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Microplastics in surface coastal waters around Plymouth, UK, and the contribution of boating and shipping activities

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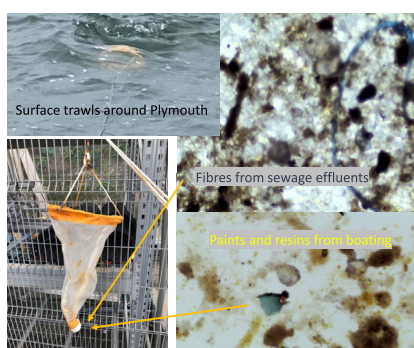
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HIGHLIGHTS

- Microplastics in surface trawls around Plymouth range from 0.26 to 0.68/m³.
- Plastics dominated by rayon and polypropylene fibres and polyester and epoxy fragments
- Concentration of fragments, but not fibres, related to floating/suspended matter concentration
- Observations suggest a decoupling of microplastic origin and transport based on shape.
- Fibres derived from land-based sources, fragments dominated by boating activities

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastics determined in surface water trawls from coastal waters around Plymouth, southwest England, ranged from 0.26 to 0.68 $n\ m^{-3}$, with a decrease evident from the lower estuaries of the Tamar and Plym to areas in Plymouth Sound more remote from urbanisation. Microplastics were dominated by fibres of rayon and polypropylene and fragments of polyester and epoxy resins, with fragment concentration demonstrating a significant and positive linear relationship with concentration of floating and suspended matter retrieved by the trawls. Observations are attributed to the suspension of land-based (e.g., treated municipal waste) sources of textile fibres, and the flotation of land-based and in situ emissions of paints and resins from boating and shipping activities. The implied decoupling of microplastic transport based on shape and origin merits further investigation while the more general determination of floating and suspended matter concentration in microplastic studies is recommended.

1. Introduction

As well as commonly supporting centres of urbanisation and industrialisation, estuaries and coasts are often important areas for marine resources and recreational activities. Consequently, many studies have sought to address the scale and impacts of contamination arising from local municipal, industrial and agricultural sources (Zoller, 2006; Wu and Wang, 2008; Nie et al., 2018; Luo et al., 2022). One particular type of

contaminant that has been receiving growing, recent interest in estuaries and coastal areas is microplastics, or primary and secondary plastic particles in the size range of 1 μm to 5 mm. Sources of microplastics may be both local and distant, and can be introduced to the coastal zone via land, sea and the atmosphere. Specific examples of microplastic sources include water treatment effluents, storm water runoff, waste disposal activities, agricultural and industrial discharges, fishing activities, and boating and shipping.

The generation of microplastics from boat hull coatings or the hull itself, and both in situ and during repair or maintenance, has been referred to in estuaries and coastal areas (Bayo et al., 2019; Ramirez et al., 2019;

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Kumar and Varghese, 2021; Prata et al., 2021). However, relatively little analytical, quantitative or mechanistic information exists in this regard. This is perhaps surprising since several studies suggest that the majority of microplastics, or at least non-fibrous microplastics, in the extended coastal zone or open marine environment are derived from hulls in situ (Song et al., 2014; Dibke et al., 2021; Turner et al., 2022), and shore-based boat facilities are known to generate a great deal of plastic and paint waste (Singh and Turner, 2009; Ciocan et al., 2020). Reasons why information is limited could relate to a lack in consensus as to whether paint particles and composites of plastic and glass should be classified as microplastics or the neglect of these materials in conventional microplastic surveys involving sediments on separation grounds because of their relatively high densities (Turner, 2021).

The aims of the present study are to investigate microplastic concentrations and characteristics in the coastal zone and estuaries around Plymouth, southwest England, and evaluate the contribution of boating activities to the microplastic pool. The area is largely urbanised, and a diversity of maritime, boating and shipping activities take place within a variety of sectors (fishing, military, commercial, transportation, recreational). Samples are trawled from surface waters in various locations with different anthropogenic and maritime influences and are categorised by conventional means and analysed by infra-red spectrometry to determine their polymer composition. We also measure the content of floating and suspended matter captured by the trawls in order to determine whether microplastic concentrations or characteristics are related to the accumulation of other, natural and anthropogenic debris.

