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A temporal sediment record of microplastics in an urban lake, London, UK

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Abstract A radionuclide-dated (^{210}Pb and ^{137}Cs) sediment core collected from Hampstead Pond No. 1, a North London lake, was used to provide novel data on the historical accumulation of microplastic waste in the urban environment. Microplastics were extracted from sediments by sieving and dense-liquid separation. Fibres of anthropogenic origin dominated the assemblage. Microplastics were first identified by microscopy before Raman spectroscopy of selected particles was used to determine the composition of synthetic polymers and dyes. Polystyrene microplastic particles were identified, in addition to synthetic fibres of polyacrylonitrile, polyvinyl chloride and fibres containing synthetic dyes. Concentrations of total microplastics in the sediment samples ranged from detection level to 539 particles per kilogram of dried sediment. Proliferation of microplastics is evident in the core from the late 1950s to the present. Relatively low numbers of particles were found in older sediments, comparable to laboratory blanks, highlighting the difficulty of extending a plastic chronostratigraphy back to the early twentieth century. This study shows that, with optimisation, routine extraction of microplastics from radionuclide-dated lake sediments can add an important temporal perspective to our understanding of microplastics in aquatic systems.

Keywords Microplastics · Freshwater · Lead-210 · Sediment · Plastic pollution

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Introduction

The accumulation of plastic waste in freshwater aquatic systems has become a relatively recent focus of research (Moore et al. 2011; Zbyszewski and Corcoran 2011; Wagner et al. 2014; Eerkes-Medrano et al. 2015), despite its high visibility, potential for association with other contaminants (Rochman et al. 2013) and known resistance to degradation. Microplastics (including synthetic fibres) < 5 mm in size are of interest due to the likelihood of ingestion by freshwater organisms and, because they have large specific surface area, their potential for adsorption and leaching of associated contaminants and additives (Lobelle and Cunliffe 2011; Sanchez et al. 2014; Faure

et al. 2015; Näkki et al. 2017). Pellets and 'microbeads' specifically manufactured as microplastic forms (primary microplastics) may be found in freshwaters subject to large inputs of industrial drainage or effluent (Castañeda et al. 2014; Lechner et al. 2014; Corcoran et al. 2015; Driedger et al. 2015; Baldwin et al. 2016; Horton et al. 2017a; Horton and Dixon 2018), although secondary microplastics (derived from the breakdown of larger plastic items) dominate in freshwater sediments subject to more diffuse plastic waste sources (Free et al. 2014; Driedger et al. 2015; Faure et al. 2015; Zhang et al. 2016; Blettler et al. 2017; Matsuguma et al. 2017; Sruthy and Ramasamy 2017; Imhof et al. 2018).

In the absence of long-term monitoring of plastic waste (Barnes et al. 2009), analysis of environmental archives capable of capturing and accumulating plastic, should enable retrospective assessment of the occurrence, usage patterns and changing types of plastics through time. Plastics are highly persistent, with many estimated to take hundreds of years to degrade, and are therefore likely to accumulate in sediment deposits (Andrady 2003; Barnes et al. 2009). To understand previous and current amounts and varieties of plastics in the environment, a temporal perspective is invaluable. Unlike other global contaminants, however, plastic waste accumulation has not, until now, been analysed within the context of a well-constrained lake sediment sequence.

Lakes provide excellent depositional contexts for recording the transport of contaminants within and through catchments (Boyle 2001; Engstrom and Rose 2013). Especially valuable in the historical framework of plastic waste (Zalasiewicz et al. 2016) is the presence of well-defined, datable sediments over the last ca. 150 years that, compared to marine cores (Woodall et al. 2014; Martin et al. 2017), are more accessible and proximal to terrestrial sources of plastic contamination. Lakes therefore represent catchment-scale sinks for microplastic debris compared with oceans, which are subject to global-scale long-range transport from multiple catchments (Hidalgo-Ruz et al. 2012; Hardesty et al. 2017). The volume and composition of plastic waste found at one point in time and space in an aquatic system, will comprise a cumulative component, as a consequence of temporary storage and reworking in the catchment. Benthic sediment is an end member of microplastic transport in freshwater systems, but more research is required to

quantify transport pathways, burial and biological interactions within lacustrine systems.

Microplastics have been found in lake sediments, with inputs linked to urbanization, industrial activities and wastewater influences (Castañeda et al. 2014; Corcoran et al. 2015; Driedger et al. 2015; Su et al. 2016; Blettler et al. 2017), but high-resolution sediment evidence of the historical incorporation of microplastics in lake sediments, indeed in any co-dated or cross-correlated aquatic/estuarine sediment, is surprisingly rare (Cundy et al. 1998; Corcoran et al. 2015; Matsuguma et al. 2017; Willis et al. 2017). Previous age/depth measurements of lake-sediment microplastic abundance were estimated either by comparison with historical trends of other contaminants or by correlation with other cores (Corcoran et al. 2015; Matsuguma et al. 2017). In freshwater environments, however, variations in sediment accumulation affect burial of microplastics, and so historical records must be reliably dated to quantify rates of incorporation in benthic sediments.

This study aimed to:

a. Obtain the first record of microplastics in lake sediment core samples dated with radionuclides (^{210}Pb, ^{137}Cs) and co-measured with paleolimnological indicators of human activity.
b. Provide a first look at the changing composition and abundance of microplastic debris in lake sediments that span the twentieth century to present day.
c. Provide a straightforward method for separation of microplastics from lake sediment matrices that can be added to the paleolimnological toolset.

