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Synthetic microplastic abundance and composition along a longitudinal gradient traversing the subtropical gyre in the North Atlantic Ocean

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ABSTRACT

Plastic pollution has been reported in the North Atlantic Ocean since the 1970s, yet limited data over subsequent decades pose challenges when assessing spatio-temporal trends in relation to global leakages and intervention strategies. This study quantified microplastics within the upper ocean along a longitudinal transect of the North Atlantic and its subtropical gyre. Microplastics were sampled from surface and subsurface (–25 m) water using a manta trawl and NIKSIN bottle respectively. The surface water polymer community varied significantly between geographic positions ('inshore', 'gyre', 'open ocean'), and was significantly influenced by fragment quantity. Compared to other positions, the North Atlantic gyre was associated with high concentrations of polyethylene, polypropylene, acrylic and polyamide fragments. Subsurface water was dominated by polyamide and polyester fibres. Backtracked 2-year Lagrangian simulations illustrated connectivity patterns. Continued monitoring of microplastics throughout the water column of the North Atlantic Ocean is required to address knowledge gaps and assess spatio-temporal trends.

1. Introduction

Plastic pollution of the marine environment is now widely evidenced to threaten resources and cause detrimental impacts to a diversity of species which can present conservation challenges (Avery-Gomm et al., 2018; Beaumont et al., 2019; Galloway et al., 2017; Kühn and van Franeker, 2020). The annual emissions of plastic from land into the aquatic environment are estimated to increase to 35–90 million tons within the next decade under a 'business as usual' scenario (Borrelle et al., 2020). The persistence and ubiquity of plastic within the ocean, along with the range of environmental, social and economic impacts caused by this pollutant (Beaumont et al., 2019) has led to plastic pollution becoming categorised as a planetary boundary threat (Arp et al., 2021; MacLeod et al., 2021; Villarrubia-Gómez et al., 2018).

Over half of the plastics produced have a density which is less than, or similar to seawater (Andrady, 2011; Geyer et al., 2017). When introduced to the marine environment from land-based and marine-based sources, buoyant plastics can disperse over large distances influenced by ocean currents and wind (van Sebille et al., 2020).

Observational and modelling studies find that a large proportion of plastics converge and may accumulate within subtropical oceanic gyres (Cozar et al., 2014; Eriksen et al., 2014; Law et al., 2010; Lebreton et al., 2018) whereas others remain within coastal regions (Chenillat et al., 2021; Lebreton et al., 2019; Onink et al., 2021). However, not all plastics remain floating at the ocean surface. Under the influence of weathering processes and interactions with organic particles and biota, the physical properties of plastics constantly change which alter their behaviour and transport pathways (Ballent et al., 2012; Coppock et al., 2019; Kaiser et al., 2017; Khatmullina and Chubarenko, 2019; Kvale et al., 2020). Additionally, vertical mixing from wind and turbulence can distribute plastics through the upper ocean (Enders et al., 2015). Indeed microplastics are documented within the water column globally (Courtene-Jones et al., 2017; Egger et al., 2020; Kanhai et al., 2018; Li et al., 2020; Pabortsava and Lampitt, 2020; Tekman et al., 2020; Wang et al., 2022; Zhao et al., 2022) and as such it is important to sample microplastics within this environmental compartment when assessing quantities present in the ocean.

While research within the field of (micro)plastic pollution has grown

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considerably over the last few decades, the geographic range over which in-situ data have been collected remain heterogeneous, with entire regions still not sampled (Khatmullina and Chubarenko, 2019; van Sebille et al., 2015). Computational modelling approaches have offered a tool to ‘fill in the gaps’ and consider microplastic distribution over broad scales (Lebreton et al., 2012; Maximenko et al., 2012; van Sebille et al., 2015), yet these do not include all processes impacting on plastic transport and/or removal from the surface, and much require verification from empirical studies (van Sebille et al., 2020). Validating models can be confounded by limited data and by the wide variety of sampling and extraction techniques utilised by researchers, presenting challenges when comparing data spatially or temporally (Hartmann et al., 2019; Rochman et al., 2017).

Effectively addressing the issue of marine plastic pollution requires information on the abundance, distribution and composition of plastic. While some of the earliest accounts of plastic within the ocean are from the North Atlantic (Carpenter and Smith, 1972; Wilber, 1987), over the subsequent decades, while studies examining beach litter have increased (Edo et al., 2019; Pieper et al., 2015) there have been limited studies published examining and quantifying the geographic range of oceanic plastic pollution (Eriksen et al., 2014; Law et al., 2010; Pham et al., 2020; Reisser et al., 2015) when compared to other ocean basins, such as the North Pacific (Desforges et al., 2014; Egger et al., 2020; Goldstein et al., 2013; Law et al., 2014; Lebreton et al., 2018; Martinez et al., 2009; Rios Mendoza and Jones, 2015). The lack of continued monitoring within the North Atlantic Ocean presents challenges when trying to assess trends in the quantity and distribution of marine plastic pollution in relation to global emissions, and the reduction strategies and regional management plans implemented (Karasik et al., 2020; OSPAR Commission, 2014; Wilcox et al., 2020; Xanthos and Walker, 2017).

The present study examined microplastics within the upper ocean (surface and –25 m depth) of the North Atlantic Ocean across a longitudinal transect from 10 °W to 61 °W and aimed to contribute towards providing further data of plastic quantities. Sampling surface and subsurface water from the same location enabled a more holistic overview of microplastic pollution within the upper ocean. Additionally, samples were collected from regions where data are sparse, such as towards the eastern boundary of the North Atlantic gyre (Eriksen et al., 2014; Isobe et al., 2021; Wilcox et al., 2020). Coupling empirical data with 2-year backtracked Lagrangian particle simulations enabled the dynamic pathways and connectivity of plastics to be considered.

2. Methods

2.1. Sample collection

Sampling was performed from the sailing vessel TravelEdge as part of the ‘Round the World’ voyage organised by eXXpedition. Two transects took place between September–November 2019 (Fig. 1 and Supplementary data) transiting from the UK to the Azores, and from the Azores to Antigua. These cross the North Atlantic anticyclonic subtropical gyre from north east to south west, encountering first the eastward flowing remainder of the Gulf Stream passing to the north and south of the Azores, then traversing the centre of the gyre to finally pass through the westward flows that feed the North Equatorial Current (Talley et al., 2011). In total, 28 surface water and 5 sub-surface (collected from –25 m depth) water stations were sampled (Fig. 1 and Supplementary data). Contamination mitigation measures were implemented throughout (Section 2.3).

