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Airborne microplastic particles detected in the remote marine atmosphere

Miri Trainic^{1✉}, J. Michel Flores¹, Iddo Pinkas², Maria Luiza Pedrotti³, Fabien Lombard^{3,4}, Guillaume Bourdin^{3,5}, Gabriel Gorsky³, Emmanuel Boss⁵, Yinon Rudich¹, Assaf Vardi^{6✉} & Ilan Koren¹

Anthropogenic pollution from marine microplastic particles is a growing concern, both as a source of toxic compounds, and because they can transport pathogens and other pollutants. Airborne microplastic particles were previously observed over terrestrial and coastal locations, but not in the remote ocean. Here, we collected ambient aerosol samples in the North Atlantic Ocean, including the remote marine atmosphere, during the *Tara* Pacific expedition in May–June 2016, and chemically characterized them using micro-Raman spectroscopy. We detected a range of airborne microplastics, including polystyrene, polyethylene, polypropylene, and poly-silicone compounds. Polyethylene and polypropylene were also found in seawater, suggesting local production of airborne microplastic particles. Terminal velocity estimations and back trajectory analysis support this conclusion. For technical reasons, only particles larger than 5 μm , at the upper end of a typical marine atmospheric size distribution, were analyzed, suggesting that our analyses underestimate the presence of airborne microplastic particles in the remote marine atmosphere.

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Marine microplastic particles (MP) are plastic particles of sizes up to 5 mm^{1,2}. Primary microplastic is produced either for direct use or as a precursor for other products, while secondary microplastics are formed in the environment from breakdown of larger plastic material, especially marine debris^{1,2}. Marine MP are a subject of elaborate research, and a matter of great concern in the fields of marine pollution, chemistry, and ecology^{3–13}. MP may pose a risk to aquatic environments due to their documented ubiquity in the marine ecosystems, their long lifetime in the environment, their ability to transport potential pathogens and persistent organic pollutants¹⁴, their role as a new source of toxic compounds in the marine environment, and their propensity to be ingested by diverse organisms^{1,2,15–20}. Marine microplastic persistence is a major source of concern, since plastic entering marine ecosystems doesn't degrade, but rather undergoes mechanical erosion and photooxidation via interaction with ultraviolet (UV) radiation, resulting in fragmentation to micro- and nanoplastic sizes (<1 µm)¹. The microplastic and nanoplastic fragments are easily incorporated in an increasing number of environmental matrices, making them impossible to remove. The effect of MP toxicity on marine biota increases with decreasing size, since smaller sizes facilitate ingestion, as well as chemical processes such as leaching¹. Estimates of recent years suggest that ~5.25 trillion plastic fragments are currently floating on the oceans' surface²¹. Airborne microplastic particles (AMP) from terrestrial origins were found in both polluted and remote areas over land, as well as in coastal regions, ranging from micron to mm scales, and can travel tens of kilometers^{12,13,22–33}. However, despite the overwhelming amounts of marine MP and their effect on the marine environment, atmospheric microplastic particles over the ocean are given little attention¹³. Marine AMP may have implications on marine ecology; facilitated by the winds, AMP can spread potential pathogens and persistent organic pollutants adsorbed onto their surface¹⁴, by an order of magnitude faster than the ocean currents^{34,35}. Furthermore, the components adsorbed to the AMP are exposed to UV radiation and oxidation, which can increase their toxicity to marine biota^{1,2,36}.

The ubiquity of AMP from terrestrial sources, and the ubiquity of MP in the world's oceans raise the question whether AMP are present in the remote marine atmosphere. We searched for marine AMP during the *Tara* Pacific expedition, a 2.5 year research project that investigates coral reefs and plankton communities^{37–39}. In this report, we focus on marine AMP collection and analysis conducted on May–June 2016, during the first North Atlantic crossing of the *Tara* Pacific Expedition.

Results and discussion

The main goals of this study were to determine whether AMP are present in the open ocean, to chemically identify them, and to investigate their possible sources. For these purposes we collected ambient atmospheric aerosol samples along the first North Atlantic transect of the *Tara* Pacific expedition (see Fig. 1) onto polycarbonate membranes. Overall, 43 samples were analyzed, and AMP were found in nine of them (~20%). Using micro-Raman spectroscopy, a highly novel and reliable method for polymer identification and characterization^{40–43} (see “Methods” section for details), we chemically identified AMP as small as 5 µm in the collected marine aerosol samples. Strict measures were taken to assure that the AMP we identified did not originate from the boat, nor from any stage of the sampling and analysis. The R/V *Tara* surfaces were thoroughly sampled, and we found that they may be a source of plastic colorants, polyester and cellulose in the marine environment (see “Methods SI” section for details). Materials found in the handling blanks or on *Tara* surfaces were

excluded as AMP, since their source could be the R/V *Tara* itself. Only materials detected exclusively in the collected atmospheric samples were considered as AMP (full details in the “Methods” section).

We detected AMP in the marine atmosphere from the airborne samples collected in the North Atlantic Ocean, and found that most of them correspond to the most ubiquitous MP found in the marine environment^{6,44–47} (see Fig. 1 and Table 1). AMP samples 2, 5, 17 and 30 contained polystyrene (PS), where the Raman spectrum of sample 17 is a combination of PS with the polycarbonate filter on which the samples were collected and analyzed. Polyethylene (PE) and polypropylene (PP) were observed in samples 38 and 39, respectively. The spectrum of sample 38 shows two peaks in the region of 500 cm⁻¹ wavenumber (Figs. 1b), attributed to rutile (TiO₂), which may serve as a whitening agent or part of a colorant for plastics. The PP found in sample 39 is low ethylene RCP (random co-polymer), i.e., PP containing low levels of an ethylene co-monomer (1–8%). We found mainly poly-silicone compounds in samples 10, 11, and 23, identified as Tegomer E-Si-2330, which is polydimethylsiloxane (PDMS).

