of sizes around 25 µm and thickness < 100 µm. There is also the possibility of an overlap of polymer bands which make it difficult to distinguish between identified polymers (Käppler et al. 2016; Käppler et al. 2018).

3.3.3 Scanning Electron Microscopy (SEM)

The use of scanning electron microscopy for identification of MPs has gained popularity for several reasons such as its ability to provide clear images of vital physical properties of the particle (Hanvey et al. 2017). It is good for showing physical degradation such as pits and cracks among other advantages and has been reported to be useful in differentiating between MPs and other organic/inorganic components (Crawford and Quinn 2017). It has also been combined with energy-dispersive X-ray spectroscopy (SEM-EDS) to examine the composition as well as additive content of plastics (Crawford and Quinn 2017; Fries et al. 2013). The laborious prepar-atory processes involved in using this method is however a major disadvantage (Wang et al., 2017).

3.3.4 Pyrolysis-gas chromatography - mass spectrometry (PyroGC-MS)

The basic principle of this method involves thermal decomposition of the particle of interest, followed by identification of the cryo-trapped evolved gases using mass spectrometry (Dümichen et al. 2017; Shim et al. 2017). This method is not only useful in identifying MPs but very effective for determining the polymer type, as well as associated organic matter (Mai et al. 2018). However, it is time consuming and destructive (Mai et al., 2018), and has difficulty correctly identifying polymers with polar sub-units (Dekiff et al. 2014a; Fries et al. 2013; Käppler et al. 2018).

3.3.5 Thermogravimetric Analysis coupled with Thermal desorption gas chromatography with mass spectrometric detection (TGA-TD-GC/MS)

TGA-TD-GC/MS works basically by interfacing thermo-gravimetric analysis connected to a solid-phase sorbent, with thermal desorption GC/MS (Dümichen et al. 2017; Elert et al. 2017). It has been successfully applied to measure MPs with the notable advantages of being more efficient even with complex samples. However, the method is destructive and time consuming (Dümichen et al. 2015).

3.4 Concerns about identification/quantification of MPs

Following the identification and quantification (sometimes by counting or weighing) of MPs using the previously discussed procedures, the results are often presented in varying units. A major concern is the lack of harmonization in the units of previous reports on the quantity of MPs in sediment (Hanvey et al. 2017). As a way of checking if this trend has changed since the Hanvey review, we conducted a search of the ISI web of knowledge database with the keywords "sediment" and "microplastics", restricted to papers published in 2019. This revealed that the inconsistency in result reporting remains a concern, although there are tentative indications of a movement towards a consensus on the use of items or particles per kg of dried sediment. A total of 38 publications met our search criteria and of these, only 7 quantified the measured MPs in terms of some form of units. Four of these reported the concentrations of MPs in items or particles per kg or g dry sediment (Mu et al. 2019; Yuan et al. 2019; Zhang et al. 2019); two reported as particles or items per kg (without specifying whether dry weight or wet weight of sediment) (Bordós et al. 2019; Li et al. 2019), while the remaining study reported as items/km of river (Xiong et al. 2019). It is recommended that items per kg be adopted as a standard as some recent studies have further classified MPs into fibres, particles, fragments,

filaments, microbeads, foams etc. based on their shape (Bertoldi et al. 2021; Kumar et al. 2021; Sang et al. 2021). Also, the use of both weight and volume is recommended where possible.