2.1.1. Surface water sampling

The surface water protocol was based on that of Collignon et al. (2012); Eriksen et al. (2014); Eriksen et al. (2013). Samples ($n = 28$) were collected using an aluminium manta trawl (rectangular mouth, inner diameter: 13.6 cm height \times 64.4 cm wide) which had a 1.5 m long net with a detachable cod-end (30 \times 10 cm) both made from 335 μ m

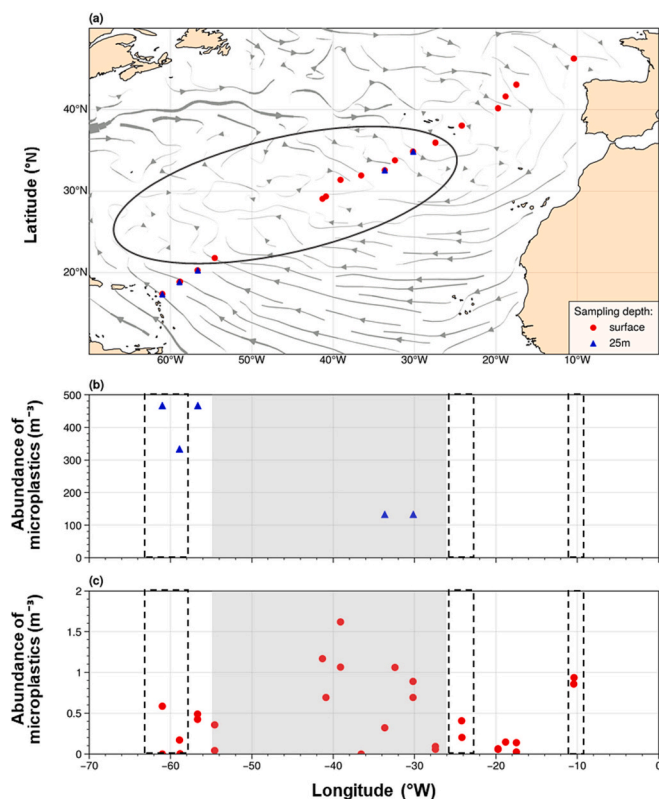


Fig. 1. (a) Map illustrating the surface water (red points) and subsurface water (blue triangles) sampling locations across the North Atlantic Ocean, overlaid with the cmems GLOBAL model annual mean surface current velocity streamlines for 2019. The ellipse indicates the broad location of the central region of the North Atlantic subtropical gyre. The abundance of microplastics m^{-3} isolated from (b) subsurface (blue triangles) water at a depth of –25 m and (c) surface water samples across the longitudinal transect are presented. Longitudes shaded in grey indicate sampling within the North Atlantic gyre central region, those within hatched boxes are from ‘inshore’ sampling locations (≤ 200 nm from land) and the remaining longitudes are ‘open ocean’ locations. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

polyamide mesh. The manta trawl was towed for 30 min, using the spinnaker pole to position the towline outside of the wake of the vessel. Sampling was conducted in Beaufort Sea State ≤ 3 , and for all trawls environmental data including the sea state and wind speed were recorded. The GPS locations were used to calculate the distance travelled (see Jones-Williams et al., 2020; Suaria et al., 2020). The volume passing through the trawl was subsequently calculated by multiplying the distance travelled by half of the net aperture (0.04 m^2): this assumes a laminar flow through the cod end of the net and that half of the net aperture was submerged. The water volume was used to normalise the abundance of microplastics (m^{-3}) in surface water. Once the manta trawl was recovered, the cod-end was removed and carefully inverted over three stacked sieves, of mesh sizes 4.75 mm, 1 mm, and 0.335 mm. A gentle flow of filtered seawater (50 μ m aperture filter, see Section 2.3) was used to transfer all material into the sieves and the cod-end was inspected thoroughly to ensure no microplastics remained on the net. Any large natural debris was thoroughly rinsed with filtered seawater over the sieve and examined to ensure there were no microplastics adhered to its surface, and then discarded. For the size fractions 4.75 mm and 1 mm from each trawl, all potential plastics were transferred into glass vials, separated by size, and for the 0.335 mm all material on the sieve was transferred. Samples were stored in the dark at ambient temperatures.

2.1.2. Subsurface water sampling

Triplicate subsurface bulk water samples were collected from a depth of -25 m at each sampling location ($n = 5$) using a 5 L OSIL NISKIN bottle. The bottle was deployed open from the spinnaker pole using a blue polypropylene rope. Once at -25 m, a brass messenger weight was used to trigger and close the caps of the bottle before it was recovered to the surface. NIKSIN bottles were only deployed in Beaufort Sea State ≤ 2 , and the vessel was held in a stationary position. The GPS location and time of deployment was recorded as was environmental data on the wind speed and direction.

Once on-board, the NISKIN bottle was filtered below deck through a $20\ \mu\text{m}$ cellulose filter paper ($\varnothing 70$ mm) using a stainless steel hand vacuum filtration system. A clear-silicon tube was used to deliver the water from the NISKIN bottle into the filter funnel. Once the entire 5 L volume of water had been filtered, the filter paper was immediately placed into a petri dish and sealed for further analysis at the University of Plymouth. This entire process was repeated for each of the triplicate samples, using separate filter paper for each NISKIN bottle deployment.

2.2. Characterisation of microplastic

Microplastics were categorised according to their morphology as either i) fragment, ii) film, iii) monofilament line, iv) foam, v) pellets, vi) fibres (fibres included for subsurface sampling only) (Hidalgo-Ruz et al., 2012). The colour of each particle was also recorded (Supplementary information). Sizes of plastics obtained in the manta trawls were categorised according to the sieve size fractions (Eriksen et al., 2013), while microplastics in subsurface water were measured along their longest dimension using the ocular scale of the Lumos dissecting microscope and were subsequently grouped into the same categories as the manta trawls. Fourier Transform infrared (FTIR) spectroscopy was undertaken for every putative plastic (see Supplementary information) to determine polymer types. Plastics (1 mm and 4.75 mm size fractions) collected by manta trawl were identified using a PerkinElmer Spectrum Two, Attenuated total reflectance FTIR spectrometer which has a spectral resolution of $0.5\ \text{cm}^{-1}$. Each spectrum was the average of four co-added scans within the range $400\text{--}6000\ \text{cm}^{-1}$ wavenumbers, with a background scan performed prior to each sample. Spectral data were visualised in the complementary software Spectrum IR v.10 (PerkinElmer, 2017) with use on the inbuilt PerkinElmer 'Polymer' spectral library to facilitate sample identification. Surface water microplastics retained on the $335\ \mu\text{m}$ sieve, along with those sampled from subsurface water were analysed with a Bruker Vertex 70 μFTIR coupled with a Bruker Hyperion 1000 microscope run in transmission mode. Each spectrum was the average of 32 co-added scans in the wavenumbers $600\text{--}4000\ \text{cm}^{-1}$. A background scan, also the result of 32 co-added scans, was performed prior to each sample. Microplastics were analysed on a diamond compression cell (Specac DC2; 2 mm diameter). Spectra were visualised in the complementary OPUS v7.5 software, and the spectral libraries 'BPAD', 'synthetic fibres' and the database developed by Primpke et al. (2018) were utilised to facilitate identifying samples along with manual inspection of each spectrum to confirm and quality control the matches.