2. Methods

2.1. Study area

Plymouth Sound, southwest England (Fig. 1), is a macrotidal inlet of the English Channel whose inner waters of up to 40 m deep are sheltered by a 1600 m breakwater. The majority of freshwater is supplied from the northwest via the Tamar Estuary and from the northeast via the Plym Estuary. The Sound and lower estuaries (i.e., the region under study) are contained within a designated Special Area of Conservation (Joint Nature Conservation Committee, 2022). However, the coastline landwards of the breakwater is largely urbanised by the city of Plymouth (population ~ 260,000) and other, smaller settlements, and supports various shipping, naval and fishing industries and recreational maritime activities. Specifically: the lower Tamar is home to the largest naval dockyard in western Europe; passenger ferries and commercial vessels operate from port facilities located between the Tamar and Plym; up to 40 fishing boats are based at Sutton Harbour; various boatyards, sailing clubs and marinas (accommodating several thousand moorings) are located throughout the region.

Six locations of contrasting hydrographies, anthropogenic signatures and maritime activities were selected within the region for sampling transects (Fig. 1). The Plym Estuary was sampled at the entrance of the sheltered, tidal inlet of Hooe Lake that houses a variety of abandoned boats and receives effluent from a wastewater treatment plant (T1), and in its lower, urbanised reaches in the vicinity of a road bridge, an old landfill site and various boatyards (T2). The partly mixed Tamar Estuary was sampled in the vicinity of the only road and rail bridges crossing the estuary (T3) and in the Hamoaze alongside the naval dockyard (T4). The inner and outer Sound (T5 and T6) were sampled along the eastern edge, with the latter location encompassing a seagrass conservation zone and being the most remote from any urbanisation or sources of treated wastewater.

2.2. Sampling and sample processing

The six transects were sampled at slack water during morning high tides at the beginning of August 2022. Surface samples were collected in a 53- μ m nylon plankton net with a circular aperture of 25 cm in diameter and a threaded PVC cod-end that was towed about 7 m behind a small, motorised,