PS, PE, and PP were previously identified as common plastic pollutants in the waters of remote ocean locations and shores worldwide^{6,44–46,48}. Here, they were detected in the marine atmosphere. Moreover, PS, PP, and PE were recently identified as the most abundant plastic compounds that were carried in the atmosphere to a remote terrestrial location²². PE and PP are the main components of airborne plastics in urban environments, according to several studies^{24,26,27}. PDMS, which was detected in three samples, is essentially insoluble in water (<1 ng L⁻¹ at 23 °C), and therefore is preserved in the seawater. Since PDMS is less dense than water it is expected to remain in the ocean's surface microlayer, from where it can be subjected to aerosolization via wind driven processes. PDMS has been found in aerosol samples in several US cities⁴⁹.

AMP concentrations are presented in Fig. 2, together with total and coarse mode aerosol concentrations³⁸. Wind speed and direction prevailing during AMP sampling periods are presented for reference. AMP concentrations are in the order of 1–10% of the coarse mode aerosol concentrations for sizes >10 µm. The AMP concentrations obtained here are in good agreement with concentrations attributed to marine source AMP, which were obtained from measurements conducted at the French Atlantic coast³⁰. Based on their coastal measurements, the authors suggested that AMP can be formed via bubble bursting, which may also occur in the open ocean. In order to investigate the open ocean as a source of the AMP obtained in our study, we conducted 48 h back trajectories at 250 m altitude (HYSPLIT; Hybrid Single-Particle Lagrangian Integrated Trajectory) for each point along the *Tara* North Atlantic transect where AMP were detected. The vast majority of the air masses passed over the ocean during the full 48 h trajectories (see Fig. 2c). Next, we assessed the atmospheric residence times of the AMP we found, by calculating their settling (terminal) velocities (Table 1; see “Methods” section for the detailed calculations). The residence time of atmospheric particles is likely to be affected by their atmospheric terminal velocities. Slower velocities (≤0.01 m s⁻¹) imply higher likelihood to be lifted from near the water surface by eddies, and to be carried by the surrounding air⁵⁰. We found AMP settling velocities in the range of 0.001–0.2 m s⁻¹ for the particles (Table 1, see “Methods” section). By combining the HYSPLIT back trajectories with the obtained residence times, we can evaluate the source of the particles. Particles 2, 17, 30, and 38 were clearly a product of local emission, with residence times of minutes to several hours (Table 1). The rest, with residence times of 1–2 days, combined with the HYSPLIT back trajectories, also