This paper provides a significant contribution to our knowledge of sources, transport pathways and fate of plastic debris in freshwater environments (Schmidt et al. 2017). The data are further evidence of the ubiquity of microplastics in global environments and contribute to the discussion on plastic waste as a stratigraphic marker for the Anthropocene (Zalasiewicz et al. 2016).

Study site

Hampstead No.1 Pond is a small water body (1.5 ha, max 3 m depth) in North London, United Kingdom (Lat 51.5575°, Long − 0.1655°, 69 m above sea level). The pond was created, possibly as early as the

late seventeenth century AD, by impoundment of a stream that drains the southern slopes of Hampstead Heath. The small catchment (0.7 km^2) consists of open parkland, woodland, and housing (late-nineteenth-century stock) with gardens. The pond is the lowest in a chain of three water bodies, separated by embankments and connected by drains and culverts. In the upper part of the catchment, some sections of streams are open and in semi-natural woodland. The open parkland west of the pond was used historically for seasonal fairs. Public swimming and coarse fishing occur in the two ponds upstream.

Materials and methods

Sediment analysis and chronological framework

A 6-cm-internal-diameter piston corer was used to collect a 212-cm sediment core (HAMP1) in 2.7 m water depth in June 2009 (Hall 2013). The core was extruded for multi-proxy analyses at 1-cm intervals. The core was extruded and sliced in the field using a stainless-steel blade and 1-cm-thick ring of core tube that captured mud during vertical extrusion at measured 1-cm intervals. These tools were rinsed with tap water between collection of core slices, which were stored in individual twist-tie PVC bags. The core was measured for ^{210}Pb, ^{226}Ra, ^{137}Cs and ^{241}Am activities (Electronic Supplementary Material [ESM] 1) by direct gamma assay, using an ORTEC HPGe GWL series well-type coaxial low-background intrinsic germanium detector (Appleby et al. 1986). ^{210}Pb activity was determined via its gamma emissions at 46.5 keV, and ^{226}Ra by 295 keV and 352 keV gamma emissions from its daughter isotope ^{214}Pb, following 3 weeks in air-tight containers to allow radioactive equilibration. ^{137}Cs and ^{241}Am were determined by their emissions at 662 keV and 59.5 keV, respectively. Corrections were made for the effect of self-absorption of low-energy gamma rays within the sample. The core chronology was calculated using the constant rate of supply (CRS) ^{210}Pb dating model (Appleby 2001), with reference to the 1963 fallout maximum of ^{137}Cs and ^{241}Am derived from nuclear bomb tests (Appleby et al. 1986). Radionuclide dating of the core was complemented by a spheroidal carbonaceous particle (SCPs) chronology. SCP

analysis followed Rose (1994), with identification using criteria described in Rose (2008).

Organic matter content was estimated on contiguous 1-cm samples by weight loss following heating (loss on ignition, LOI) at 550 °C for 2 h (Heiri et al. 2001). Downcore wet density measurements at 2-cm intervals were completed by weighing a 2-cm^3 vial of wet sediment. Sediment samples (contiguous 1-cm samples, 0–20 cm; 2-cm intervals 20–212 cm) were analysed for element abundances using a Spectro XLAB2000 X-ray fluorescence (XRF) spectrometer. 1 g d.w. (weighed to 4 d.p.) of freeze-dried and milled sediment was placed in nylon cups with a base of prolene foil (4 μm thickness). Two reference sediment samples, JLK-1 (Imai et al. 1996), of the same mass were measured every 9th sample, to identify instrument drift error and assess measurement accuracy. Recovery rates for elements Ti (% d.w.), Zn and Pb ($\mu g \ g^{-1}$ d.w.) in this study, were 112.0%, 97.9% and 113.8%, respectively (Hall 2013).

Microplastic extraction

Prior experience with extracting low numbers of microplastic particles (< 20 per 100 g d.w.) from littoral sediments of an urban UK lake (Vaughan et al. 2017), and small volumes of mud as a consequence of HAMP1 having been used for multiple analyses, required 1-cm core intervals be combined into 5-cm depth intervals (Table 1). Combined sediment samples were mixed before oven drying (48 h, 40 °C) for dry mass calculation. Dried sample masses ranged from 20 to 90 g dry mass. The dried sample was disaggregated with deionised water and washed through 1-mm and 500-μm stainless steel sieves, again with deionised water. The > 1-mm and 1-mm to 500-μm fractions retained from each sample were collected for analysis. For two samples (ESM 2) the < 500-μm fraction was also retained to check microplastic presence in this finer fraction. The focus on the 1-mm to 500-μm size range was selected to remove fine silts and clays, while retaining easily visible microplastics for microscopic assessment and Raman spectroscopy (Horton et al. 2017a; Vaughan et al. 2017).

Ten mL of sodium polytungstate (SPT) solution (2.1 g cm^{-3} density) was added to a 50-mL centrifuge tube. This solution was used, as most common polymers remain buoyant at this density

Table 1 Dated time intervals used for microplastic (MP) assessment

^{210}Pb dating and sedimentation rate				MP depths	MP time periods
Depth (cm)	Year AD	g cm^{-2} yr^{-1}	cm yr^{-1}		
0–1	2009 ± 2	0.1827	1.261	0–5[a]	2009–2005
				5–10[a]	2005–2000
10–11	2000 ± 2	0.167	0.951	10–15[a]	Mid-late 1990s
				15–20[a]	Early-Mid 1990s
20–21	1989 ± 3	0.1838	0.942	20–25[a]	Mid-late 1980s
				25–30[a]	Mid-early 1980s
30–31	1978 ± 4	0.1554	0.761	30–35[a]	Late 1970s–1960s
				35–40[a]	Late 1960s–early 1960s
40–41	1963 ± 5	0.1466	0.627	40–45[a]	Early 1960s–mid 1950s
				45–50[a]	Mid 1950s to 1940s
50–51	1935 ± 15	0.0486	0.193	50–55	1940s–1930s
				55–60	1930s
70–75				70–75	Early twentieth century
80–85				80–85	Late nineteenth century
90–95				90–95	∼ Nineteenth century

