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Article

Plastics from Surface to Seabed: Vertical Distribution of (Micro)plastic Particles in the North Pacific Ocean

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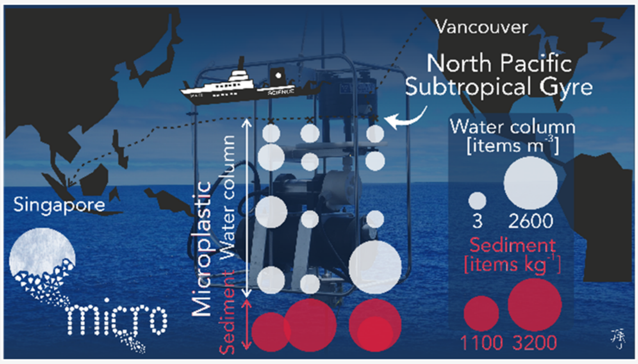
ABSTRACT: In this study, the vertical distribution of microplastic items across the water column and deep-sea sediments in the North Pacific Ocean, at depths exceeding 5 km, was investigated, covering two previously detected hotspots of surface-floating plastic items, the North Pacific Subtropical Gyre and the World Heritage Site Papahānaumokuākea Marine National Monument, and a less polluted open ocean site between them. Concentrations ranged from 8 to 2600 items m^{-3} in the water column and 1100 to 3200 items kg^{-1} in sediments, with distinct distribution patterns between stations. Our results demonstrate that microplastics are ubiquitously distributed vertically throughout the study sites in the North Pacific Ocean. The similar polymer composition across the samples, dominated by polyethylene and polypropylene, suggests that the plastic items originate from surface fallout, indicating vertical transport as a crucial dispersion pathway. The high abundance of small plastics at the analytical size detection limit of 11 μm is particularly concerning and underscores the necessity to analyze even smaller particles. This study substantially contributes to narrowing critical knowledge gaps regarding distribution and transport pathways of marine plastics in the North Pacific Ocean and provides crucial information for environmental management and policy actions.



KEYWORDS: *marine pollution, marine litter, anthropogenic debris, water column, sediment, FT-IR imaging*

INTRODUCTION

The pollution of the environment with micro- and nanoplastics (MNPs) is widely recognized as a global issue and, as part of "novel entities", classified as a planetary boundary threat.[1,2] Due to the ever-increasing global production of plastics, which reached 413.8 Mt in 2023, in combination with insufficient waste management strategies, the amount of plastics ultimately reaching the environment is expected to increase further.[3−5] Plastic particles are a ubiquitous contaminant across a wide range of environmental compartments, including the atmosphere,[6] agricultural soils,[7] freshwater bodies,[8] and the marine environment, from beaches[9] to surface[10−12] and subsurface waters.[13−15] Furthermore, plastic particles were found in the most remote regions, such as the Arctic[12] and the Southern Ocean,[16] and ranging from the highest mountains[17] to the deepest regions of the world's oceans.[18,19]

In the marine environment, plastic particles, especially in the size range of microplastics (1−5000 μm), have been found in organisms across various trophic levels, ranging from zooplankton[20] to fish,[21] marine mammals,[22] turtles,[23] and seabirds.[24] The ingestion of plastic items has been associated with physical obstruction, inflammation, reduced nutrient uptake and toxic effects on both, cellular and physiological processes, indicating their harmful potential on the marine ecosystem.[25,26] Furthermore, plastic items may act as vectors for hydrophobic contaminants, heavy metals, pathogenic microorganisms, and invasive species, potentially amplifying their environmental and health impacts.[26] To better understand such impacts, a comprehensive understanding of the fate and distribution of plastic items in the marine environment is crucial to realistically assess the effects on marine life and to design efficient mitigation strategies.[25]

Despite the recent increase in research addressing marine plastic pollution, including advancements in analytical techniques and numerical modeling, our understanding of