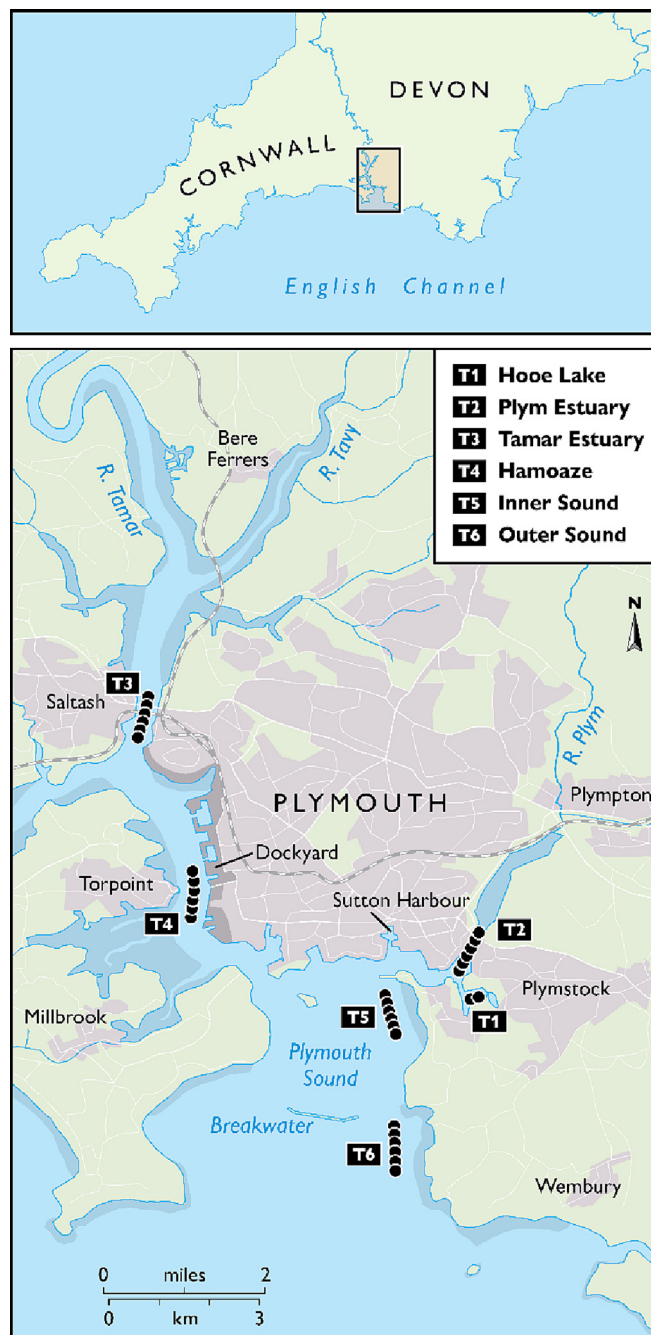


Fig. 1. Plymouth Sound and the lower Plym and Tamar Estuaries, with locations of the six sampling transects annotated as a series of black circles (successive circles denote the beginning and end of each tow).

rigid inflatable boat. At each location, five successive tows were trawled for 3 min at a time and about 200 to 250 m in distance at a constant speed (between 1.8 and 2.3 knots), with the precise positions and distances ascertained by GPS. Transects were about 1 to 1.2 km in length except for Hooe Lake (T1) where parallel tows (whose summed distance equalled 1 km) were performed. With the aperture area of the plankton net submerged of about 0.05 m², the volume of water passing through the plankton net during each tow was estimated to be 9800 L.

After each tow, the outside of the plankton net was thoroughly washed with distilled, deionised Millipore Milli-Q water (MQW) and the material that had accumulated in the cod-end was rinsed into a pre-cleaned, screw-capped, 150 mL glass jar. In the laboratory, the contents of each jar were vacuum-filtered using a ceramic-glass Buchner filtration kit, with

the funnel covered in aluminium foil while not being filled, through a series of (between three and six) 0.45 µm pore size, 47-mm diameter Whatman cellulose nitrate filters. Filters were transferred to covered glass petri dishes and dried at 40 °C in a drying cabinet for 5 h before being weighed on an aluminium foil boat using a five-figure Sartorius microbalance. Two new filters subject to filtration of MQW served as controls, and the average weight of five blank filters was subtracted from dried sample filter weights to determine the quantity of total particulate matter above 53 µm in size that was floating (with a density lower than that of seawater) and suspended (with a density similar to seawater) (hereafter termed FSPM).

2.3. Microplastic identification and quantification

Taking precautions to minimise contamination (e.g., wearing latex gloves and a cotton laboratory coat, washing work surfaces with ethanol), dried filters were inspected under a NIKON SMZ800 stereomicroscope fitted with a 1 × Achro objective (and attaining a magnification up to 63 ×) that was connected to an Olympus SC30 camera operated by Olympus Stream software. Suspected microplastics were identified visually and with the aid of a metal probe using criteria outlined in MERI (2014) based on structure, homogeneity, colour, flexibility and lustre. Suspected microplastics were also classified according to size or length (<3 mm; 3 to 5 mm; >5 mm), colour and shape (fibres: with a length to diameter ratio > 10; fragments: secondary, angular and irregularly-shaped particles; pellets: primary, regularly-shaped spheres, discs and ovoids).

Microplastics were converted to concentrations on a number to volume basis by summing the number of particles identified per transect and dividing by the total volume of water passing through the plankton net aperture. Total water volume was calculated from the area of the aperture that was submerged, the average speed of the boat and the total time of sampling.

2.4. Polymer identification by FT-IR

With the aid of stainless steel tweezers or the wetted tip of a 000 sable hair paint brush, all suspected microplastics were transferred from their filters to the diamond compression cell of a Bruker Vertex 70 Fourier transform-infrared (FT-IR) spectrometer coupled to a Hyperion 1000 microscope. Spectra were obtained using OPUS 7.2 software in transmission mode with the co-addition of 32 scans in the region of 4000 to 500 cm⁻¹ and at a resolution of 4 cm⁻¹. Spectra were compared with various spectral databases of polymers and common materials, with a hit rate of >75 % defined as a positive identification and a hit rate between 65 % and 75 % requiring manual inspection and confirmation of key spectral peaks relative to reference spectra.