[a]Samples analysed by Raman spectroscopy

(Scientificpolymer.com 2013; Nuelle et al. 2014) except polytetrafluoroethylenes (PTFE) and some composites. Solutions rinsed from the sieves with deionised water were gently poured on top of the SPT solution. The 50-mL tubes were capped and centrifuged at 1500 rpm for 5 min. Each lower-density fraction, containing the microplastics, was pipetted off and vacuum-filtered through a 0.45-μm micropore filter paper. Filter papers were covered by a watch glass and oven-dried at 40 °C before microscopic inspection. Sieves, pipettes and centrifuge tubes (following SPT treatment) were rinsed (3 ×) with deionized water to prevent particles from adhering to surfaces (Hidalgo-Ruz et al. 2012). Between samples, all sieves and vacuum filter equipment were washed and backwashed three times. Blank samples of deionised water were also passed through the sieves and floated on top of SPT before being transferred to vacuum filter papers every 5th sample to check for contamination (ESM 2).

The filter papers were examined at up to 40 × magnification with a stereomicroscope (Leica M60, Germany), with selected particles transferred by stainless steel forceps into covered, white cardboard trays for storage. Microplastics were distinguishable and selected using the following criteria (Hidalgo-Ruz et al. 2012; Nor and Obbard 2014; Horton et al. 2017b): (1) bright/unnatural coloured granules, films

and fibres; (2) granules, films and fibres not visibly organic or cellular and (3) microparticles selected by the above criteria that maintain integrity when gently stretched and are elastic when squeezed with forceps. Fibres were inspected for changing thickness along their length and any branching that would suggest organic origin (hairs, root/plant fibres). If branching was present, such fibres were rejected. Particle colour was noted and the longest dimension measured using a 1-mm gridded sheet or microscope graticule. Fibres were stretched by forceps to measure maximum length. Plant root/leaf fibres were distinguishable by their brown/woody colour, branching features and low tensile strength.

The effectiveness of the method was assessed by visual inspection of microplastics present in the > 1-mm and < 500-μm sieved fractions and also in the remaining, > 2.1 g cm^{-3} solution, following SPT flotation and centrifuging. This was to check for presence of macroplastics in the coarser sediment matrix, < 500-μm microplastics, and whether any dense microplastic particles had sunk during density separation. The > 1-mm and < 500-μm sieved fractions of each sample were rinsed with deionised water, SPT-treated and vacuum-filtered through 0.45-μm filter papers and oven-dried before inspection and particle selection as described above. Denser sediment and SPT solution remaining in the centrifuge tube was

rinsed with deionised water, vacuum-filtered and dried as above for each sample. The few microplastics in the > 1-mm sieve fraction were added to the sample counts of the 1-mm to 500-μm fraction. Microplastic counts in the < 500-μm fraction of two samples (0–5 cm and 35–40 cm) and in the post-centrifuging SPT solution (ESM 2 and 3) were not included in calculations or Raman composition assessment. Concentrations of particles were calculated as numbers of particles per dry mass of sediment analysed (n particles kg^{-1}). Microplastic accumulation rates (n m^{-2} yr^{-1}) were calculated by multiplying concentration by the ^{210}Pb-derived sediment accumulation rate (Table 1).

Contamination control and blank processing

Incorporation of contaminant microplastics into sediment samples during laboratory processing (Willis et al. 2017) is of particular concern for stratigraphic studies. Potential microplastic contamination can occur during core collection (from clothing, ropes, atmospheric deposition) and laboratory processing. Core HAMP1 was collected using protocols for paleoenvironmental and trace metal work that employ clean materials during core slicing and avoid sediment transported downward during core collection and extrusion. In the laboratory, a white cotton coat was worn to avoid synthetic clothing to sample transfer. Metal sieves were washed and back-washed in deionized water prior to use and kept covered when not in use. The vacuum filter flask (moulded transparent polysulfone, 'Nalgene$^{®}$') was similarly washed, before and between samples, with deionized water and covered while in use to avoid airborne dust.

Three blanks of deionized water were run as samples through the whole sieving, density separation and filtration process (ESM 2). The numbers of microplastics in the blanks were low (2 to 3 white, red or blue coloured fibres) and comparable to previous blank numbers run in the same laboratory (Vaughan et al. 2017). These low numbers are significant, however, given the presence of comparable numbers of fibres found in pre-twentieth century and younger HAMP1 sediments. Numbers of fibres in the sediment reported are therefore calculated minus the total number (+ 1 for caution) of fibre types found in the blanks (ESM 2). Other particles were not found in the blanks.

Microplastic composition

Only the upper 50 cm of the core was selected for Raman analysis, as plastic was expected to have only been deposited in significant amounts after the mid-twentieth century. Subsamples of particles taken from the > 1-mm and 1-mm to 500-μm fractions were analysed by Raman spectroscopy (HR800UV, Jobin–Yvon Horiba, with an integrated Olympus BX21 microscope). Particles were selected to represent the range of shapes and colours found in the sample. A minimum of 20% of the particles from each sample were analysed. Where highly diverse particle shapes and colours were present, more particles were analysed to give a better representation of particle types, up to a maximum of 66% particles from one sample (ESM 4).