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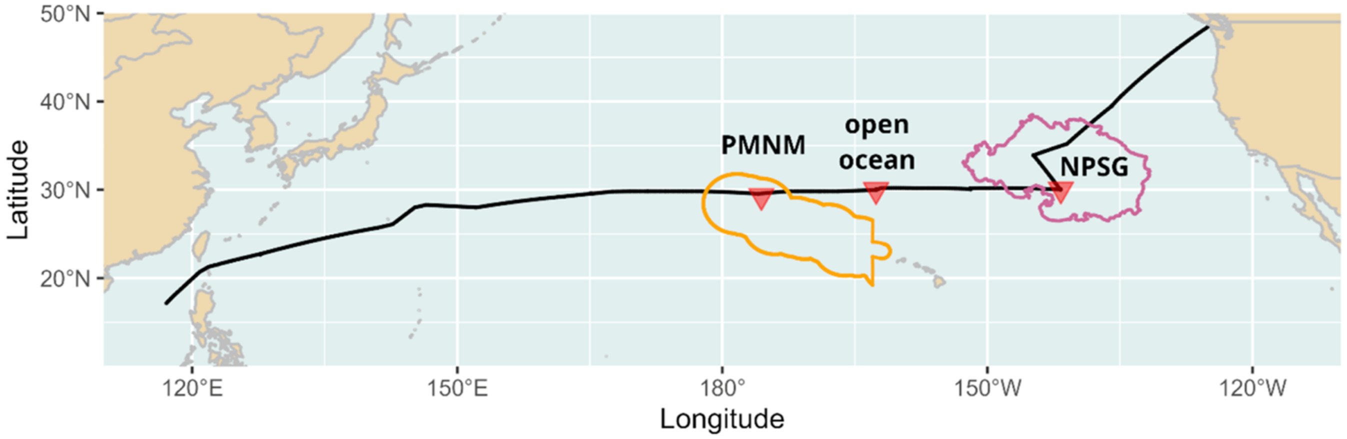

Figure 1. Map of the North Pacific Ocean with the cruise track of RV SONNE expedition SO268/3 in 2019 (black line), the boundary of the North Pacific Subtropical Gyre (red), and the border of the Papahānaumokuākea Marine National Monument (orange).[31,32] Red triangles represent the positions of the sampling stations. Samples were collected within the typical location of the Subtropical Convergence Zone (STCZ) along 30°N.

the distribution of MNPs in the oceans is still limited. Due to the extension, complexity, and variability of marine environments, the sampling, quantification, and modeling of the distribution of plastic items is still challenging. Microplastic distribution is influenced by oceanic features such as currents, gyres and fronts, leading to the formation of hotspots.[27] However, there are still critical knowledge gaps regarding their vertical transport, deposition in the deep sea, and long-term environmental fate. Understanding the distribution, transport, and accumulation mechanisms of plastic pollution is ultimately key to developing effective mitigation strategies.[28] Stable conditions in the deep-sea and the deep-ocean sediments (absence of light, movement, and sheer forces at constant temperatures of about 4 °C) favor the persistence of plastics. For these areas, data is scarce but key to assess the overall plastic pollution and to understand the gap between suspected and observed quantities of MNPs in the environment.[29,30]

In this study, we assessed the vertical distribution of microplastic items larger than 11 μm directly beneath two previously identified hotspots of surface-floating plastic in the North Pacific Ocean, as well as a third area with a substantially lower surface plastic concentration in the open ocean.[10] The aim of this study was to compare the concentrations, polymer types, and size distributions of plastic items from the sea surface to the deep-sea sediments of the three stations to (i) detect patterns throughout the sea surface, water column, and sediment, (ii) identify possible vertical transport mechanisms, and (iii) address existing knowledge gaps regarding the occurrence and distribution of plastic items in the marine environment.

■ MATERIALS AND METHODS

Sampling. Samples were collected during the research vessel RV SONNE expedition SO268/3 (30 May to 5 July 2019) from three stations between Vancouver (BC, Canada) and Singapore. Sampling sites were selected based on a previous study on surface-floating plastic particles, covering two areas with high concentrations, the North Pacific Subtropical Gyre (NPSG, station SO268/3-6) and a World Heritage Site, the Papahānaumokuākea Marine National Monument (PMNM, station SO268/3-17), as well as one area with a lower concentration of plastic items in the surface waters (station SO268/3-10), hereafter referred to as "open ocean" station (Figure 1).[10] Detailed sampling-related information is provided in the Supporting Information (SI, Tables S1 and S2).