3. Results

3.1. FSPM concentrations

Table 1 summarises the data for FSPM (>53 µm) over the six transects and comprising five tows each. Mean concentrations ranged from 0.0325 mg L⁻¹ in Hooe Lake (T1) to 0.0943 mg L⁻¹ in the Hamoaze of the Tamar Estuary (T4). Under the microscope, it was established that FSPM consisted largely of biogenic material (decaying fragments of organisms and faecal matter and dominating the floating component)

Table 1

FSPM concentrations for each transect (as mean ± one standard deviation for the component tows; n = 5), and the total number of microplastics by shape and the overall concentration of microplastics (n MPs) per m³ over each transect.

| Transect | Location | FSPM, mg L ⁻¹ | Fibres, n | Fragments, n | Pellets, n | Total, n | MPs, n m ⁻³ |
|----------|---------------|--------------------------|-----------|--------------|------------|----------|------------------------|
| T1 | Hooe Lake | 0.0325 ± 0.008 | 19 | 5 | 1 | 25 | 0.52 |
| T2 | Plym Estuary | 0.0356 ± 0.006 | 16 | 6 | 3 | 25 | 0.52 |
| T3 | Tamar Estuary | 0.0723 ± 0.015 | 19 | 11 | 3 | 33 | 0.68 |
| T4 | Hamoaze | 0.0943 ± 0.028 | 5 | 15 | 0 | 20 | 0.40 |
| T5 | Inner Sound | 0.0904 ± 0.004 | 4 | 10 | 0 | 14 | 0.28 |
| T6 | Outer Sound | 0.0746 ± 0.032 | 4 | 9 | 0 | 13 | 0.26 |

with smaller contributions from grains or aggregates of silt (the suspended component).

3.2. Abundance and classification of microplastics

The total number of suspected microplastics sampled was 146. However, the results of the FT-IR analysis (see Section 2.4) established that twelve particles were natural (albeit anthropogenic) cotton fibres and four fragments were not identifiable. The control filters were also found to contain a total of two white, cotton fibres. Neglecting cotton-based fibres and unidentifiable particles, microplastic numbers in the transects (summed for the five component tows in each case) ranged from 13 to 14 in the Sound to 33 in the Tamar Estuary, with respective overall concentrations for the transects ranging from 1.3 n m⁻³ to 3.4 n m⁻³ (Table 1). In the Plym and Tamar Estuaries (including Hooe Lake), fibres were the dominant microplastic particle type and pellets were detected. Elsewhere, fragments were the most important type and pellets were absent. Overall, fibres and fragments made up 52 % and 43 % of microplastics, respectively, and the order of abundance by colour was: black > blue, white-transparent > red; with no green or yellow-orange particles observed. The number of microplastics increased with decreasing size (53 µm to 3 mm = 102; 3 to 5 mm = 17; >5 mm = 11) and among the different shapes, this distribution was most pronounced for fragments (<3 mm = 49; 3 to 5 mm = 6; >5 mm = 1). Particles in the largest size category were encountered throughout T1 to T4 but were absent from Plymouth Sound (T5 and T6).

While the concentration of FSPM was not related to the total concentration of microplastics, a significant, linear relationship was observed with the concentration of microplastic fragments (Fig. 2).

3.3. Polymeric make up of suspected microplastics

Table 2 summarises the results from the FT-IR analyses. Among the microplastics with a positive identification (n = 130), 74 % were synthetic, petroleum-based polymers, and 26 % were constructed of fibres of the semi-synthetic cellulosic, rayon. Regarding the synthetic polymers, polypropylene was distributed among the three different shape categories, whereas all particles of epoxy resin and polyester or vinyl ester resins were fragments and all polyamides were fibres. FT-IR also identified glass fibres as a filler in some epoxy and polyester resins, with absorbance peaks characteristic of silicate glass evident in the region 1000–1200 cm⁻¹ (Hopkinson et al., 2021).