Spectra were obtained using a near-infrared laser (785 nm). Acquisition time was 40 s and accumulation was set at 2×, with the range set to acquire between 200 and 3200 cm^{-1} to account for the entire polymer spectrum. Depending on particle colour, laser intensity was adjusted accordingly, to prevent particle damage by the laser. The spectra were analysed using the BioRad KnowItAll$^{®}$ Informatics System—Raman ID Expert (2015) software. This allowed automatic and manual matching of unknown spectra to known compounds within the Biorad KnowItAll$^{®}$ Raman database (containing > 24,000 known compounds). The most suitable match was identified based on corresponding peak positions. For a more detailed description of identification methods see Horton et al. (2017a).

Results

Core chronology

Unsupported ^{210}Pb and ^{137}Cs activities provide a reliable sediment chronology post 1935 ± 15 (50 cm depth). Between 1935 ± 15 (50 cm) and 1963 ± 5 (40 cm), sediment accumulation was low (0.0486 g cm^{-2} yr^{-1}; 0.193 cm yr^{-1}). Sediment accumulation since 1963 ± 5 has been relatively uniform, with a mean rate of 0.167 g cm^{-2} yr^{-1} (Table 1). Below 40 cm, particularly between 40 and 110 cm, there is little net decline in unsupported ^{210}Pb,

which may indicate some sediment mixing (ESM 1). Profiles of both LOI 550 and Ti concentrations (Fig. 1), however, show that if a hiatus or mixing occurred, the sediment sources and the depositional environment remained similar.

Assigned dates below 50 cm are derived from 1935 ± 15 sedimentation rates and historical records of SCP and trace metal concentrations in other ^{210}Pb-dated core sequences from London (Hall 2013). Very low concentrations of SCPs are found in HAMP1 below 100 cm depth, indicating a mid-late nineteenth-century age, but they increase above 90 cm, most likely signifying the late nineteenth- to twentieth-century expansion of small-scale power generation plants in London at that time (Rose and Appleby 2005). The concomitant increase in Zn and Pb in the core (Fig. 1) from the late-nineteenth century and their tracking of the increase in SCPs to their peak in the mid-1970s also supports evidence of historical lake-sediment trace-metal accumulation from fossil-fuel combustion in London (Hall 2013).

Microplastic abundance

The assemblage of microplastics recovered from HAMP1 core samples is dominated by fibres (40–100% of particles, variable by slice) (Table 2). Microplastic fragments were identified only in sediments younger than the late 1950s (45–40 cm). Fibres occur in samples down to 95 cm that match microplastic features identified in younger sediments by Raman spectroscopy. Fibres were found both entwined with particles of mud/organic matter and on their own. Many fibres were visibly twisted, knotted and showed signs of abrasion/tearing (Fig. 2). The smallest fibres observed were ∼ 100 µm. Calculation of the abundance of fibres in relation to numbers found in the associated blanks, show red, blue and white fibres (ESM 2) in sediments below 45 cm occur at near or non-detectable/below blank level (Fig. 1).

Microplastic concentration calculated for each depth/age interval assumes that microplastics are uniformly distributed through the whole core slice. Total microplastic maximum concentrations, equating
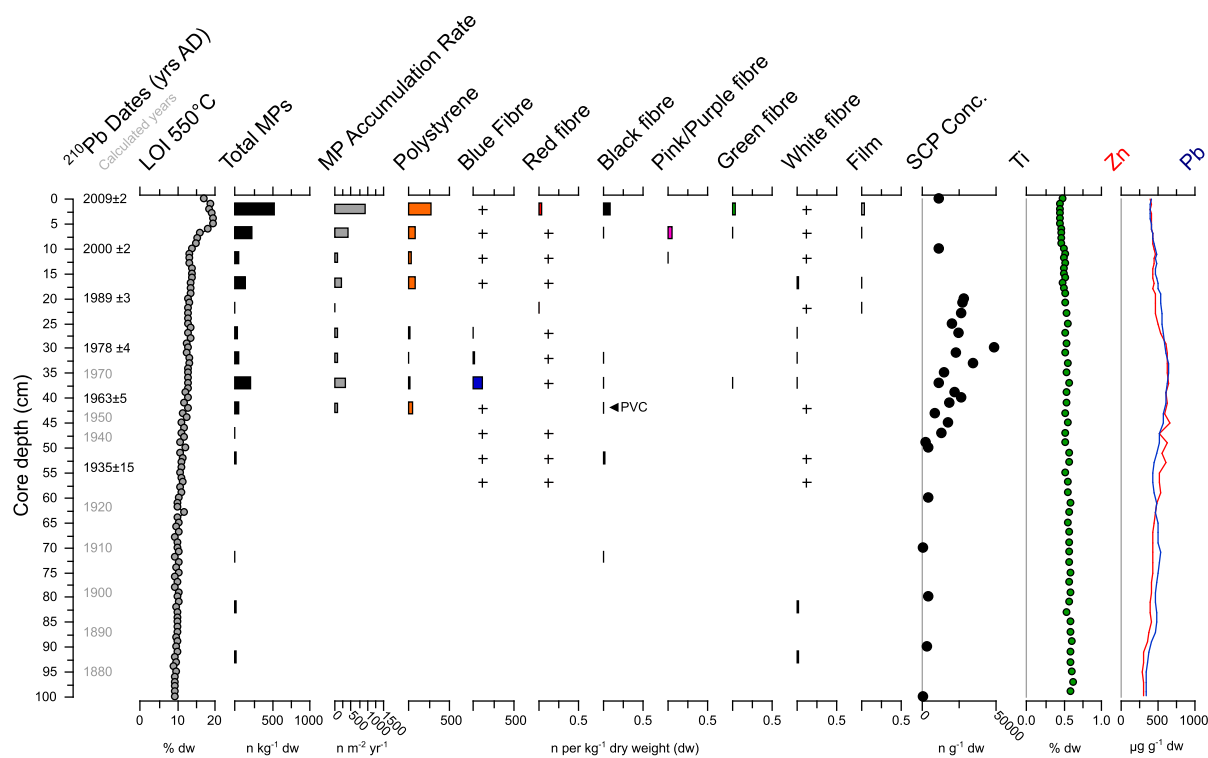