2.3. Contamination mitigation

Prior to use, the sieves used to sort the manta trawl samples were backwashed thoroughly with a deck hose fitted with a $50\ \mu\text{m}$ aperture stainless steel mesh screen. Fibres were not included in manta trawl data, however pieces of monofilament line were considered. The funnel used to filter subsurface waters, was kept covered with aluminium foil, apart from a small gap where the tube delivering water into the funnel was placed. Techniques widely used in forensic science and within the field of microplastics research were implemented during this research (Prata et al., 2021; Wesch et al., 2017; Woodall et al., 2015). Atmospheric controls were implemented, involving placing a damped filter

paper ($11\ \mu\text{m}$ particle retention, $\varnothing 90$ mm) in an uncovered petri dish adjacent to filtration, to sample any airborne microplastics which may be deposited into our samples (Woodall et al., 2015). There was one instance where a synthetic microplastic was identified within the atmospheric controls (polyester fibre) and where appropriate this was subtracted from the raw count for the triplicate samples, to obtain a 'corrected count'. Samples of putative contaminants (manta trawl net, NISKIN bottle, lines and ropes, boat hull and deck paints) were collected and spectroscopically analysed concurrently with samples. Sample counts were adjusted where necessary (see Supplementary information), for any particles with colours and polymer types matching the contaminants. Laboratory analysis was performed in a positive pressure microplastics laboratory (air filtered to $0.5\ \mu\text{m}$). Personnel inside the laboratory wore natural fibre clothing under a cotton laboratory coat and specific laboratory shoes. The number of personnel within the laboratory at any one time was kept to a minimum.

2.4. Statistical analysis

Microplastic counts, as confirmed by FTIR analysis, were corrected for any contamination detected (polymer-specific corrections) (see Supplementary information). Cellulosic fibres detected in subsurface water samples were excluded from all reporting and statistical analyses, to only consider synthetic polymers within the North Atlantic. Counts were normalised by the volume of water sampled, to derive quantities per m^3 , and all statistical tests were performed on these values (raw counts and plastics/ km^2 provided in Supplementary data files to facilitate inter-study comparisons). Pearson correlations were applied to determine the influence of environmental variables (wind speed and sea state) on the abundance of plastics identified within water samples. To explore variation in the spatial distribution of plastics, sampling locations were broadly categorised as i) 'inshore water' (≤ 200 nautical miles (nm) from shore, within a state's exclusive economic zone), ii) 'North Atlantic gyre', considering the central convergence area as identified from previous studies (Law et al., 2010; van Sebillie et al., 2015; Wilcox et al., 2020) and surface water velocity data (Fig. 1), and iii) the wider 'open ocean' (>200 nm from shore and not classified as 'subtropical gyre').

Non-metric multidimensional scaling (NMDS) coupled with Permutational Multivariate Analysis of Variance (PERMANOVA) (Anderson, 2001, 2003) were used to evaluate the significance of observed patterns (based on Bray-Curtis dissimilarity and 9999 permutations of the data), and test the null hypothesis that there was no significant difference in polymer composition or morphologies according to spatial or environmental factors. Surface and subsurface water were considered separately, due to the different sampling methods used. PERMANOVA and NMDS could not be applied to subsurface samples due to the limited dataset. Statistical analyses were performed in R Studio v. 1.3 (R Core Team, 2019) with the packages *vegan* (Oksanen et al., 2019), *ggplot2* (Wickham, 2016).

2.5. Lagrangian particle tracking experiment

To examine the dispersion and connectivity pathways of the floating plastic debris that reached the different surface sampling locations, Lagrangian particle tracking experiments were carried out using the Ocean Parcels Lagrangian framework (Delandmeter and van Sebillie, 2019). To obtain a comprehensive ensemble of potential particle trajectories, virtual particles representing floating plastic were seeded within 1° radius from the site location (surface water locations) and released repeatedly in time from 10 days prior to the sampling time to 10 days after every 12 h, resulting in an ensemble of 5220 particle trajectories for each site. Particles were back calculated – using a Runge Kutta 4 scheme – using the daily averaged surface current velocities of the E.U. Copernicus marine service global ocean analysis product (GLOBAL_ANALYSIS_FORECAST_PHY_001_024) (Lellouche et al., 2018)

together with the daily-averaged wave-induced Stokes Drift component of the Copernicus marine service wave analysis product (GLOBAL_ANALYSIS_FORECAST_WAV_001_027) to account for the wave effect on particle movement. Products have a resolution of $1/12^\circ$ horizontally. We assume particles do not experience direct wind drag. In addition, random noise with constant tracer diffusivity of $K = 10 \text{ m}^2/\text{s}$ was added to represent dispersion induced by subgrid scale processes (Kaandorp et al., 2020). Particle positions were backtracked for up to 2 years. Particle trajectories were examined to determine the last coastal point of contact of the particle before reaching the sampling site to gain an indication of the dispersion of particles. A particle was considered to contact the coast when located within less than one model gridcell distance from the coastline ($<12 \text{ km}$). For each trajectory, the last point of contact was recorded along with the time taken to connect with the sampling site.

3. Results

3.1. Microplastic community (composition and quantity) varied spatially

Microplastics (MP) were found in all but two of the water samples collected (26 out of 28 surface water samples and all subsurface water locations; see Supplementary information for particle numbers analysed). Quantities of microplastics and their polymeric composition varied spatially (Figs. 1–4). Within surface water (av. $0.45 \pm 0.44 \text{ S.D.}$; median: 0.34 MP/m^3), the greatest abundance was recorded within the region $30\text{--}42^\circ\text{W}$, which is within the North Atlantic subtropical gyre (1.62 MP/m^3) and a second relatively high abundance ($0.87\text{--}0.94 \text{ MP/m}^3$) around 10°W , lying approximately 350 km (189 nm) from the coast of Spain (Fig. 1). On average, samples collected in the gyre contained the highest concentrations of plastic ($0.62 \pm 0.52 \text{ MP/m}^3$), followed by inshore samples ($0.40 \pm 0.37 \text{ MP/m}^3$), and then open ocean locations ($0.19 \pm 0.19 \text{ MP/m}^3$).