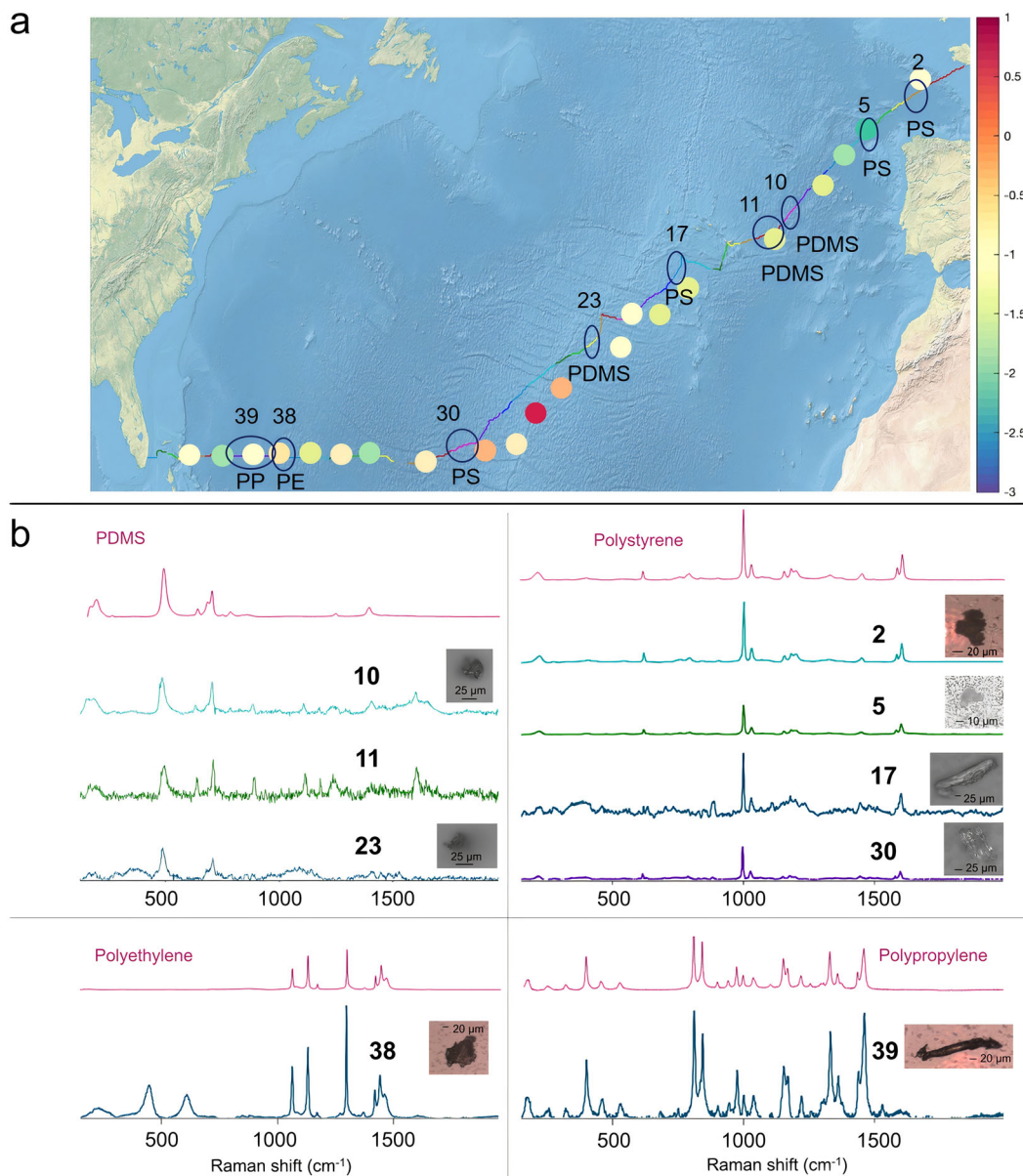


Fig. 1 AMP collected in the atmosphere of the first North Atlantic transect of the *Tara Pacific Expedition* during May–June 2016. **a** The presence of AMP and MP along the Atlantic transect. The different colors of the transect presented on the map represent the segments that the *R/V Tara* crossed during each airborne filter collection. Forty-six airborne samples (filters) were collected in total, with typical sampling durations of 12–24 h. Each airborne sample number where AMP was detected along the transect (2, 5, 10, 11, 17, 23, 30, 38, and 39) is presented on the map, along with the corresponding sampling location (circled in blue), and the detected AMP type (abbreviated; full names of the plastic compounds are provided in Table 1). The colored full circles along the transect represent seawater microplastic particle (MP) log abundance per m^3 . Note that the seawater concentrations vary by 4 orders of magnitude. **b** Raman spectra of the AMP types collected during the transect. The obtained Raman spectrum (arbitrary intensity vs. Raman shift in wavenumber) of each airborne AMP and its light microscope image are presented. The AMP sample number as appears on the map is written next to each Raman spectrum. The Raman spectra of the plastic polymer standards, corresponding to the sampled AMP types are presented in magenta above the samples' spectra.

appear to have been formed in the ocean. This suggests that the AMP found in this study are emitted from the seawater, indicating wind-related processes via bubble bursting.

AMP particles with irregular and elongated shapes can have long atmospheric residence times. For the most elongated AMP, despite having long major axis, their atmospheric settling velocities are in the range of 0.001 m s^{-1} , equivalent to typical sub-micron particles, which are suspended in the marine atmosphere in a timescale of days, and dominate the marine boundary layer⁵¹. We summarize that the AMP we found can remain in the atmosphere up to ~ 2 days, and for a typical marine wind speed of

8 ms^{-1} can be transported to 100's–1000's km. However, AMP with settling velocities in the order of $\sim 0.1 \text{ m s}^{-1}$ will remain in the atmosphere for ~ 5 min, and travel $\sim 3 \text{ km}$ ⁵¹.

To further explore the role of the open ocean as a source of AMP, we compared the chemical composition of the AMP samples with microplastic particles collected from seawater sampled at the same approximate location to where the AMP were sampled. We chose AMP samples collected at $26^\circ\text{N } 66^\circ\text{W}$ – $26^\circ\text{N } 68^\circ\text{W}$ and $26^\circ\text{N } 68^\circ\text{W}$ – $26^\circ\text{N } 72^\circ\text{W}$, (samples 38 and 39, respectively; Fig. 1a), where the HYSPLIT trajectories indicate the air masses are of purely oceanic source. Microplastic particles

Table 1 The sample (filter) numbers in which AMP was detected, the types of plastic polymers detected by micro-Raman spectroscopy and their density, diameters, and calculated equivalent spheres, AMP terminal settling velocities, and estimated residence times.

Filter number	Polymer detected	Polymer density (g cm ⁻³)	Particle major and minor axis (a, b; μm)	μ_s = Terminal velocity of droplet/sphere (m s ⁻¹)	$\alpha = (b/a)^2$	μ = AMP terminal velocity (m s ⁻¹)	AMP residence time ^a
2	Polystyrene	0.96–1.04	66, 53	0.13	0.64	0.08	~ 10 min
5	Polystyrene	0.96–1.04	25, 18	0.017	0.52	0.009	~1 day
10	Polydimethylsiloxane (PDMS)	0.97	22, 17	0.014	0.6	0.008	~1 day
11	Polydimethylsiloxane (PDMS)	0.97					
17	Polystyrene	0.96–1.04	124, 45	0.23	0.13	0.03	~ 5 h
23	Polydimethylsiloxane (PDMS)	0.97	17, 13	0.01	0.58	0.006	~1 day
30	Polystyrene	0.96–1.04	100, 70	0.22	0.49	0.11	~5 min
38	Polyethylene	0.88–0.96	102, 97	0.22	0.90	0.20	~5 min
39	Polypropylene	0.86–0.95	172, 17	0.26	0.01	0.003	~2 days

^aBased on the calculations in ref. ⁵¹ (Table 8 in the chapter).

collected from the seawater on the 22.06.2016 at the area of 26°N 68°W were compared with airborne sample collected at 26°N 66°W–26°N 68°W (sample 38) containing polyethylene AMP, which was collected on the same day, and with airborne sample collected at 26°N 68°W–26°N 72°W (sample 39) containing polypropylene AMP, which was collected the next day (23.06.2016) (see “Methods” section and Fig. 1a).