Fig. 1 Stratigraphic plot of ^{210}Pb-dated sediment variables and occurrence of microplastic (MP) types in core HAMP1. Crosses indicate below-blank o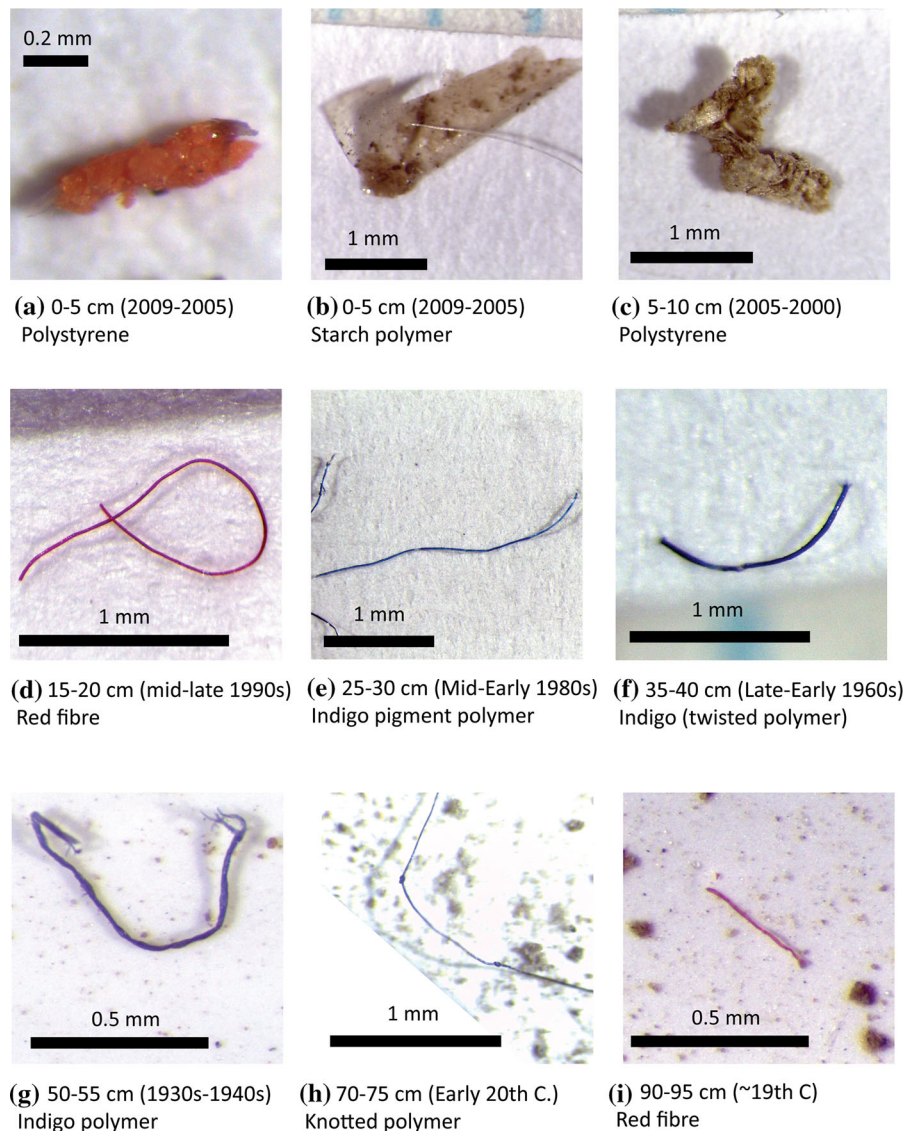ccurrence. LOI 550 °C (dry mass loss on ignition); SCP Conc (spheroidal carbonaceous particle concentration); Ti, Zn and Pb element abundance in sediment samples measured by XRF (see text)

Table 2 Colour and size classification summary of materials found in (a) 500-μm to 1-mm and (b) > 1-mm sieved fractions following SPT flotation. Fibres, measured length; Fragments*, longest axis. Total Microplastic (TMP) concentration includes all particles and synthetic fibres. TMP-blank concentrations (italics) calculated by subtracting fibre numbers found in blanks (ESM 2)

Time periods	2009–2005		2005–2000		Mid-late 1990s		Early-mid 1990s		Mid-late 1980s		Mid-early 1980s		Late 1970s–1960s	
Depth	0–5		5–10		10–15		15–20		20–25		25–30		30–35	
	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)
Fibres (n)														
Blue		3	1	1		1		1			2	2	5	1
Red	3	1	1	1	1		1	2	3	1		1	2	
White		1		2		1		4	1			3		3
Black		2		1										1
Green		1	1											
Pink/purple			1	2	1									
Fragments (n)														
Orange foam*	5	1	4		2		5				2		1	
Film*		1		1				1		1				
Dry mass (g)	20.4		41.2		46.7		53.5		64.0		67.8		76.7	
TMP (n)	18		16		6		14		6		10		13	
TMP minus blank	*11*		*10*		*3*		*8*		*2*		*4*		*6*	
TMP (n kg^{-1})	882.4		388.3		128.3		261.7		93.7		147.3		169.4	
TMP minus blank	*539.2*		*242.7*		*64.2*		*149.5*		*15.6*		*58.9*		*78.2*	