A total of 15 synthetic polymers were identified within surface water (Fig. 2), however their distribution was highly heterogenous across the

transect. PERMANOVA analysis revealed significant differences between the community of polymers in surface water and their geographic position (i.e. ‘inshore’, ‘gyre’ and ‘open ocean’) ($F\text{-perm}_{2,23} = 3.415$, $p = 0.005$). NMDS indicated that samples from within the gyre had a similar polymer community (indicated by the clustering of these points; Fig. 3) and were associated with a composition numerically dominated by polyethylene, polypropylene, acrylic, and polyamide, compared to other sampling positions. Samples collected from the ‘open ocean’ broadly clustered, illustrating similarity between samples, and were most closely associated with polystyrene, PVC and a co-polymer of polyethylene and vinyl acetate. ‘Inshore’ samples, collected $\leq 200 \text{ nm}$ from shore, showed a more disparity in their polymer composition (points are spread with a lack of clustering). Interestingly, paint (which has a higher density than seawater) was associated with samples collected in all three geographic positions (indicated by the proximity of sampling points around this polymer; Fig. 3). Within surface water samples, the quantity of fragments was found to significantly ($p < 0.001$) influence the association between polymer composition and geographic position, with a greater quantity of fragments present in samples from the North Atlantic gyre. Overall, fragments dominated the morphology of microplastics (accounting for between 62 to 100 % of plastics; Supplementary data Fig. S1), and so the aforementioned result can be attributed to the gyre containing a greater overall quantity of microplastics than other geographic regions.

Five polymer types were identified within subsurface samples, collected from a depth of -25 m , (excluding cellulosic fibres which were removed from the analysis; Fig. 4), the most common of which was polyamide, which is negatively buoyant in seawater. Polyester and polyamide were the only two polymers found in subsurface samples from the central region of the North Atlantic gyre, while a greater diversity of polymers ($n = 5$) were isolated from water sampled to its west, close to the Caribbean (Fig. 4). Fibres dominated the samples (50–100 % of synthetic plastic particles; Supplementary data Fig. S2), with fragments accounting for the remaining items. Overall, when data were normalised (MP/m^3), subsurface water quantities were on average

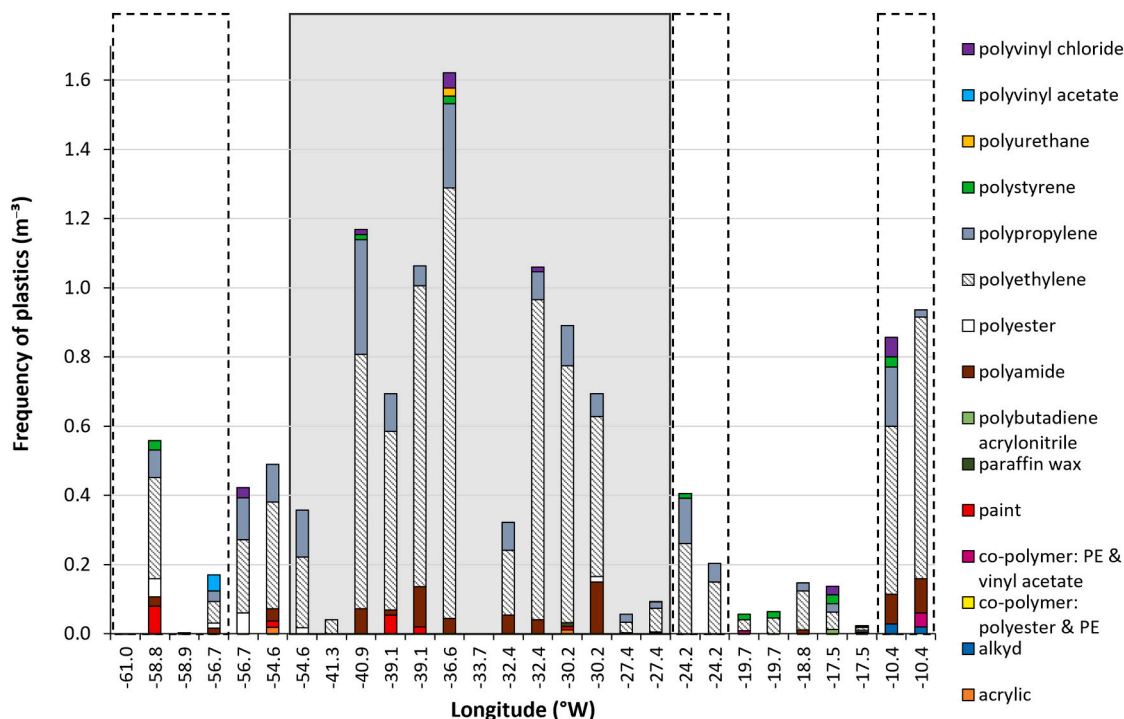


Fig. 2. The abundance of microplastics (m^{-3}) and the polymeric composition recorded during each surface water trawl across the longitudinal transect. ‘Inshore’ sampling locations are depicted within the hatched boxes, the shaded region indicates sampling points from within the North Atlantic gyre and the remaining locations are ‘open ocean’.