Particle samples from the water column were obtained *in situ* using four Large-Volume Water Transfer Systems (WTS-LV, McLane Research Laboratories, Inc.) which were attached to a standard CTD cable. The samples were collected during CTD profiling at the following depths: the maximum chlorophyll-a concentration (110−130 m), 300, 2500, and 50−70 m above the seafloor (5000−5400 m). Each WTS-LV was equipped with two stacked stainless-steel filters with mesh sizes of 35 and 10 μm (Drahtweberei Pausa GmbH, Germany). The pumps were held at the desired depth for 1−2 h to filter water volumes ranging from 260 to 744 L (Table S1), resulting in a total time of 6 to 7 h per sampling including lowering and lifting the equipment to the desired depths. Strict measures were taken to prevent contamination of the samples on board as described in Rynek et al.[10] Filters were transferred from the WTS-LV into precleaned 0.5-L wireframe glass jars (Flaschenland GmbH, Germany) directly on deck, covered with aluminum foil to prevent contamination from jar lid seals, sealed, stored, and transported frozen until processing in our laboratories. For the water filtrates, we generated sampling controls near the end of the sampling campaign as follows: one McLane pump system was successively operated two times with 52.2 and 50.0 L of water obtained from the RV's Milli-Q supply in the identical setup as used for sampling of the water column. The water was stored in 50 L stainless-steel containers and covered with aluminum foil during processing. The blank sample filters were immediately transferred to identical glass jars as used for the samples, sealed, stored, and shipped to our institute where they were processed alongside the samples.

Deep-sea sediments were sampled using a multicorer (MUC, MC 12-20 × 100, Oktopus GmbH, Germany, inner diameter of core liners 9.6 cm) at depths of 5020 m (NPSG), 5660 m (PMNM), and 5300 m (open ocean), resulting in a total sampling time of around 4 h per station. Upon reaching the RV, the core liners containing the sediment samples were immediately sealed and left standing on the deck for several hours to allow the suspended material to settle. Then, the upper 5 cm of sediment were subsampled on deck into

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aluminum trays that were immediately sealed with aluminum foil, placed in sealed polyethylene (PE) bags, stored, and shipped frozen until they were processed. As blank controls, bulk sediment material was collected from the lower 20−25 cm of two sediment cores at the NPSG and "open ocean" stations, sealed, stored, shipped, and processed in parallel to the samples.

Water Sample Preparation. After defrosting, the water column samples ($n = 24$) and process control samples ($n = 4$) from the WTS-LV were transferred into stainless-steel reactors semienclosed with stainless-steel filters (10 μm, Haver & Boecker OHG, Oelde, Germany).[33] The samples and sample controls were treated according to an enzymatic purification protocol, following the procedure used in the previous study on surface-floating plastic items.[10,34] Briefly, the samples were consecutively treated with sodium dodecyl sulfate (SDS, J.T. Baker), Protease A-01, Cellulase TXL (both ASA Spezialenzyme GmbH, Germany), hydrogen peroxide (H_2O_2, CHEMSOLUTE, Germany), Chitinase (ASA Spezialenzyme GmbH, Germany), and again with H_2O_2.

After the last step of the purification protocol, the steel mesh, the 10-μm steel filters, and the inner surface of the reactors were thoroughly rinsed with 300 mL of Milli-Q water (Milli-Q Direct 8, Merck KGaA, Germany) into glass beakers. The resulting suspension was filtered onto a stainless-steel mesh (10 μm, Haver & Boecker OHG, Oelde, Germany) and afterward placed in glass centrifuge vessels. The vessels were filled with a prefiltered (0.1 μm) high-density solution of sodium iodide (ρ = 1.7 g/cm^3, CHEMSOLUTE, Germany), shaken for 1 min, and centrifuged for 3 min at 2000 rpm. Afterward, the lower half of the centrifuge vessel was placed in a cooling bath (ethanol/dry ice) to immobilize high-density particles by freezing. The supernatant containing the plastic particles was filtered onto aluminum oxide filters (Anodisc, 0.2 μm pore size, 25 mm diameter, Whatman PLC, U.K.), rinsed with a minimum of 200 mL of Milli-Q water, placed in Petri dishes, and dried overnight at 50 °C in an oven (Heraeus T6120, Thermo Fisher Scientific).