Time periods	Late 1960s–early 1960s		Early 1960s–mid 1950s		Mid 1950s—1940s		1940s–1930s		1930s		Early twentieth century		Late nineteenth century		~ Nineteenth century	
Depth	35–40		40–45		45–50		50–55		55–60		70–75		80–85		90–95	
	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)
Fibres (n)																
Blue	12	2	1			2	1		1							
Red	1										3			2	1	1
White	1	3		1			2			1				4	2	3
Black		2	1			1	2					1				
Green	1															
Pink/purple																
Fragments (n)																
Orange foam*	3		4													
Film*																
Dry mass (g)	83.9		70.4		77.7		56.5		60.3		71.1		72.3		89.9	
TMP (n)	25		7		3		5		2		4		6		7	
TMP minus blank	*19*		*5*		*1*		*2*		*0*		*1*		*2*		*3*	
TMP (n kg^{-1})	297.8		99.4		38.6		88.5		33.2		56.2		83.0		77.8	
TMP minus blank	*226.4*		*71*		*12.9*		*35.4*		*0*		*14.1*		*27.7*		*33.3*	

Fig. 2 Selected microplastic particles and fibres found in HAMP1 core

(a) 0-5 cm (2009-2005) Polystyrene

(b) 0-5 cm (2009-2005) Starch polymer

(c) 5-10 cm (2005-2000) Polystyrene

(d) 15-20 cm (mid-late 1990s) Red fibre

(e) 25-30 cm (Mid-Early 1980s) Indigo pigment polymer

(f) 35-40 cm (Late-Early 1960s) Indigo (twisted polymer)

(g) 50-55 cm (1930s-1940s) Indigo polymer

(h) 70-75 cm (Early 20th C.) Knotted polymer

(i) 90-95 cm (~19th C) Red fibre

to 539 particles per kg^{-1} dry weight, occurred in sediment deposited between 2005 and 2009. Total microplastic concentration decreased below the surface (242 particles kg^{-1}, 2000–2005) and further declined to 16 particles kg^{-1} in the late-to-mid-1980s, before increasing (226 particles kg^{-1}) in the late-mid 1960s. This latter sample had the greatest number of fibres in any of the depth intervals (Table 2). Total microplastic concentrations were low (< 70 particles kg^{-1}) in sediments dated between the nineteenth century and mid-1960s. Accumulation rates of total microplastics were low in the mid-1950s to 1960s (104 particles m^{-2} yr^{-1}) with a mid-late-1960s plastic

accumulation peak (Fig. 2) equating to 350 particles m^{-2} yr^{-1}. Accumulation rates in the surface 5 cm (2005 to 2009) are three times higher than those in the mid-late 1960s (984 particles m^{-2} yr^{-1}). Microplastics observed in the > 1-mm sieved fraction, except for one orange fragment in the surface sample, were fibres. Three and one red fibres were found in the 0–5 cm and 35–40 cm sample depths, respectively, in the < 500-μm fractions (ESM 2). Extrapolating the number of microplastic particles in HAMP1 core samples to the whole lake points to a conservative estimate of hundreds of millions of microplastic

particles incorporated into the benthic sediment of Hampstead Pond No. 1.

Microplastic fibre colour and composition

The majority of microplastics retained by the 1-mm and 500-μm sieves were different-coloured fibres, < 5 mm in length. Blue fibres were, overall, the most abundant in the sediments (25% particles), followed by white (22%), red (17%), black (8%), pink/purple (3%) and green/turquoise (2%). Blue, red and white fibres were, however, the only coloured fibres found in the blanks, which indicate their likely prevalence as contaminants in the sediment samples. White fibres are especially problematic as they occurred above blank levels in older nineteenth-century sediments, suggesting either systematic contamination or error in their characterization as synthetic fibres.

A few blue fibres generated spectra characteristic of artificial dyes; indigo and Deorlin blue—but the majority of blue fibres remained unidentified (Table 3). Synthetic dye compounds (Indigo, Evans blue), rather than polymer composition, were similarly identified for black fibres, although one black fibre was identified as PVC in sediment from the mid 1950s–early 1960s (Table 3, highlighted on Fig. 1). Spectra for red fibres analysed were matched to a nitrocellulose composite with red pigment and polysulphide rubber (although this match was not conclusive). Measured white/clear and pink-coloured fibres comprised polyacrylonitrile composites (Table 3). Natural polymers identified were polymerisable starch and mucopolysaccharides (Fig. 2b) in the top 10 cm. Synthetic polymers or substances dominated the identifiable composition of the fibres examined by Raman spectroscopy (90%).

Non-fibre microplastics

Compared to the relative abundance of fibres in the sediments, the only fragments found were distinct orange (0.2–2 mm), foam-like particles and a crumpled film particle (Fig. 2a, c). Orange particles were found only in sediments younger than the early 1950s–1960s (57 particles kg^{-1}), with a maximum in the surface 5 cm (294 particles kg^{-1}). The orange particles have a vesicular/crumb-like texture and deform when squeezed. Representative samples of these orange foamlike particles and the crumpled film were

found to be polystyrene, with various copolymers (Table 3). The accumulation rate of microplastic polystyrene indicates a decline from its appearance in the early 1960s (80 m^{-2} yr^{-1}) to the middle 1980s (not found), before increasing to the surface (540 m^{-2} yr^{-1}).