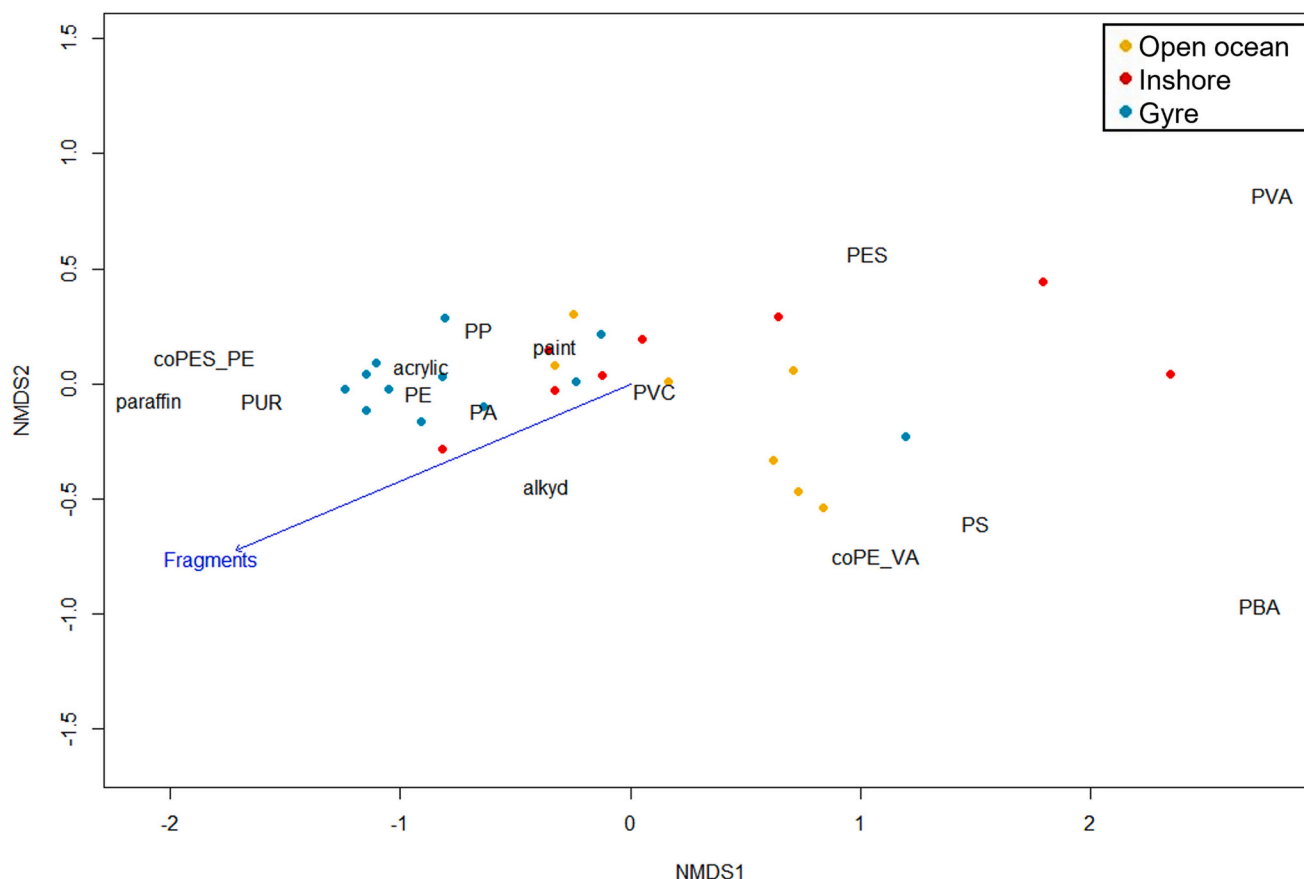


Fig. 3. NMDS plot of Bray-Curtis dissimilarity between the polymer community of surface water sampling sites, classified by their position: 'open ocean', 'inshore' and 'N. Atlantic gyre'. Polymers are displayed in black text (standard abbreviations are used except coPES_PE: co-polymer of polyester and polyethylene, and coPE_VA: copolymer of polyethylene and vinyl-acetate) and point colour definitions are shown in the legend. The quantity of fragments was identified as a significant variable associated with the spatial distribution of polymers, and is overlaid in this plot. 2D stress = 0.04.

306.70 ± 167.33 S.D (median: 333.33 MP/m³; Fig. 1).

There were no significant correlations identified between the abundance of plastics/m³ within surface or subsurface water and either wind speed or sea state.

Overall microplastics 1 mm–4.74 mm in size dominated all surface water samples, with plastics 0.335–0.99 mm in size the second most numerous size category (supplementary data Fig. S3). There was no trend identified between the size of plastic and the geographic location or position (in or out of the gyre). An intact bottle top and an item of plastic cutlery were recovered from two separate trawls in the gyre, however these were the only recognisable objects found (supplementary data Fig. S4). The most prevalent size categories in subsurface water were 0.335–0.99 mm and 1.00–4.74 mm (equally common across samples). The bulk sampling method enabled the retention of smaller sized particles than the manta trawl, consequently plastics 0.02–0.334 mm were also reported in 4 out of the 5 locations (Supplementary data Fig. S5).

3.2. Lagrangian backtracked particle experiments

The connectivity of particles, i.e. the time taken for simulated particles to be backtracked to land, and to which geographic region, varied widely between the location of the sampling points considered in this study (Fig. 5; supplementary data Fig. S6). Overall, the connectivity patterns observed, driven by ocean circulation and waves, illustrate that the closest coastlines were not necessarily the likeliest source for the observed plastic pollution (Fig. 5). For example, simulations performed from surface water sampling sites located in the eastern North Atlantic

(between 46.270 °N, 10.416 °W and 35.950 °N, 27.448 °W) were relatively rapidly backtracked to the coast of North America, South-eastern Canada and southern Greenland, with the vast majority of particles reaching these locations within the 2-year simulation (69–93 % of particles). As sampling locations moved into the North Atlantic gyre, the proportion of particles reaching land within the 2-year simulation decreased (22–80 %), indicating the circulation of particles within the central region of the convergence zone. Also, stations closest to the centre of the subtropical gyre had the broadest potential origins, with modelled particle trajectories suggesting that waste leakages from a number of regions including the Caribbean islands, North America, Canada, western Europe and North western Africa may contribute to the plastics found within these locations (Fig. 5; Supplementary data Fig. S6). As sampling locations approached the Caribbean islands, particle trajectories were backtracked to the coast of Europe, North West Africa and Canada (Fig. 5; Supplementary data Fig. S6). These plastic particles were likely to be distributed by the Canary current and the North Atlantic gyre's northern and southern boundary currents.