The seawater microplastic particles selected for analysis and comparison with the AMP underwent thorough morphological scanning, in order to assure their identification as plastic prior to micro-Raman spectroscopy measurements (see “Methods” section). Eight microplastic particles were analyzed from the seawater sample at 26°N 68°W, seven of which contained polyethylene or polypropylene (Fig. 3). Five were identified as polyethylene, out of which two were polyethylene compositions (one with Cobalt phthalocyanine and the second with acrylic acid), and two were identified as polypropylene (see Fig. 3).

The oceanic origin of the air masses as indicated by the HYSPLIT back trajectories, together with the chemical evidence from samples 38–39 and their corresponding seawater MP samples strongly suggest marine source of the AMP. Additionally, AMP sample 38 has a calculated terminal velocity of 0.2 m s⁻¹, and 39 of 0.003 m s⁻¹, yielding atmospheric residence times of ~5 min and ~48 h, respectively (Table 1). This strongly suggests that both were produced in the ocean. AMP sample 38 was most likely locally produced, and sample 39 with a potential residence time of 48 h, may have been produced far from the sample collection point (the residence time only indicates the maximal time a particle can spend in the atmosphere, and therefore could have also been emitted very shortly before sampling), but is also likely of oceanic source, as the full 48 h duration of the HYSPLIT back trajectory was spent over the ocean.

Furthermore, the specific densities of most AMP we found, and specifically polyethylene and polypropylene found in the seawater and in AMP samples 38 and 39, are lower than the specific density of seawater (Table 1), and are therefore likely to accumulate in the sea surface microlayer, and thus can be readily aerosolized.

Our findings indicate a new possible source and formation mechanism for AMP, different to those found for the terrestrial and coastal AMP detected to date^{13,22–33}. We have evidence from the open ocean, which suggest that AMP is emitted from the seawater into the atmosphere through wind related processes. MP is accumulated in the ocean surface microlayer, where it can be scavenged by rising air bubbles. The bursting of the bubbles

injects the MP into the atmosphere in a process associated with sea spray aerosol formation^{51,52}.

This implies that the ocean can be a constant global source of AMP, which may be transported to coastal regions and remote locations. However, we cannot dismiss the possibility that some AMP particles may have originated from other boats, continents or islands³³, and transported into the remote marine atmosphere.

We found AMP in ~20% of our atmospheric samples. We postulate that these results underestimate the actual AMP concentration in the marine atmosphere due to the current technical challenges, which did not allow us to analyze airborne microplastic particles smaller than 5 μm (see “Methods” section for more details). Moreover, nanoplastic particles (<1 μm) were only very recently identified in the environment at the single particle level²⁸, due to methodological limitations in nanoplastic collection and detection^{44,53}. In a typical marine aerosol size distribution, the diameter range between 10 nm–1 μm constitutes >99% of the total marine airborne particle concentration⁵², as was also measured during the R/V *Tara* Pacific expedition³⁸ (see Table 1 and Fig. 5 of their paper). It is therefore likely that a higher concentration of microplastic and nanoplastic particles, below our current size detection limit are suspended in the air. Despite the technical limitations in tracing them, it is widely accepted, and recently confirmed, that nanoplastic particles are abundant in the marine environment^{9,28,44,45,53,54}. Therefore, we suggest that the marine atmosphere contains nanoplastic particles (10 nm–1 μm), which would be much more easily introduced and sustained in the marine atmosphere than the large particles presented in this study.

The presence of nanoplastic and microplastic in the marine atmosphere may have implications on the atmospheric composition, and may also increase microplastic toxicity due to exposure to UV radiation in the atmosphere^{1,2,36}, with possible severe consequences for marine microbial ecology. The consequences may include increased stress response, and a decrease in metabolism, growth, and reproduction, which are some of the phenomena observed in aquatic organisms interacting with microplastics⁵⁵. It is known that microplastics and nanoplastics, particularly polystyrene, polyethylene, and polypropylene (all found in our AMP samples), accumulate persistent organic pollutants⁵⁶ as well as heavy metals^{44,57,58}. The products of plastic degradation, particularly polystyrene, found in our AMP samples, are toxic^{58–60}. Given that some AMP can persist in the air for days, according to our terminal velocity calculations, allowing