Discussion

Temporal patterns in microplastic abundance

Low numbers (above blank) of largely unconfirmed-composition synthetic fibres, particles of polystyrene and other polymers comprise the sedimentary sequence of microplastics in HAMP1. Excluding unconfirmed synthetic fibre types as microplastics has a significant effect on microplastic concentration; between 20 and 100% reduction in this study. Synthetic fibres are prevalent in microplastic studies of lacustrine and other freshwater surface sediments (Free et al. 2014; Baldwin et al. 2016; Ballent et al. 2016; Fischer et al. 2016; Su et al. 2016; Vaughan et al. 2017). Derived from the breakdown of synthetic textiles, they are released directly into freshwaters with industrial and domestic wastewater (Napper and Thompson 2016; Miller et al. 2017) and via treated wastewater and reworked sewage sludge spread on agricultural land (Zubris and Richards 2005). Wastewater and sewage inputs of microplastic and synthetic fibres into Hampstead Pond No. 1 are likely to have been low because of the pond's small, parkland catchment. Other sources of fibres could be those shed from clothing and textiles, synthetic swimwear and fishing line used in upstream ponds. Prevalence of fibres in Hampstead Pond No. 1 may also be from breakdown products of woven synthetic fabrics, released by degradation processes occurring in the lake and along the shoreline of the two upstream ponds.

Benthic sediment microplastic concentrations (maximum 539 particles kg^{-1}) in HAMP1 are generally comparable to the number of particles reported in surface sediments of Lake Ontario, Canada (87–616 particles kg^{-1}) (Corcoran et al. 2015), Tai Hu, China (11–234.6 kg^{-1}) (Su et al. 2016) and lakes in central Italy (109–266 particles kg^{-1}) (Fischer et al. 2016), but an order of magnitude lower than the number of plastic particles found in lake sediments of the

Table 3 Raman compositional analysis results; (●) identified, (○) identified but not 100% conclusive, (■) Peaks visible, but not able to interpret, (□) no visible peaks therefore unidentifiable

Sample Age (Depth cm)	Particle description	Polystyrene	PVC	Other polymer	Manmade (e.g. dye)	Natural substance	Unidentifiable	Description
2009–2005 (0–5)	Blue-black fibre						■	–
	Blue-black fibre 2						■	–
	Blue fibre				●			Deorlin blue dye
	Blue fibre 2						○	–
	Clear fibre	○						Inconclusive spectra
	Clear film					●		Starch - polymerisable
	Orange fragment 1	●						Composite (27% syrene/isoprene)
	Orange fragment 2	●						Composite (31% syrene/butadene)
	Red fibre				●			Nitrocellulose + pigment red composite
	Red fibre 2						□	–
2005–2000 (5–10)	Clear fibre					●		Mucopolysaccarides
	Pink/purple fibre			●				Polyacrylonitrile composite
	White flexible fragment	●						Polystyrene-containing composite
	Red–brown fibre				●			Copper phthalocyanine dye
Mid-late 1990s (10–15)	Beige/white fragment						□	–
	Orange–brown fragment						□	–
Early-mid 1990s (15–20)	Blue black fibre						■	–
	Red fibre			○				Polysulphide rubber
	Purple fibre						□	–
Mid 1990s–late 1980s (20–25)	Red fibre			○				Polysulphide rubber
	White fragment						□	–
Mid-early 1980s (25–30)	Blue fibre				●			Indigo dye
	Blue fibre_2						□	–
	Red fibre			○				Polysulphide rubber
	Blue–black fibre				●			Indigo dye
Late 1970s–1960s (30–35)	Clear fibre			●				Poly(acrylonitrile methyl acrylate)
	Curly grey fibre				●			Cibanon dark blue
	Pink/purple fibre			○				Aminobenzaldehyde polymer
	Red fibre						□	–
Late 1960s–Early 1960s (35–40)	Black fibre				●			Composite containing Evans blue dye
	Black fibre 2						■	–
	Black fibre 3						■	–
	Blue fibre				●			Indigo dye - twisted suggests polymer
	Blue fibre 2						■	–
	Blue fibre 3						□	–
	Brown fibre				●			Burnt sienna dye
	Clear fibre						□	–
	Clear fibre 2						□	–
	Blue/clear fibre				●			Indigo dye
	Long black fibre						□	Composite containing Evans blue dye
Early 1960 – mid 1950s (40–45)	Black curved fibre		●					PVC
	Black fibre				●			Indigo dye
	Orange fragment 1	●						Composite (25% syrene/isoprene)
	Orange fragment 2	●						Composite (23% syrene/isoprene)
Mid 1950s to 1940s (45–50)	Blue fibre						□	–
	Blue fibre 2						□	–
	TOTAL	6	1	6	11	2	20	

Sakurada-bori moat in Tokyo (1845–5385 particles kg^{-1}) that receives street runoff and combined sewer overflow (Matsuguma et al. 2017). Although the increased particle and fibre concentration in the upper 10 cm may be concomitant with the slightly more organic (15–20% LOI) sediment in the top of the core (Fig. 1), this effect is not observed in the increased fibre concentration of the 1950s–1960s (35–45 cm). This earlier increase in fibre concentration coincides with the first occurrence of orange polystyrene

particles and significant above-blank abundance of blue and black fibres (dyes/polymers). This coincidence of particle composition and significant concentration increase corresponds with the start of global mass production, use and waste-generation of plastic (Barnes et al. 2009; Zalasiewicz et al. 2016; Geyer et al. 2017).