4. Discussion

This study quantifies plastic pollution in the upper ocean across a longitudinal transect conducted in the North Atlantic Ocean. Samples were collected within inshore waters (≤200 nautical miles (nm) from shore), the central region of the North Atlantic subtropical gyre and the wider open ocean (>200 nm from shore, and not classified as 'gyre'). Results show that the polymer community in surface water (the diversity and relative abundance of the polymers) varied significantly

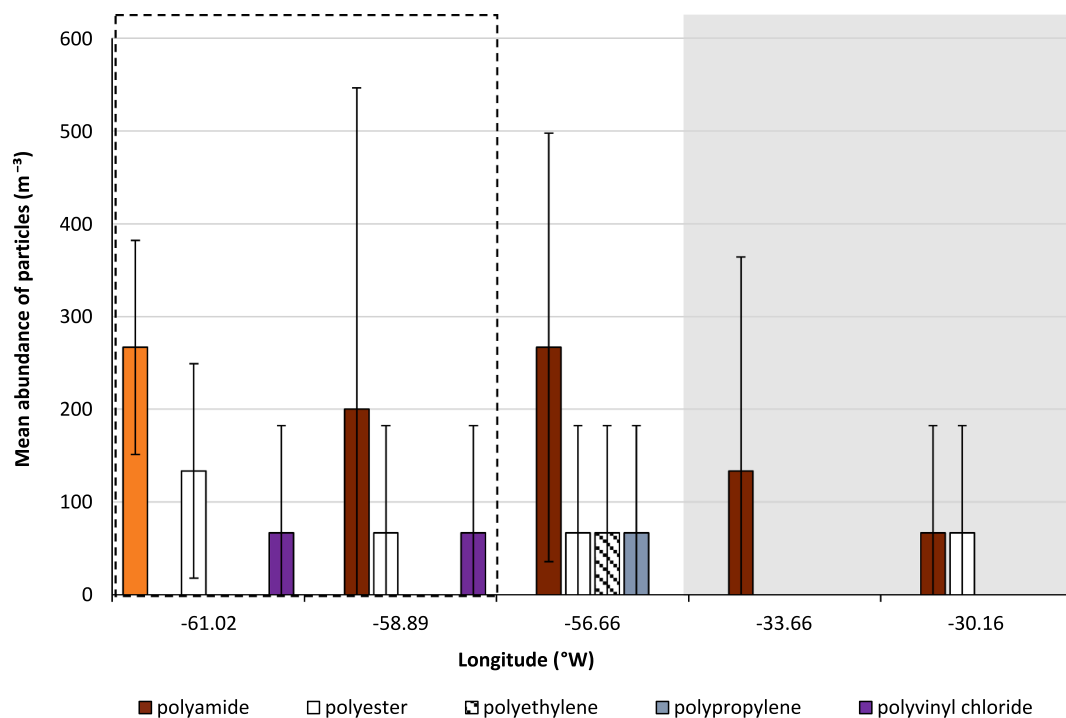


Fig. 4. The mean ($n = 3$) abundance of synthetic particles m^{-3} of water sampled from a depth of -25 m in the North Atlantic Ocean across the longitudinal transect. 'Inshore' sampling locations are depicted within the hatched boxes, the shaded region indicates sampling points from within the North Atlantic gyre and the remaining locations are 'open ocean'. Error bars show standard deviation.

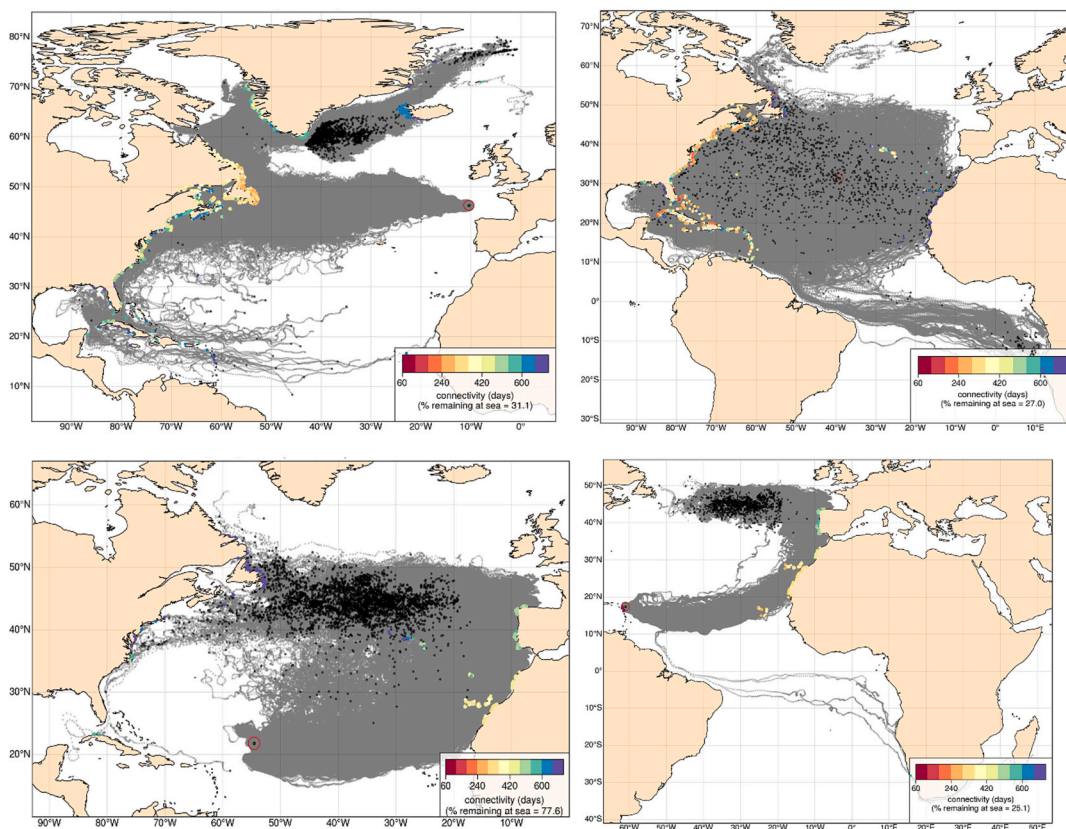


Fig. 5. Example maps illustrating the connectivity of particles released from surface water sampling locations (points circled in red) over the 2-year simulation. The backtracked trajectories are shown in grey, the duration in days until a particle connected with land is illustrated via the coloured points (see legend for interpretation of the colour) and black points represent particles which were not tracked back to land within 2 years. Note that the latitude and longitude boundaries differ between the panels. All maps are provided in Supplementary information. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

geographically. Samples from the North Atlantic gyre were closely associated with polyethylene, polypropylene, acrylic, and polyamide; while those from the open ocean were more associated with PVC, polystyrene and a copolymer of polyethylene and vinyl-acetate; and those from inshore water showed intra-variability in their polymer composition with an absence of a clear association. The variability in inshore samples may be influenced by the relative proximity of these sampling locations to land-based sources which are diverse in their polymeric composition (Pedrotti et al., 2016; Pieper et al., 2019).