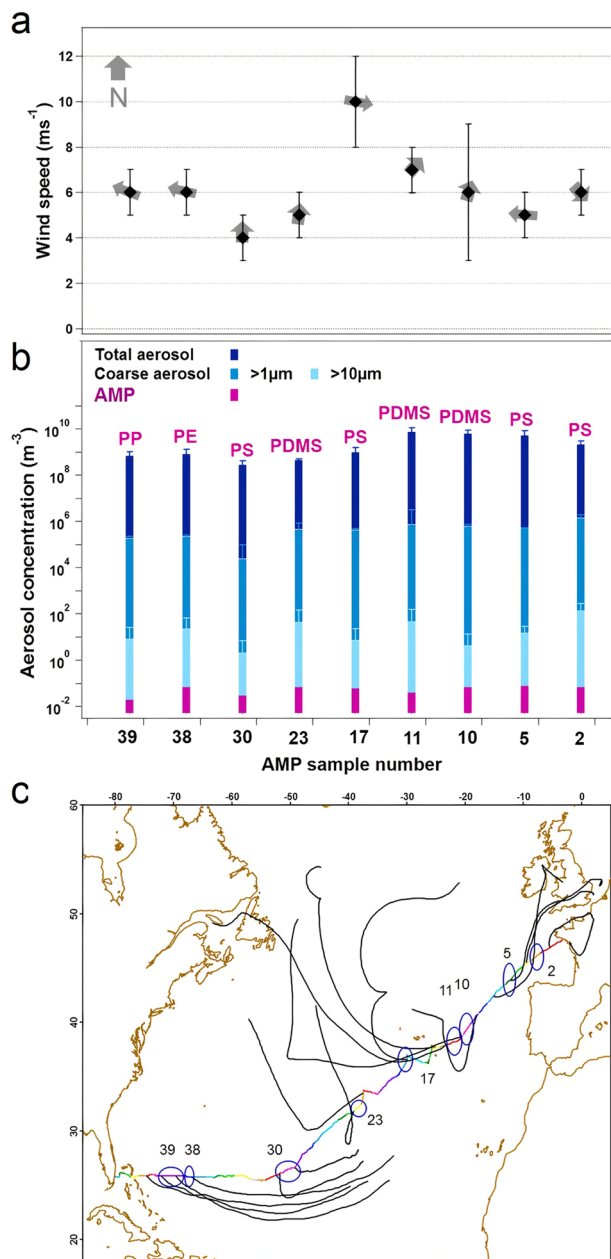


Fig. 2 AMP and total aerosol concentrations related to wind and air mass back trajectories. **a** Average wind speed and direction (gray arrows)³⁸ during AMP sample collection. The error bars represent the standard deviation of the wind speed. **b** AMP, total, and coarse mode aerosol concentrations per m^3 for each sample. AMP are marked in magenta, total aerosol are dark blue, and coarse mode aerosol are marked in cyan and blue for $>10\ \mu\text{m}$ and $>1\ \mu\text{m}$, respectively. Sample numbers are presented in the direction following the R/V *Tara* transect presented on the map. **c** Map of 48 h HYSPLIT back trajectory ensembles for each point along the R/V *Tara* transect in the Atlantic Ocean where AMP were detected³⁸. These are average trajectories of the HYSPLIT “Ensemble option” that were calculated based on an endpoint at 250 m height, the minimum height for the optimal configuration of the ensemble. The vast majority of the air masses originate from the ocean for the full extent of the 48 h.

long exposure times to UV radiation and oxidation, they can become highly toxic^{1,2,36} before being redeposited into the seawater, where they can cause further harm to the marine ecosystem than previously observed for non-airborne microplastic particles^{55,61}. The ability of microplastic to carry pathogens^{1,2,14–}

16,55,61–66 may also increase the harm posed by AMP to the marine ecosystem, as airborne microplastic can carry pathogens further than typical ocean currents. According to our calculations, some AMP can travel 100’s to 1000’s of km, rendering distant marine ecosystems in danger of infection by these pathogens, provided they remain infective in the atmosphere, as shown possible in several studies^{67,68}. Therefore, the effect of AMP can be devastating for marine microbial ecology, and must be thoroughly investigated¹⁷.

This study is the first to detect and identify AMP in the remote marine atmosphere. The implications of these findings on AMP emissions and their fate, the possible effects on the marine atmosphere and on marine microbial ecology, as suggested above, require further investigation¹⁷.

Methods

Atmospheric samples were collected during the *Tara* pacific expedition 2016–2018^{37,39} using the collection system illustrated in Fig. 4³⁸. The collection system is composed of an inlet comprised of a funnel mounted on the rear backstay of the R/V *Tara*, and connected with conductive tubing (1.9 cm inner diameter) to filter holders (AHF analysentechnik AG). The air was pulled into the system by a vacuum pump at a flow rate of ~ 20 LPM (liter per minute). Aerosol samples were collected on polycarbonate membranes (Isopore membranes, Millipore Ltd., $0.8\ \mu\text{m}$ pore size), with collection volumes of $\sim 12\text{--}60\ \text{m}^3$.

We used micro-Raman spectroscopy to analyze the samples collected in the North Atlantic Ocean (28th May–28th June 2016) in order to detect and chemically identify airborne microplastic particles (AMP). We note that during this period the inlet was installed halfway up the backstay, ~ 15 m above sea level.

We collected 46 filters (samples 1–46) from the Atlantic transect presented in Fig. 1. Three of the 46 samples had to be discarded for technical reasons, and therefore could not be analyzed.

We used the HORIBA micro-Raman system to detect and identify microplastic particles. The LabRAM HR Evolution (HORIBA, France) set-up was used for this analysis. The excitation was performed mostly with a 633 nm laser (but 532 and 785 nm were occasionally used as well). The set-up uses an 800 mm spectrograph, which allows for a high spectral resolution and low stray light. The pixel resolution is $\approx 1.8\ \text{cm}^{-1}$ when working with a 600 g/mm grating and a 633 nm laser. The sample was illuminated using a $\times 50$ LWD NA 0.5 objective (LMPlanFL N, Olympus). The Raman spectra were measured using a 1024×256 pixel open electrode front-illuminated CCD camera, cooled to $-60\ ^\circ\text{C}$ (Syncerity, HORIBA, USA). The system utilizes an open confocal microscope (Olympus BXFEM) with a spatial resolution greater than $1\ \mu\text{m}$. The measurements were done with the laser beam focused on a single microplastic particle at a time. Raman spectroscopy is a documented method for polymer characterization, and micro-Raman spectroscopy can be used for marine microplastic detection due to its high spatial resolution. It is well suited for the study of (marine) microplastic due to the small spot size, addressing even minute samples^{40–43}. Spectra were collected between $100\text{--}1800\ \text{cm}^{-1}$ or $100\text{--}3500\ \text{cm}^{-1}$, depending on the sample. For consistency, all spectra shown in this paper are in the $100\text{--}1800\ \text{cm}^{-1}$ range.