Sediment Sample Preparation. Sediment samples (n = 4, duplicate at the NPSG station) and process controls ($n = 2$) were freeze-dried in aluminum sample containers (α 1-4 LSCplus, Martin Christ Gefriertrocknungsanlagen GmbH, Germany). Afterward, manganese nodules present in the sediments were removed, and aggregates in the fine sediment were carefully disintegrated using a spatula. To extract putative plastic particles via density separation, 40.0 g of homogenized sediment was weighed into a 1 L glass beaker ($n = 2$ per sample and $n = 1$ for process control), followed by the addition of 800 mL of the prefiltered sodium iodide solution. The beaker was covered with aluminum foil, and the suspension was stirred thoroughly using a magnetic stirrer for 30 min, left to settle for 1 day, stirred again gently for 30 min, and finally left to settle for another 48 h. Then, the beaker was transferred into a cooling bath of ethanol and dry ice to immobilize the precipitate by freezing and then transferred to a freezer (−80 °C) to completely freeze the suspension.

After 1 day, the frozen suspension was taken out from the freezer, and the upper 200 mL was evenly rinsed into a 600 mL glass beaker using Milli-Q water (40−50 °C). The resulting suspension was filtered onto a stainless-steel mesh (10 μm), placed in a glass beaker and treated with Fenton's reagent to remove organic matter.[35] To this end, 10 mL of a prefiltered (0.1 μm) 7.2 mM solution of $FeSO_4$ (CHEMSOLUTE, Germany) were added to the beaker. The beaker was then placed in a water bath, and 20 mL of H_2O_2 was added. After 15 min, the suspensions were filtered onto aluminum oxide filters (Anodisc, 0.2 μm pore size, 25 mm diameter, Whatman PLC, U.K.), rinsed with a minimum of 200 mL of Milli-Q water, placed in Petri dishes, and dried overnight at 50 °C before analysis.

FT-IR Imaging. Filter samples containing putative plastic items were analyzed in transmission mode using a Cary 620 Fourier transform infrared spectroscopy (FT-IR) microscope equipped with a 15× objective and a 128 × 128 Lancer MCT FPA detector array (Agilent Technologies). The Anodisc filters containing the samples were placed on a steel filter holder and covered with a BaF_2 window (Korth Kristalle GmbH, Germany). FT-IR measurements of the whole filter area were conducted with 8 scans in the spectral range of 3800−1250 cm^{-1} and a spectral resolution of 8 cm^{-1}, resulting in a spatial resolution of 5.5 × 5.5 μm per spectrum. Data analysis was carried out using Microplastics Finder v. 4.09 (Purency GmbH, Austria).[36] Postprocessing was done by excluding the polypropylene (PP) support ring of the Anodisc filter and particle assignments with less than two pixels, implementing a minimum particle size of 11 μm or with relevance and similarity scores below 0.6. The particle size was determined based on the FT-IR images and reported as the Feret diameter.

Calculation of Concentrations. Concentrations of plastic items >330 μm at the sea surface were taken from our previous study on surface-floating plastic items, corrected for wind-induced vertical mixing caused by differences in sea states during sampling to account for varying sea states.[10] Concentrations of plastic items from water column samples for each polymer $c_{\text{water,polymer}}$ were calculated using the number of detected particles per polymer $N_{\text{sample,polymer}}$, the average number of particles of the respective polymer in the blank samples ($n = 2$) $N_{\text{blank,polymer}}$, and the volume of water that was pumped by the WTS-LV $V_{\text{WTS-LV}}$ extracted from the pump logs (eq 1).

$$c_{\text{water,polymer}} = \frac{(N_{\text{sample,polymer}} - N_{\text{blank,polymer}})}{V_{\text{WTS-LV}}}[\text{items m}^{-3}] \tag{1}$$

Concentrations of plastic items in sediment samples $c_{\text{sed,polymer}}$ were calculated using the number of detected particles per polymer, the combined number of particles of the respective polymers detected in the blank samples ($n = 2$), and the dry mass of sediment used for the polymer analysis $m_{\text{sediment,dry}}$ (eq 2).

$$c_{\text{sed,polymer}} = \frac{(N_{\text{sample,polymer}} - N_{\text{blank,polymer}})}{m_{\text{sediment,dry}}}[\text{items kg}^{-1}] \tag{2}$$