Overall, low-density polymers (polyethylene (PE), followed by polypropylene (PP)) dominated the surface water samples, as has been reported in other studies globally (Courtene-Jones et al., 2021; Egger et al., 2022; Enders et al., 2015; Erni-Cassola et al., 2019; Ferreira et al., 2020; Ory et al., 2020; Pedrotti et al., 2016). Collectively polyolefins (PP & PE) account for over half of the total plastics manufactured (American chemistry council, 2021; PlasticsEurope, 2020) and are used widely in disposable or single-use applications such as packaging (Chen et al., 2021). Consequently PE and PP contribute towards a large proportion of plastics entering the waste-stream and leaking into the environment (Geyer et al., 2017). Comparatively high quantities of PE and PP were identified within the gyre than the other ocean regions sampled. The buoyant nature of these polymers may enable them to remain afloat within the upper ocean for long periods of time, where they can be dispersed by surface currents before entrainment within the gyre (Lebreton et al., 2012; Ryan, 2015; van Sebille et al., 2020). However, polymers with densities greater than seawater, such as polyester, PVC and paint flakes were also isolated from the surface water samples. While the mechanisms by which high-density polymers can persist on the sea surface have yet to be fully ascertained, the influence of surface tension, turbulence and wind driven mixing can act to keep particles in afloat, and studies globally have reported high-density polymers at the ocean surface (Song et al., 2014; Soroldoni et al., 2018; Suaria et al., 2016; Wang et al., 2020). Paint flakes were identified in all three regions ('open ocean', 'inshore' and 'gyre'), and paraffin wax was isolated from one sample from the north Atlantic gyre, inferring direct maritime inputs from recreational or commercial vessels (Suaria et al., 2016; Turner, 2021; Turner et al., 2021).

It was noted that those surface samples collected in inshore waters showed high variability in their polymer compositions and on average contained numerically higher quantities of microplastics than open ocean samples. This may be due to these location being closer to the numerous and diverse sources of plastics emanating from land (Pedrotti et al., 2016; Pieper et al., 2019) and additionally in the case of samples collected from around the Azores, its proximity and ocean connectivity to the North Atlantic gyre (Cardoso and Caldeira, 2021; Pham et al., 2020). Large quantities of plastics, including pellets, have been recorded on the beaches of the Canary Islands (Alvarez-Hernandez et al., 2019; Edo et al., 2019; Herrera et al., 2018) and the Azores (Pham et al., 2020; Pieper et al., 2015). There are no plastic processing industries within these archipelagos, indicating pellets are transported within ocean currents. In the present study individual pellets were identified within four surface water trawls in the North Atlantic gyre and one within Azorean water, which Lagrangian modelling indicates may have been transported over large distances via ocean currents from North and/or Central America.

The dispersive nature of marine plastic pollution provides challenges in asserting responsibility for the problem. To this end Lagrangian particle tracking provides a useful tool to consider the pathways and potential origins of marine litter (Cardoso and Caldeira, 2021; Lebreton et al., 2012; Maximenko et al., 2012; van Gennip et al., 2019), yet also highlights the complexity of tracing plastic debris (Khatmullina and Chubarenko, 2019). Overall, the connectivity patterns driven by ocean circulation and waves illustrate that closest coastlines were not necessarily the likeliest origin for the observed plastic and that sampling locations closest to the centre of the subtropical gyre had broadest sources. Over the two-year simulation, the majority of particle trajectories from

the locations 34.8648°N, 30.1802°W to 29.0668 °N, 41.3125 °W were strongly confined, indicating convergence and re-circulation within the gyre region. Those particles which were backtracked to land within such timescales primarily made contact with countries along the western boundary (i.e. North America and Canada). Similar results are evidenced by Cardoso and Caldeira (2021), who found that plastics originating from the west coast of North America were a dominant source of plastic litter to the islands of Macaronesia (Azores, Madeira, Canary Islands and Cape Verde). The method used within the present study only accounted for the last continental point of contact and a maximum drift of two years, providing only a first-order indication of the dispersion of particles, and suggests that connectivity timescales with the eastern boundary coastline are longer. In addition, the computer models do not account for vertical movements or the beaching and remobilisation of plastics from shore, and as such the simulation presents a somewhat simplified interpretation of the dynamic pathways of marine plastics and serves only as an indication of the possible trajectories of particles. Nonetheless, Lagrangian models enable for a more comprehensive picture of the transport dynamics of plastics which would not be achieved through environmental monitoring alone.

The numerical quantity of plastic fragments was significantly greater within the North Atlantic gyre, than in the open ocean or inshore areas. This is likely explained by the hydrodynamics of these features which can retain debris for long-periods for periods (Cardoso and Caldeira, 2021; Onink et al., 2019), and is illustrated by the Lagrangian particle tracking simulations in this study. Subtropical gyres have previously been identified to accumulate marine plastic (Eriksen et al., 2014; Lebreton et al., 2018; Moret-Ferguson et al., 2010; Ryan, 2014) and our results are in line with these observations. The majority of studies have not quantified plastics across the entire North Atlantic Ocean basin longitudinally (Eriksen et al., 2014; Law et al., 2010) impeding attempts to contextualise our findings. Cozar et al. (2014) undertook a similar trans-Atlantic sampling campaign nearly a decade earlier than the present research and reported a similar overall trend in the abundance of plastic, however comparing specific quantities is not possible due to differences in reporting units. A latitudinal transect from Germany to South Africa in 2015 reported heterogeneous concentrations of microplastics which were influenced by the location and environmental variables (water temperature, wind direction and speed and salinity) (Kanhai et al., 2017), however broadly microplastic concentrations were similar to those identified across our longitudinal voyage. While evidence widely supports the convergence and accumulation of plastic within the North Atlantic gyre, few studies have reported plastic concentrations sampled from this region since 2015 (Law et al., 2010; Poulain et al., 2019; Reisser et al., 2015), which presents challenges when trying to assess trends in the quantity and distribution of marine plastic pollution in relation to global emissions, and the reduction strategies and regional management plans implemented (Karasik et al., 2020; OSPAR Commission, 2014; Wilcox et al., 2020; Xanthos and Walker, 2017).

The numerical quantity of plastics within the gyre accumulation zone identified in the present study are comparable Kooi et al. (2016), yet are lower than documented in other research (Law et al., 2010; Reisser et al., 2015). Such variation may be due to spatial differences, as the latter studies sampled the western portion of the gyre (Eriksen et al., 2014; Law et al., 2010; van Sebille et al., 2015), whereas the present research primarily sampled its eastern region – projected to have larger concentrations of surface plastic (Chassignet et al., 2021). Periods of bad weather precluded sample collection along the entire transect; in particular when leaving the UK and at the south-western boundary of the North Atlantic gyre. Generally, the quantity and geographic extent of plastics towards the eastern boundary of the gyre are not well documented (Law et al., 2010; Wilcox et al., 2020) and baseline data presented in this study contributes towards ongoing monitoring efforts.