4. Discussion

Comparisons of the present results with literature data on microplastics in coastal waters have to be undertaken with caution because variations in the precise means of sampling and identification can have considerable impacts on the microplastic abundance reported. In Plymouth Sound and the English Channel, Lindeque et al. (2020) found that microplastic concentration in surface waters determined after enzymatic digestion of FSPM was highly sensitive to the trawl net mesh size selected. Thus, mean concentrations were 10.03 ± 2.21 n m⁻³ for a 100 µm net, 4.08 ± 1.32 n m⁻³ for a 333 µm net, and 1.03 ± 0.16 n m⁻³ for a 500 µm net, with no differences in size-specific concentration evident between sampling sites. Using a single, smaller net size of 53 µm but extending sampling into the lower estuaries

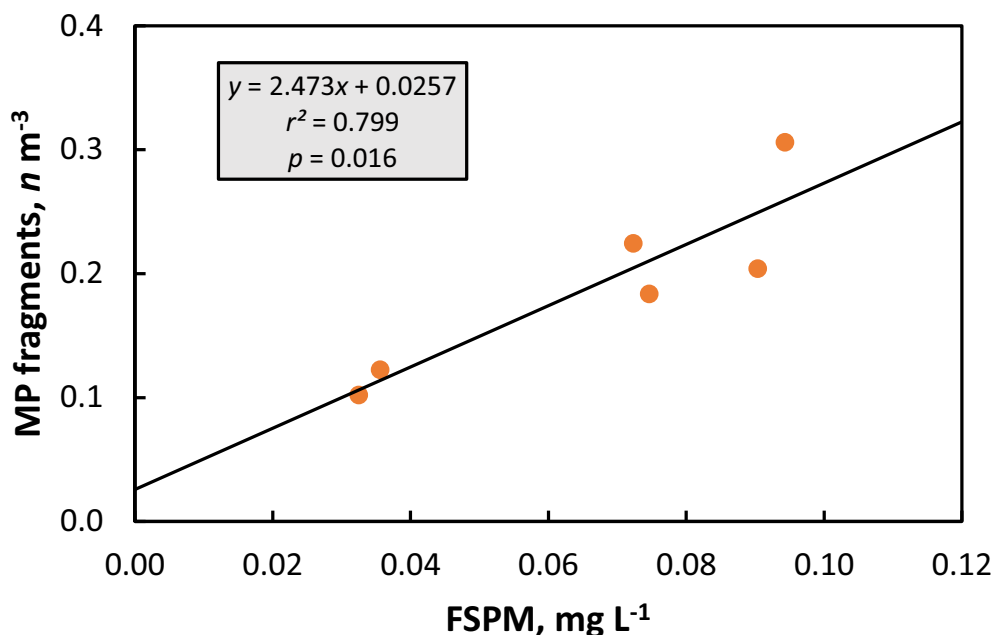


Fig. 2. Concentration of microplastic fragments versus FSPM concentration for the six transects and the best-fit regression line and equation defining the relationship.

of the Tamar and Plym, we found microplastic concentrations averaging 0.44 n m^{-3} (and closer to concentrations reported for other anthropogenically impacted coastal waters; Rodrigues et al., 2020) and evidence of an inverse relationship between concentration and particle size. However, our microplastic concentrations were more sensitive to location, with values lower in the Sound than in the estuarine reaches of the Tamar and Plym. We also found a higher proportion of larger and more fibrous particles in the Tamar and Plym estuaries compared with the Sound, and that pellets were restricted to the most landward reaches of the estuaries sampled. This suggests that land-based sources are more important for fibres constructed of rayon, polypropylene and polyamides and for polyolefin-based pellets than for fragments that were dominated by polyester and epoxy.

One of the most important, land-based sources of microplastic fibres to the environment, and in particular the coastal environment, is wastewater from treatment plants (Leslie et al., 2017; Petroody et al., 2020). Here, textile fibres (including those composed of the polymers identified here) are largely derived from the washing of clothes and, regardless of polymer density but because of their size and shape, a significant proportion of these fibres can evade capture during sludge settlement (Cesa et al., 2017; De Falco et al., 2019). Other land-based, urban sources of fibres and other microplastics may also be transported to the coastal zone via stormwater flow (Schemewski et al., 2021) and the atmosphere (Szewc et al., 2021).