Another concern from the foregoing is the cost and efficiency of the methods discussed above. The need for "a cheaper, faster and more easily applied method" is glaring, especially if sufficient data from lower income regions of the world are to be obtained in order to establish temporal and spatial trends on a global scale (Maes et al. 2017). One positive step in this direction, as suggested by (Andrady 2011) is the use of lipophilic fluorescent dyes such as Nile Red to enhance visual identification/quantification of MPs under microscopes. Nile Red is a solvatochromic fluorescent dye that fluoresces with varying emission spectrum depending on the polarity of its environment (Maes et al., 2017). It has been applied successfully to the identification and quantification of microplastics in environmental samples (Nel et al. 2020; Maes et al., 2017, Erni-Cassola et al., 2017, Shim et al., 2016) and can cut analysis time down by 30–58% compared to the analysis of unstained samples (Nel et al. 2021a). Overall, this approach in combination with fluorescence microscopy and image analysis software is - according to Erni-Cassola et al. (2017) - a fast, reliable and cost-effective method of detecting, quantifying and sizing polyethylene (PE), polypropylene (PP), polystyrene **(**PS), and Nylon-6 particles in the size range of 20 µm to 1 mm. Shim et al. (2016) described the Nile Red staining approach as "straight forward and quick for identifying/quantifying polymer particles in laboratory-controlled samples" and reported a recovery rate of 98% for PE (100–300 µm). In comparing the recovery rates of LDPE particles (in the size range of 100–300 µm) using the Nile Red staining method and FTIR, Shim et al. (2016) reported 96±7% for FTIR and 98±3% for the Nile Red method. Furthermore, the Nile Red method gave on average a 1.4 times higher particle count than FTIR, following comparison of the quantitative analysis of field samples using both methods (Shim et al., 2016). This shows that the fluorescent dye staining method, which is a lot cheaper and faster, is also efficient. Despite its limitation of staining other non-plastic organic materials, it provides (among other advantages) a good solution to the difficulty in visual identification of transparent and white MPs (Shim et al., 2016). It is also highly unlikely that Nile Red would pose any risk of secondary contamination or interference with further analysis of additives present in the MPs (e.g., phthalates or OPEs) (Ramirez et al. 2019).

3.5 Quality assurance/quality control (QA/QC)

Considering the ubiquity of MPs, it is extremely important to put measures in place to prevent contamination of samples and ensure data accuracy. It is also important to guard against over- and under-estimation (Dekiff et al. 2014). The level of such accuracy largely depends on the

extent to which standard QA/QC measures are adhered to (Ibe and Kullenberg 1995). Rinsing all apparatus 3 or 4 times with purified water (Wen et al. 2018, Jiang et al. 2018, Yuan et al. 2019b) and then cleaning the workspace with 70% alcohol (Wen et al, 2018) has been reported. A strict use of 100% cotton lab coats as well as nitrile gloves, in addition to ensuring that all equipment and glass containers are covered by aluminum foil has also been reported (Wang et al., 2019). Some researchers pre-filter all liquids before use (Wang et al, 2019, Yuan et al 2019). Other quality assurance and quality control measures that are recommended include validation studies and procedural blanks. Validation studies are laboratory tests performed to verify that a technique or procedure is accurate and reproducible, specifically highlighting any limitations. Hanvey et al., (2017) reported that only 7 out of the 43 reviewed studies conducted some form of validation test. Some more recent studies have used procedural blanks as QA/QC measures (Wang et al., 2019; Yuan et al., 2019b, Jiang et al., 2018). Other steps that have been undertaken in previous studies to ensure the integrity of the results of MPs studies, include the use of non-plastic sampling, storage, and processing tools (Hanke et al. 2013). It is recommended that publishers insist on proper QA/QC measures as a condition for publication as this will ensure the integrity of reported data. Also, something similar to the work of Andrade et al., (2020) that proposed a minimum information for publication of IR-related data on MPs characterization (Andrade et al., 2020) is strongly recommended for the study of MPs in sediments.

3.6 Possible effect of MP extraction methods on the analysis of associated additives

The bioavailability of additives such as OPEs that are present in sediments, will very likely vary substantially depending on whether they are sorbed to MPs or to naturally occurring sediment organic matter. It is therefore important to be able to differentiate between OPEs bound to MPs present in sediment and those associated with the sediment itself. Moreover, in order to achieve such differentiation, it is crucial to understand the impact exerted on the determination of OPEs by the procedures used to isolate MPs from sediments - e.g. will the use of Nile Red dye chemically affect OPEs and/or introduce chemical interferences? Although not much has been done in freshwater environments, a few studies have looked at the presence of plastic additives in MPs isolated from marine environments (see Fries et al. 2013; Hirai et al. 2011; Rani et al. 2015) each identified MP types and associated organic plastic additives (OPAs) following their extraction from sediments using the density separation approach. QA/QC measures such as the use of metal spoons, covering all materials with aluminium foil after each step and rinsing of equipment with ultra-pure water were reported, but no mention