It is apparent that the record of accumulation in HAMP1 does not follow the exponential historical increase in global plastic production (Thompson et al. 2004; Zalasiewicz et al. 2016). Unlike large-catchment, continental lakes (e.g. Lake Ontario, US–Canada) and marine basins that receive expansive atmospheric and wastewater sources, where the proportion of different microplastics is more commensurate with national production patterns (Corcoran et al. 2015), microplastic abundance and types in small catchments of lakes with limited wastewater inputs such as Hampstead No. 1 Pond are influenced far more by local sources and atmospheric deposition (Dris et al. 2016). These differences among types, sources and transport modes must be considered if a 'plastic horizon' (Corcoran et al. 2014, 2015) is to be used to define or signpost the Anthropocene (Zalasiewicz et al. 2016).

The colour range of fibres found at depth in the Hampstead No.1 Pond core is like that found in previous microplastic surveys, i.e. black, red, white and blue. Colour alone provides ambiguous information regarding the origin of the fibres, as the range of synthetic fabrics (clothes, carpets, ropes) that shed fibres is vast (Browne et al. 2011; Boucher et al. 2017), but placed in a stratigraphic sequence may provide insights into historical changes in microplastic sources to benthic lake sediments (ESM 5).

Discounting the low number found in pre-twentieth-century mud and likely occurrence from modern contamination (Woodall et al. 2015; Wesch et al. 2017; Willis et al. 2017), it may be significant that the few coloured fibres found were red. Fibres of the same colour and form were identified in the upper 50 cm of the core as possibly being made of polysulphide rubber (Table 2). This is an early family of plastics used in hoses and linings of gas/water-tight fabrics. The move towards blue fibres, peaking in the 1960s, followed by a post-1970s decline in concentration of fibres containing synthetic dyes, might reflect trends in fashion and fabric use (Schneider 1994). We observed the presence of indigo, a common synthetic dye used

for polyester and cotton. Twisted forms (McIntyre 2004) (Fig. 2e–h) and high tensile strength noted during microscope selection, however, suggest these blue fibres are synthetic polymers (Burkinshaw 1995; Kunttou et al. 2005; Nor and Obbard 2014). Separating synthetically dyed organic fibres from synthetic fibres can be achieved using chemical digestion (e.g. peroxide or nitric acid) techniques (Nuelle et al. 2014), but was not done in this study because of potential discoloration (bleaching) effects, and the fact that multiple treatments are often required to remove all organic matter from benthic lake sediments. Polyacrylonitrile polymer fibres found in both 1960s–1970s (30–35 cm) and 2000–2005 (5–10 cm) sediments indicate a half century of acrylic-fibre transport to the lake. Concentrations of identifiable polystyrene (PS) particles found in the core range from 60 particles kg^{-1} (1950s–1960s) to 300 particles kg^{-1} of sediment in the upper 5 cm. They occur only post-1950, corresponding with their production and usage, up to the present. Orange foam particles were not present in any of the blanks. The concentration of PS particles of homogenous orange and foam-like structure are similar through the core, suggesting a continuous common source of coloured material for the last ca. 50 years. The absence (Table 2) of larger plastic particles in the sediment record reflects the particle sorting that occurs in benthic environments, determined by depth and local wind/current activity (Vaughan et al. 2017), as well as the lake's position at the bottom of a chain of lakes. Only one larger particle ($\sim$ 2 mm) of orange foam was encountered in the > 1-mm sieved fraction (Table 2).

Methodological limitations

Only a representative selection of microplastic particles from each sediment depth was analysed by Raman spectroscopy and we recognize that this limits the stratigraphic compositional interpretation. Of the particles selected for Raman analysis, 24% contained synthetic material (e.g. dyes), 28% were identified as plastic polymers (ESM 6) and around half (48%) of the particles were unidentifiable or were natural substances. This proportion of identified versus unidentified particles is common in aquatic microplastic studies using Raman spectroscopy (identified 41–67%) (Ballent et al. 2016; Clunies-Ross et al. 2016; Frère et al. 2017). Because of the low number

and size of microplastics found, and the ubiquity of microfibres found in urban atmospheres, contamination during coring and processing (Woodall et al. 2015; Wesch et al. 2017) must be acknowledged as a possible source of error. The number of fibres in the blanks examined during processing, and their potential influence on the interpretation of early plastic occurrence, is recognized.

Conclusions

This ^{210}Pb-dated sediment-core assessment of microplastic concentrations, colour abundance and measured composition, yielded a realistic stratigraphy of historical microplastic deposition in Hampstead Pond No. 1. Dominance of fibres in the sediment and the lake's geographic position and relative isolation from wastewater inputs suggest that atmospheric fallout was an important source of microplastics.

This study indicated that downcore changes in microplastic abundance, type and colour in a sediment sequence may reflect changes in microplastic production and usage over time. It also showed that routine extraction of microplastics from radionuclide-dated lake sediment cores is feasible and best measured alongside other contaminant and environmental proxies. Possible input of contaminant microplastics to sediments during sampling and processing is recognized as a potential problem, and future stratigraphic work must aim to further improve contaminant reduction and blank-control sample methods. With improved contaminant detection and a greater proportion of microplastic polymers identified, future paleolimnological work will undoubtedly assist in quantifying the historical flux of microplastic waste, from terrestrial, through freshwater, to marine environments.

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