We used a library of Raman spectra of polymers available in the BioRad know-it-all software, searching for matches for the spectra of our particles. Our method allows us to detect particles as small as $\sim 5\text{--}10\ \mu\text{m}$, which is a limiting factor in this study, as plastic debris is known to produce also microplastic and nanoplastic particles smaller than $5\ \mu\text{m}$. The optical system can detect particles in this size range, or even smaller (up to $1\ \mu\text{m}$ particles), but as the particles are deposited on polycarbonate filters, even the small depth of focus of the confocal microscope produces a strong background signal from the filter, overshadowing the signal from the smaller particles.

We used the particle finder module (Horiba, France) to detect particles on each filter, and automatically analyze each particle detected by the software using Raman spectroscopy. The spectra were treated for background subtraction as is common in Raman spectroscopy. An area of $\sim 870\ \text{mm}^2$ was scanned in search of particles in each filter (total of 43 filters). The area scanned was the same distance from the edge of the filter for each of the 43 filters. Upon finding a particle or group of particles, we obtained Raman spectra for an area of $0.018\ \text{mm}^2$ in each scan of the particle finder software. Depending on the amount of particles found during the search executed for the entire filter (the $\sim 870\ \text{mm}^2$ scan), 30–100 scans were performed using the software, resulting in a total scanned area between 0.54 and $1.8\ \text{mm}^2$. Since polycarbonate is the material from which the filters are composed, the Raman signal of polycarbonate often appeared in the obtained spectra, and was removed from the analyzed spectra of the particles when possible.

Sampling and handling blanks. In order to assure that the AMP we identified were not emitted from any surface onboard the R/V *Tara* or any part of the boat, and are not contaminations from any stage of sampling onboard the *Tara*, sample handling in the laboratory or the Raman spectra analysis, very strict

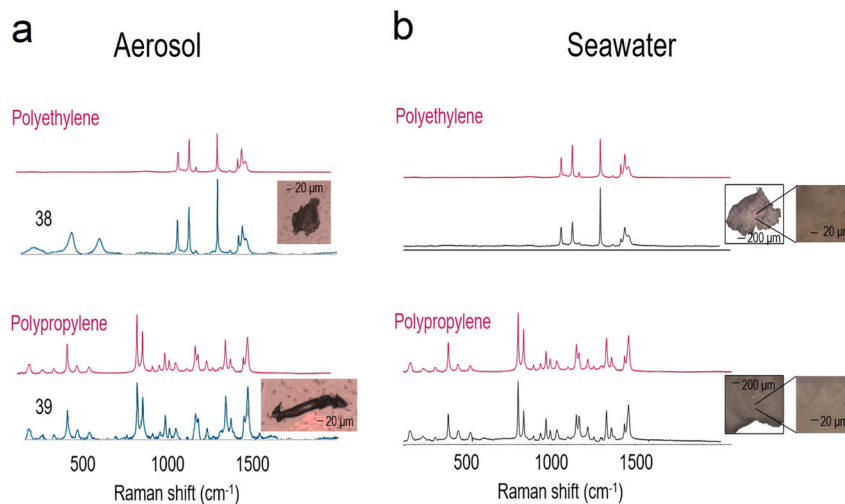


Fig. 3 Comparison between airborne and seawater microplastic particles. **a** Raman spectra of airborne microplastic samples collected between June 22nd and 23rd 2016, between 26°N 66°W and 26°N 72°W (filters 38–39). **b** Raman spectra of seawater microplastic samples collected on June 22nd, 2016 at 26°N 68°W. Optical microscope images are presented for both airborne and seawater particles. Close up images of the locations where the Raman spectra were retrieved in the seawater microplastic particles are presented. The Raman spectra of the corresponding plastic polymer standards are shown in magenta.

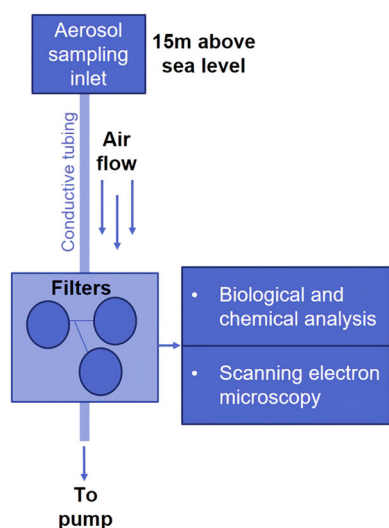


Fig. 4 Illustration of the atmospheric aerosol collection system onboard the R/V Tara. The system is composed of an inlet, positioned 15 m above sea level, which is connected to a flow splitter leading to the filter holders. Each filter holder contains a different filter, for biological analysis, for chemical characterization, and for morphology using SEM. Air is being pulled from the inlet through all filter holders by a vacuum pump. The air flow rate through each filter was approximately 20 LPM (liter per minute), as measured by a flow-meter³⁸.

measures were taken: two types of blanks were collected and analyzed using Raman spectroscopy to obtain their chemical identity in the exact same manner as the collected atmospheric samples. Firstly, all surfaces onboard the R/V *Tara* were sampled: the ropes (blue and red), the electricity cords, the flags, the masts and the sails. Raman spectra of the particles sampled from all the surfaces were obtained. The main component found was cellulose. Figure 5 shows Raman spectra of the R/V *Tara* surface blanks compared with the Raman spectra standards of the chemical compound corresponding to the blanks, which was cellulose. Figure 5a shows the Raman spectrum of cellulose, Fig. 5b shows typical Raman spectra of the sampled surfaces, corresponding to the spectra of cellulose, demonstrating the abundance of cellulose in various parts onboard the R/V *Tara*. Many of the surfaces onboard *Tara* emitted cellulose fibers and particles into our sampling inlet, and therefore cellulose particles from the *Tara* surfaces