The reported data represent the total concentration of plastic items c_{total} collected on both filters, obtained by summing up the concentrations of all single polymers detected in both filter fractions of the respective station and depth (eq 3).

$$c_{\text{total}} = \sum_{\text{polymers}} c_{\text{polymer}} \tag{3}$$

Statistical tests (Pearson correlation, Spearman rank correlation) were carried out, and plots were generated in RStudio (v. 2024.12.1) using R (v. 4.4.0) and packages ggplot2

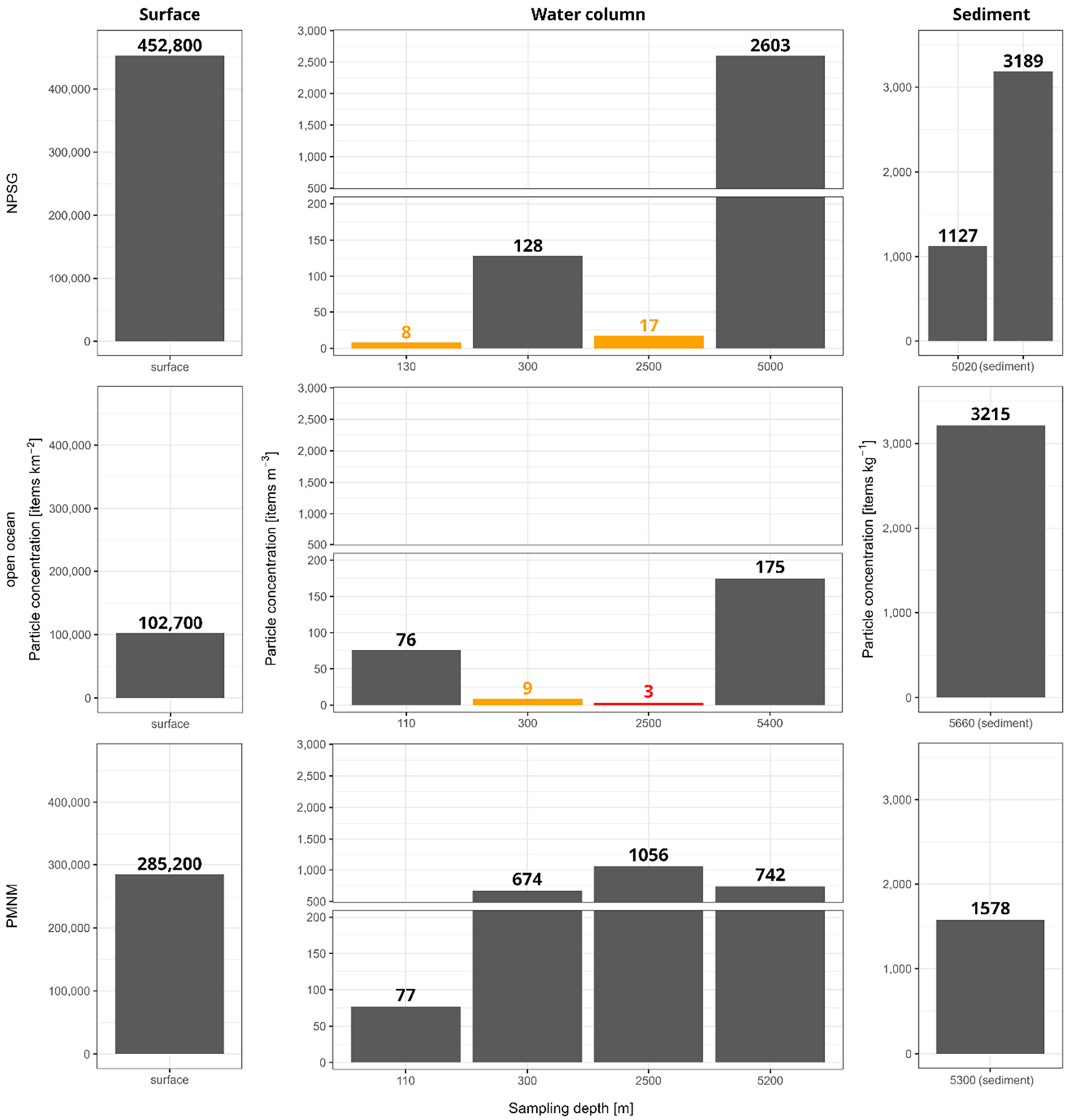

Figure 2. Distribution of plastic item concentrations at the sea surface (larger microplastics of >330 μm size, left), in the water column at different depths (smaller microplastics of >11 μm size, middle), and in the sediment (right) in the North Pacific Subtropical Gyre (NPSG), at the open ocean station and in the Papahānaumokuākea Marine National Monument (PMNM). Samples were gathered during RV SONNE expedition SO268/3 in 2019. The sample marked in red (open ocean, 2500 m depth) must be viewed with caution as lowest estimate due to loss of the filter fraction >35 μm during cleanup; for details, see the text. The concentrations marked in orange are calculated based on less than 10 particles above blank levels and therefore require cautious interpretation.

(v. 3.5.1), ggpubr (v. 0.6.0), tidyverse (v. 2.0.0), scales (v. 1.3.0), ggbreak (v. 0.1.2), patchwork (v. 1.3.0), cowplot (v. 1.1.3), ggplotify (v. 0.1.2), sf (v. 1.0-17), and oce (v. 1.8-3).

Quality Assurance and Quality Control (QA/QC). To minimize process contamination, the water column filter samples were directly transferred to airtight containers on the ship's deck, sealed, stored, and shipped frozen to our laboratories. Sediment samples were directly transferred from the MUC to aluminum containers on the ship's deck, sealed with aluminum foil, placed in a PE bag, stored, and shipped frozen. Control samples were used as negative controls and treated as environmental samples to assess possible background contamination. To minimize airborne contamination during sample processing, all preparation steps in the laboratory were conducted under a safety workbench (HeraSafe HS9, Kendro Laboratory Products GmbH, Germany), in semienclosed reactor vessels or in aluminum foil-covered vessels under a laboratory fume hood. All used laboratory equipment was thoroughly rinsed with Milli-Q water and prefiltered ethanol (96%, CHEMSOLUTE, Germany, < 1 μm) and afterward covered with aluminum foil until use.