464 was made of the possible effect of MP extraction procedures on the analysis of OPEs associated
465 with MPs. In view of this, we have examined a cross section of recent studies on MP extraction
466 from sediments with a view to identifying possible sources of OPE contamination to isolated
467 MPs subsequently analyzed for these and/or related additives (Table 4). The table also presents
468 a cross section of techniques adopted by some researchers for identification and quantification
469 of MPs.

470 **Table 4**: **Examples of procedures adopted to measure MPs in sediment and possible impacts on the analysis of OPEs and/or**
471 **related MP additives.**

Reference	Sampling method	Extraction method	Clean-up	Identification	QA/QC	Possible source of OPEs/other additives
(Wen et al. 2018)	A shovel was used to collect samples into aluminum foil and kept at 5 °C.	Density Separation	Digestion with 30% H_2O_2 and Fe (II) solution (catalyst)	micro-Raman spectroscopy	The workspace was cleaned with 70% alcohol before. All pieces of apparatus were rinsed three times with distilled water and covered with aluminum foil. Blank tests were used.	No obvious source was identified but it is highly recommended that procedural blanks are incorporated as there is a possibility of contamination from indoor air.
(He et al. 2018; Liu et al. 2018; Zhou et al. 2018)	Clean stainless-steel shovels and spoons were used.	Air flotation and density separation using saturated sodium chloride solution	Use of 30% H_2O_2 for 72 h at 50 °C was the most favored digestion method.	Visual identification under an optical microscope and confirmation of MPs using micro-Fourier transform infrared (μ-FT-IR) and Raman spectroscopy	All materials thoroughly cleaned and covered with aluminum foil. Lab coats always worn during analysis. Blank tests conducted for MPs.	All studies only used blanks to monitor background contamination by MPs. A total of 33 fibers were extracted from one of the blanks and another study excluded fibers from the study entirely as they acknowledged that atmospheric contamination of samples with fibers from the air was unavoidable.

Jiang et al., 2018	Sediment (0–2 cm) from lakeshore line was collected with a stainless-steel shovel.	Density separation was used	H_2O_2 (30%, v/v) catalyzed with ferrous sulfate.	microscopic identification and Raman Spectroscopy	All equipment was pre-cleaned three times by ultrapure water and wrapped in aluminum foil when not in use. Blank tests were used to determine the background values of contamination of MPs from the laboratory	No obvious source was identified but it is strongly recommended procedural blanks be incorporated as there is a possibility of contamination from indoor air.
Wang et al., 2019	For surface sediments stainless-steel shovel was used. A plexiglass tube was used for vertical core.	Sediments were dried at 60 °C overnight. NaI solution used followed by sieving with a steel sieve The process was repeated three times.	30% H_2O_2 and 65% HNO_3 (1:3, v/v) for two days	FTIR analysis SEM	All liquids used were filtered with 0.45 μm membrane filters All glass containers were covered by aluminum foil, and a cotton laboratory coat and nitrile gloves were worn. Moreover, the entire process of analysis and identification was conducted by one person in a closed room, with blank experiments conducted to detect any ambient microplastic contamination	No obvious source was identified but it is highly recommended procedural blanks be incorporated as there is a possibility of contamination from indoor air.
(Yuan et al., 2019b)	Sediment was collected	A density separation was employed	Digestion using 30%	Visual identification as well as	Cleaning of tools with filtered pure water before sampling.	No obvious source was identified but it is highly recommended procedural

	from boat using a Van Veen grab sampler		H_2O_2 overnight. The digested solution was diluted and filtered using a 0.45 µm filter.	Stereoscopic and Raman analyses were employed for identification/ Quantification	Nitrile gloves and a cotton lab gown worn All solutions filtered through a 0.45 µm filter before use Method blank tests were carried out in the laboratory. The experiment was repeated three times.	blanks be incorporated as there is a possibility of contamination from indoor air.