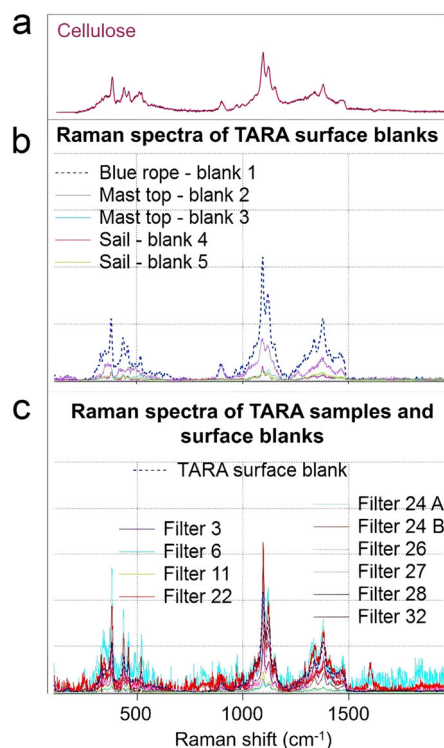


Fig. 5 The Raman spectra of Tara surface blanks identified as cellulose. **a** The reference Raman spectrum of cellulose. **b** Raman spectra of blanks collected from various surfaces onboard the *Tara*, taken from the ropes, masts, and sails, all corresponding to cellulose. **c** The cellulose spectrum obtained from the *Tara* surface blanks, and the spectra obtained from particles found in atmospheric samples 3, 6, 11, 22, 24, 26, 27, 28, and 32. A strong agreement between the spectra of the blanks and the samples is observed.

were detected in the atmospheric samples throughout the Atlantic transect (samples 1–46), as shown in Fig. 5c. The cellulose particles were disregarded as atmospheric particles (AMP) since they were found in the R/V *Tara* surface blanks, the same treatment was applied to any other materials found in any of the R/V *Tara* surfaces. The other materials identified on the surfaces were

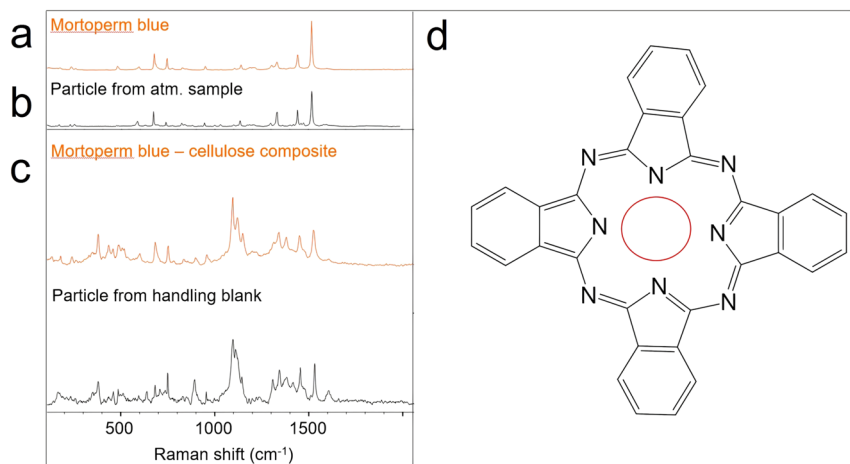


Fig. 6 Mortoperm blue in aerosol blanks and samples. **a** The Raman spectrum of Mortoperm blue. **b** The Raman spectrum of an atmospheric sample containing Mortoperm blue. **c** The Raman spectrum of a handling blank containing a composite of Mortoperm blue with cellulose. **d** The basic molecular structure of Mortoperm blue and Copper/Cobalt phthalocyanines. Copper/Cobalt phthalocyanines contain either a Copper (Cu) or a Cobalt (Co) ion at the center of the molecule, while Mortoperm blue doesn't contain any metal ion in the center. The center of the molecule is marked with a red circle.

polyester, which was detected on one of the ropes (red rope), and Copper/Cobalt phthalocyanines (CoPhc), or Mortoperm blue, which was widely abundant in the handling blanks as elaborately explained below. Secondly, handling blanks from all aerosol sampling and analysis stages, from sampling into the filter holders (Fig. 4), and until analysis with micro-Raman Spectroscopy, were collected and analyzed with Raman spectroscopy. The handling blanks include the following treatments for filters: new filters taken out from the case, filters inserted and removed from the filter holders without aerosol collection, filters placed in the vicinity of the sample filter during microscopy and/or Raman analysis. One type of material in particular, which belongs to a family of very similar molecules called Copper/Cobalt phthalocyanines (CoPhc), or Mortoperm blue (a commercial name), was dominant in the handling blanks. It also appeared repeatedly in the atmospheric samples (in ~ 40 out of the 43 samples analyzed), as well as in a seawater sample from *Tara* from 22.6.2016 at 26°N 68°W (see main text; comparison between microplastic particles in airborne and seawater samples). It appeared in both handling blanks and the samples as a single material, or in composite with other materials, such as cellulose. CoPhc and Mortoperm blue have the same molecular structure (Fig. 6d), where the CoPhc contain either Copper (Cu) or Cobalt (Co) as the central ion of the molecule, and Mortoperm blue does not contain any additional metal in its center. An example of the Raman spectrum of Mortoperm blue (as a standard), an atmospheric sample containing Mortoperm blue, and a handling blank containing a composite of Mortoperm blue with cellulose is shown in Fig. 6a–c. CoPhc/Mortoperm blue are used as dyes and colorants in the plastic industry, and are therefore associated with plastics. However, although abundant in particles found in our atmospheric samples, and detected also in the *Tara* Pacific seawater samples, as well as in microplastic samples in aquatic environments from previous studies^{43,69}, we ruled them out as valid samples originating from the marine atmosphere, and categorized them as contamination from handling blanks during sampling and analysis. The size range of the particles, which were found in the blanks and/or airborne samples was between ~1 and 100's μm .