The presence of synthetic microplastics were confirmed within sub-surface water sampled from a depth of –25 m. Polyamide (PA) fibres

were most numerous, followed by polyester (PES) fibres. Both polymers have applications within textile and fishing/maritime industries (Carr, 2017; Napper et al., 2022). Fibres have been shown to have a lower rise velocity than fragments of the same size, indicating that fibres are more susceptible to vertical redistribution in the upper ocean (Kooi et al., 2016). Similar to other studies, fibres were the dominant morphology identified in subsurface water (Kanhai et al., 2018; Vega-Moreno et al., 2021; Zobkov et al., 2019). Cellulosic fibres which were brightly coloured, indicating they have been subjected to industrial processing, were also found throughout subsurface waters. While cellulosic fibres were excluded from further analysis within this study so as only to consider synthetic plastics, the presence of cellulosic fibres are widely documented in the global ocean (Barrows et al., 2018; Courtene-Jones et al., 2021; Suaria et al., 2020). These findings suggest that industrial treatment processes may cause naturally-derived fibres to degrade far less readily than expected (Bagheri et al., 2017; Barrows et al., 2018). The presence of acrylic (fibres and fragments) and PVC fragments were recorded in the western sampling locations, close to the Caribbean, potentially indicating the influence of land-based sources (e.g. from packaging, mismanaged waste, wastewater effluent discharge (IDB, 2020; Ivar do Sul and Costa, 2007)), as these polymers are negatively buoyant in seawater and may sink relatively rapidly (Erni-Cassola et al., 2019; Kowalski et al., 2016).

Data quantifying microplastics in the ocean interior are scarce, when compared to surface water (Isobe et al., 2021; Pabortsava and Lampitt, 2020); and while it is noted that the results presented here are based on a limited number of samples ($n = 5$ locations sampled in triplicate) and relatively small water volumes, they enable a more holistic overview of microplastics in the upper ocean. If the relatively small subsurface water volumes are extrapolated, it can be suggested that the quantities of microplastics at a depth of -25 m are numerically more abundant than in surface waters, as has been shown in some other studies (Egger et al., 2020; Pabortsava and Lampitt, 2020). However, this observation must be heavily caveated, as differing methods were used to sample the two environmental compartments. Manta trawl and NISKIN bottle were used to sample the surface and subsurface waters respectively and as such methodological differences influence the microplastics collected. Bulk water sampling methods, such as a NISKIN bottle, have been shown to have a greater efficacy at capturing small sized particles and in particular fibres, than nets (Barrows et al., 2017; Schonlau et al., 2020), but can be influenced by the volume of water collected (Ryan et al., 2020). Nets, on the other hand, enable large water volumes to be sampled but fibres and small particles can evade capture depending on the net aperture used (Underwood et al., 2017). As such it is expected that subsurface water collected via bulk sample methods will contain greater quantities of fibres than the manta net derived surface samples, as is reflected in our data. Ryan et al. (2020) found that fibres were abundant at the sea surface and as such quantities of microplastics in surface waters may be under-estimated. As such, more work is required to examine the effect that differing sampling methods have and to gather more information on the quantities of microplastics within the water column.

The majority of plastics isolated from a depth of -25 m were <1 mm in size, while plastics in surface waters were typically 1–4.74 mm in size. This finding may be explained by methodological differences (between trawl nets and bulk sampling as discussed above), but does support wider observations which suggest that small microplastics have a relatively short residence time in surface waters and are thus redistributed to depth more rapidly than larger plastics (Egger et al., 2020; Enders et al., 2015; Koelmans et al., 2017; Poulain et al., 2019). Measurements of plastics within the ocean's interior remain relatively scarce compared to surface waters. Data presented in this study emphasise the need to research microplastics within water column to more holistically understand the quantities present, their spatial distributions, transport pathways and the species which might be interacting with them.

5. Conclusion

The presence of plastic pollution within the North Atlantic Ocean and its subtropical gyre have been reported since the 1970s (Carpenter and Smith, 1972; Wilber, 1987). Over the subsequent decades research efforts tended to focus on the western Atlantic Ocean and its subtropical gyre, compared to other regions of the ocean basin (Law et al., 2010; Wilcox et al., 2020). Spatio-temporal heterogeneity of monitoring poses challenges when examining the extent and prevalence of (micro)plastics in the Atlantic and how trends may relate to global leakage and intervention strategies. This study reports plastic pollution in the upper ocean across a longitudinal gradient of the North Atlantic Ocean, contributing towards ongoing monitoring. Within surface water, the polymer community varied significantly between geographic positions (categorised as 'inshore', 'coastal' and 'gyre'), and was significantly influenced by the quantity of fragments. The North Atlantic gyre was associated with greater numerical quantities of fragments, and the polymers polyethylene, polypropylene, acrylic and polyamide. Inshore samples showed more disparity in polymer composition, potentially influenced by the numerous and varied sources of plastics arising from land. Overall quantities of plastics were similar, or less than, reported in previous accounts (Kooi et al., 2016; Law et al., 2010; Reisser et al., 2015). Synthetic microplastics (primarily polyamide and polyester fibres) were present within subsurface water sampled from a depth of -25 m. While methodological differences prevent direct comparisons of surface and subsurface microplastics (Ryan et al., 2020; Underwood et al., 2017), the ocean interior has been suggested as a substantial reservoir of plastic pollution (Pabortsava and Lampitt, 2020), requiring further investigation. The study highlights the need for ongoing research within the upper ocean of the North Atlantic Ocean to better evaluate spatial and temporal differences in microplastic abundance and composition and provide more accurate assessments of the total budget of marine microplastics.

CRedit authorship contribution statement

Winnie Courtene-Jones: Conceptualization, Methodology, Formal analysis, Investigation, Visualization, Data curation, Project administration, Writing – original draft, Writing – review & editing. **Simon van Gennip:** Methodology, Software, Formal analysis, Investigation, Visualization, Data curation, Writing – original draft, Writing – review & editing. **Juliette Penicaud:** Software, Formal analysis, Investigation, Visualization, Writing – review & editing. **Emily Penn:** Conceptualization, Funding acquisition, Supervision, Project administration, Writing – review & editing. **Richard C. Thompson:** Conceptualization, Funding acquisition, Supervision, Project administration, Writing – review & editing.

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.

Data availability

Data are attached in a supplementary file

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Appendix A. Supplementary data

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