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4. Temporal trends of MPs concentrations in freshwater sediment

Freshwater sediments may serve as long-term or temporary MP sinks (Ballent et al., 2016, Corcoran et al., 2015, Nel et al., 2018). The first published study on MPs in lake sediments, reported concentrations of 1,108 ± 983 particles/m^2 in beach sediments from the sub-alpine Lake Garda in Italy (Imhof et al., 2013). MP contamination of lake sediments is now widely documented (Carril-Ajuria et al. 2020; Gopinath et al. 2020; Hu et al. 2020; Tavşanoğlu et al. 2020). However, reliable data on temporal trends is scarce, with most studies reporting concentrations from a single moment in time (Tavşanoğlu et al. 2020; Wu et al. 2020). In order to fill this data gap long-term studies designed to capture the temporal variations in MP concentration are required. Additionally, sediment cores can be used to measure historical temporal trends. Radiometrically-dated sediment cores have been used by Turner et al. (2019) and Matsuguma et al., (2017) to investigate the historical change in MP concentrations (Matsuguma et al. 2017; Turner et al. 2019). Matsuguma et al. (2017) analyzed dated sediment cores collected from a range of water bodies (e.g., canals, lakes, coastal zones, harbors) from Japan, Thailand, Malaysia, and South Africa. Results showed a clear increasing trend, with higher MP concentrations found associated with more recent sediment layers. A correlation between plastic production and MP concentration was observed within the Asian and African samples.

Turner et al. (2019) traced the onset of MP pollution to the 1950s, which coincides with the period when large-scale production and use of plastics commenced. The Turner et al. (2019) study also reported a correlation between increased plastic fiber concentrations and global annual plastic production figures as well as reporting that identifiable concentrations of polystyrene emerged only after the onset of its production and use. These interesting correlations and trends pose questions as to whether similar trends can be identified with plastic additives.

5. Temporal trends of organophosphate esters in freshwater sediments

The occurrence of OPEs in sediments have been reported by several studies (Castro-Jiménez and Ratola 2020; Liao et al. 2020; Ren et al. 2019; Wang et al. 2018) and in one such study, ∑OPE concentrations were lowest in the oldest sediment layers but increased gradually towards the more recent sediment layers. Although the sediments were not dated, there appears

to be some degree of correlation with the global production and usage of OPEs (Liao et al. 2020). Similarly, in a study that investigated the concentration of OPEs in sediment cores and surface sediments from the Great Lakes, Cao et al. (2017) identified a possible correlation between the trend in sediment deposition of OPEs and the production/regulation trend over the period of interest. One good example of such a trend was the observed increase of the concentration of OPEs in sediment core layers dated after 2000 which coincides with the period during which worldwide production volume of OPEs tripled as a result of large-scale replacement of PBDEs with OPEs in consumer goods (Iqbal et al. 2017). It was also observed that the replacement of TCEP with TCIPP after the former was phased out, might be responsible for the exponential increase in the concentration of TCIPP in more recent Great Lakes sediment layers (Cao et al. 2017). The same study reported similar concentrations of OPEs as those reported in sediment in other parts of the world such as China (Cao et al. 2012) and Austria (Martínez-Carballo et al. 2007). Combined, such studies highlight increasing risks of ecotoxicity because of environmental pollution with OPEs that is increasing at a similar rate to that of MPs. To illustrate, a study by Cho et al. (1996) has traced the increase in OPE concentrations in a rural river (Kurose River, Japan) to the leaching of tri-cresyl phosphate (TCP) isomers from agricultural plastic films.