Other materials found in our blanks were not found in any of the atmospheric samples (1–46), or were ruled out and categorized as contamination, and are therefore not given any further attention here. We also note that cellulose and CoPhc/Mortoperm blue were detected in both R/V *Tara* surfaces, and in the handling blanks, as well as in the atmospheric samples. This suggests that these materials from the R/V *Tara* surfaces may reach the atmosphere, in which case the boat might be a source of cellulose and CoPhc/Mortoperm blue in the atmosphere, and maybe also in the seawater. Polyester, which was found on a surface onboard *Tara* was not found in any atmospheric sample, perhaps due to its high density (~1.35 g cm^{-3}). It may settle from the *Tara* surfaces into the seawater, but we have no evidence that it is emitted into the atmosphere.

Most importantly, none of the plastics found in our AMP aerosol samples were found in any of the blanks we collected and measured. This rules out with a very high confidence level that our AMP samples are a result of sampling, measurement or handling contamination, and confirms that they indeed originate from the marine atmosphere.

AMP terminal velocity calculations. AMP terminal velocity was estimated using the well-established theory and measurements of settling velocity of water droplets

and ice crystals with needle shapes, in the range of ~10–100 μm ⁷⁰. The particles shape, sizes, and densities that are close to water, allowed us to use this theory.

The terminal velocity of elongated particles in this size range, scales to the equivalent sphere of the same mass, but with a radius that scales to the largest dimension of the particle. This implies that the density of the equivalent sphere will be reduced by the ratio (α) of the real volume (V_r) of the particle and the volume (V_s) of the equivalent sphere:

$$\alpha = \frac{V_r}{V_s}$$

Then, to a good approximation, the AMP terminal velocity (μ) could be scaled as:

$$\mu = \alpha \mu_s,$$

where μ_s is the terminal velocity of a sphere (droplet) with diameter of the polymer's major axis.

More specifically: The real volume of the MP particle can be approximated as a spheroid by $V_r = \frac{4}{3}\pi ab^2$, where a is the major and b is the minor axis, and the equivalent sphere volume is $V_s = \frac{4}{3}\pi a^3$. Assuming that the MP density is close to water's density, $\rho_{mp} \approx \rho_w$, yields:

$$\alpha = \frac{V_r}{V_s} = \left(\frac{b}{a}\right)^2.$$

The terminal velocity of spheres can be described as a power-law function of the radius r : $V_t = \kappa r^\eta$, where κ and η depend on the sizes. Droplets with radii $r < 40 \mu\text{m}$ are in the laminar regime, for which $\kappa \approx 1.19 \times 10^6 \text{ cm}^{-1} \text{ s}^{-1}$, and $\eta = 2$. Droplets in the range of $40 < r < 600 \mu\text{m}$ are in the intermediate regime, between laminar and turbulent, with $\kappa \approx 8 \times 10^6 \text{ s}^{-1}$ and $\eta = 1.71, 72$.

Applying the above scaling to the measured AMP particles, yields settling velocities in the range of 0.001–0.2 m s^{-1} for the particles. For the most elongated cases, despite having long major axis, their settling velocities are in the range of 0.001 m s^{-1} (0.1 cm s^{-1}).

Seawater microplastic collection. Surface microplastics were collected using the High Speed Net (HSN) tow, deployed in the Atlantic and Pacific Oceans, and split into two equal subsamples. The first for plastic and plankton analysis by the ZooScan imaging system⁷³, and the second was used for genomic analysis³⁹.

Seawater microplastic concentrations along the North Atlantic transect presented in Fig. 1a were obtained from the microplastics collected with the HSN, and scanned with the ZooScan Imaging system. The images were separated to “living organisms” and “detritus” categories, the organisms were identified to the deepest possible taxonomic level, and the microplastics were selected from the images determined as “detritus”.

Seawater microplastic particles for micro-Raman analysis and comparison with AMP (see Fig. 3), were separated and identified after being scanned again with the ZooScan Imaging system, the second scanning was performed in order to assure that only particles identified as plastic with 100% confidence will be used for the micro-Raman analysis.

Data availability

Data are included in the article, and are publicly available through OSF at <https://doi.org/10.17605/OSF.IO/R3Q5C>. Additional data from Zooscan Tara Pacific 2016 2018 HSN 330 sn001, used in this paper (Fig. 1a), is available online: <https://ecotaxa.obs-vlfr.fr/prj/1700>.

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Author contributions

I.K. and A.V. conceived the basic idea and supervised the project. J.M.F. and M.T. designed and installed the atmospheric measurements suite onboard the Tara. J.M.F. and G.B. collected the samples. M.T. and I.P. performed Raman analysis and interpreted the data. M.L.P. and F.L. performed seawater analyses. J.M.F., G.G., E.B. and Y.R. contributed to data interpretation and writing. M.T., I.K., and A.V. wrote the paper.

Competing interests

The authors declare no conflict of interest. Yinon Rudich is an Editorial Board Member for *Communications Earth & Environment*, but was not involved in the editorial review of, or the decision to publish this article.

Additional information

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