In surface waters of San Francisco Bay, Sutton et al. (2016) noted a dominance of fragments over fibres, despite the latter having clear and

significant wastewater treatment sources. The authors suggested that fragments might be derived from stormwater runoff or in situ fragmentation of (unspecified) larger pieces of plastic. However, along the Douro River estuary, Portugal, Prata et al. (2021) noted an abundance of fragments close to boatyards and docks and suggested an origin related to boat maintenance. Consistent with Prata et al. (2021), evidence from our FT-IR analysis suggests that fragments are derived from boats, and in particular those with fibreglass hulls. Specifically, fragments were dominated by thermosetting polyester and epoxy resins and while some samples could have been paint flakes, matches to spectral library databases often identified sheet moulding composites (ready to mould glass-fibre reinforced polyester or epoxy). Given the popularity of recreational boating in the region and the density of moorings and hard-standings, we suspect that fibre-reinforced polymers (FRPs) are derived from both land-based hull repair and maintenance activities and the direct shedding of material from seaborne boats. Although information on the in situ release of FRPs from boat hulls is lacking, Song et al. (2014) reported an abundance of polyester-based particulates that included FRPs in the sea surface microlayer of coastal Korea. Despite the densities of polyester and epoxy resins (up to 1.4 g cm^{-3}) and FRPs (1.25 to 2.5 g cm^{-3}) (Abbood et al., 2021) exceeding that of coastal seawater ($\sim 1.02 \text{ g cm}^{-3}$), the authors were able to demonstrate significant retention of particles up to 4.4 mm in diameter in the microlayer by surface tension.

The positive relationship between the number or concentration of fragments and FSPM (Fig. 2) suggests that these microplastics are transported, distributed and accumulated more generally with other floating and suspended material, and in particular the floating component which dominates this pool. Thus, as well as surface currents, critical here are areas of convergence, density fronts and downwelling (D'Asaro et al., 2018). By contrast, lack of a relationship between FSPM and concentration of fibrous microplastics on a number basis suggests that this pool, comprising polymers whose densities are both below (e.g., polypropylene; $\sim 0.9 \text{ g cm}^{-3}$) and above (e.g., rayon; $\sim 1.5 \text{ g cm}^{-3}$) that of coastal seawater, are subject to dispersion by surface currents and turbulence and, for denser fibres, ultimate (albeit slow) settlement in the water column (Bagaev et al., 2017). We note Song et al. (2014) report that fibres makeup only 2 % and fragments about 95 % of microplastics floating in the sea surface microlayer of coastal Korea (captured within the pores of a metal sieve through surface tension), and while Stead et al. (2020) report microplastic fibres in the surface microlayer of Southampton Water, UK (captured on glass plates through surface tension), these were largely constructed of low-density polymers

Table 2

Total number of microplastics captured over the six transects by polymer type (and in decreasing order of abundance) and according to shape.

| Polymer | Total | Fibres | Fragments | Pellets |
|-----------------------|-------|--------|-----------|---------|
| Polypropylene | 37 | 25 | 7 | 5 |
| Rayon | 34 | 34 | | |
| Epoxy | 25 | | 25 | |
| Polyester | 22 | | 22 | |
| Cotton | 12 | 12 | | |
| Polyamide | 8 | 8 | | |
| Unidentified | 4 | | 4 | |
| Polyethylene | 2 | | | 2 |
| Acrylic | 1 | | 1 | |
| Polyethylene sulphide | 1 | | 1 | |

(mainly polyethylene; $\sim 0.9 \text{ g cm}^{-3}$). Reasons for the discrepancy in mode of transportation based on microplastic shape are unclear but could be related to differences in interfacial contact area, particle mass and inertia, and polymer deformability (Valero et al., 2022).

In the region under study at least, it appears that the microplastic stock in coastal seawater is comprised largely of floating fragments derived from boating and shipping activities and suspended fibres from various land-derived sources. Whether these characteristics apply to microplastic fragments and fibres in coastal environments more generally and whether fragments are physically associated with floating debris (through adherence or entrapment, for example) are presently unknown. However, factoring in the nature and quantity of floating and suspended particulate matter rather than destroying or disregarding it could improve our understanding of the sources, transport and fates of microplastics in surface waters.

CRedit authorship contribution statement

Chloé Higgins: Conceptualization, Investigation, Formal analysis, Writing – original draft. **Andrew Turner:** Conceptualization, Formal analysis, Writing – original draft, Writing – review & editing.

Data availability

Data will be made available on request.

Declaration of competing interest

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

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