7. Current research gaps and future perspectives

This review reveals increasing interest in the study of MPs in the freshwater environment and an urgent need to review the current trend in plastic production and work towards improved recycling methods. This is particularly important because a consistency between the trend of MP abundance in the environment and the global trend of plastic production has been reported (Matsuguma et al. 2017). In fact, Turner et al. (2019) reported a relatively low number of MPs in older sediment cores compared to earlier ones. It is also important to extend understanding of MP pollution to other parts of the world such as Africa, where only 12 studies appear to have been carried out between January 2000 and June 2021 notwithstanding the fact that some of the world's largest freshwater bodies are in Africa.

It is also clear that there are several research gaps in the study of MPs, especially in freshwater environments (Table 2). Beyond the lack of standardized procedures in sediment sampling, there is limited evidence of the comparability between the various sampling methods. This can

be addressed by investigating the possible differences between MP concentrations using the different sampling methods in a controlled environment.

Research into effective MPs extraction and detection methods with focus on standardized measurement and reporting procedures is encouraged. Further development and standardization of simple, cheap, high throughput and accurate methods like those based on fluorescence dye staining is also recommended to facilitate relevant studies in low-income and developing countries. Furthermore, international comparison and validation studies are required to ascertain the quality of data provided, as well as the comparability of results from different labs around the world. Development and production of laboratory standard reference materials for MPs in sediment is the ultimate aim for achieving a standardized QA/QC protocol for MPs analysis in sediment.

The role of MPs as conduits of chemical pollutants, widely used as plastic additives, to the freshwater environment is poorly understood. While there seems to be an apparent correlation between concentrations of MPs and chemical pollution in the few studies that have addressed this topic, it is not clear if MPs act as a source or a sink of lipophilic chemicals to the aquatic environment. In particular, nothing is known about the partitioning of chemical plastics additives in sediments between sediment particles and the MPs themselves. Thus, discerning the impacts of MPs from those of the additives present, is an important research goal. Given the potential implications of this for the bioavailability of such chemical additives, this is a significant research gap that should be addressed. Specifically, the impact on additive bioavailability of several factors including the type of plastic polymer, the concentration and physicochemical properties of chemical additives, the organic content of sediment and the temperature, is unknown. Studies are urgently required to understand the fate and behavior of hazardous chemical additives present in MPs and the factors influencing their release to the freshwater environment. This also means that while working on biodegradable plastics and other ways of promoting “Reduction, Re-use and Recycling” of plastics; sources of MP pollution and the use of chemical additives should be carefully considered because the facilitated degradation of these biodegradable plastics may provide a further source of chemical pollution to the environment.

Finally, more studies on temporal/spatial trends of MPs (and associated additives such as OPEs) in radiometrically-dated freshwater sediment cores are required, not only to provide

information on historical trends but also to enable modelling and prediction of future trends and associated risks to the aquatic environment. This will also provide valuable information for shaping pollution control strategies and the effect of hydrodynamic conditions on the distribution of MPs in freshwater systems.

8.0 Conclusion

Following a critical review of the current state-of-knowledge on MPs in freshwater sediment, certain trends such as increased interest in freshwater sediments as well as a consistency between the trend of microplastic abundance in the environment and the global trend of plastic production were observed and the need to work towards bridging research gaps in key areas was highlighted. These research gaps include; developing and producing standard reference materials for MPs in sediment, understanding the role of MPs as conduits of chemical pollutants, understanding partitioning of chemical plastics additives in sediments between sediment particles and the MPs themselves and the impact on additive bioavailability of several factors including the type of plastic polymer, the concentration and physicochemical properties of chemical additives, the organic content of sediment and the temperature.

Methods for the analysis of MPs in freshwater sediment was reviewed and although no obvious impact on the analysis of OPEs and/or related MP additives was seen, it is highly recommended that QA/QC measures such as procedural blanks be taken to prevent secondary introduction of some of the target plastic additives.

Declaration of competing interest

The authors declare that they have no known competing interests.

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Supporting Information.

Details including chemical names, structures, CAS numbers, molecular formulae, applications, environmental/health hazards of OPEs are provided in the supporting information.

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Declaration of interests

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

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