The use of plastic materials was avoided wherever possible. Plastic materials used during sampling and sample processing were polyoxymethylene (POM, pump head of the WTS-LV), polycarbonate (PC, liner of multicorer), polytetrafluoroethylene (PTFE, sealing of reactors), ethylene tetrafluoroethylene (ETFE, squeeze bottle), and PP (Anodisc support ring). While the area of the PP support ring of the Anodisc filters was comprehensively excluded from every sample, we cannot detect PTFE and ETFE due to the blockage of their IR signals by the used Anodisc filters; therefore, PTFE and ETFE are no source of blank contamination. The presence of POM and PC in control samples was monitored, and the polymers were excluded from the respective data sets if present in both controls and samples.

■ RESULTS AND DISCUSSION

QA/QC. All measured concentrations were blank-corrected for each polymer type to account for potential contamination of samples during storage, transport, sample preparation, and analysis. Raw counts of plastic items in samples and control samples (Table S3), as well as blank-corrected numbers of plastic items per polymer detected in the samples (Table S4), can be found in the Supporting Information. In two water column control samples, a total of 290 (185 and 105) plastic items was detected, predominantly composed of PE (79.7%, n = 142 and 90), followed by PP (11.7%, n = 29 and 5), polyethylene terephthalate (PET, 5.2%, n = 9 and 6), acrylonitrile butadiene styrene (ABS, 1.7%, n = 2 and 3), polystyrene (PS, 0.7%, n = 0 and 2), and polybutylene terephthalate (PBT), and polyurethane (PU, 0.3% each, n = 1). The mean particle count per polymer of the control samples was subtracted from the measured particle count in the actual samples. This method provides a particularly conservative estimate, given that the control blanks are much more likely to be contaminated by airborne plastic items than the samples due to the control blank generation process on board the RV. This assumption is supported by the fact that no relevant blank contamination was detected during the method development and validation process in our laboratory. Particles detected in the control samples (mean: 69 ± 82 μm; median: 45 μm) were slightly larger compared to the samples from the NPSG (mean: 60 ± 60 μm; median: 45 μm), open ocean (mean: 58 ± 61 μm; median: 37 μm), and comparable to the PMNM station (mean: 67 ± 80 μm; median: 43 μm). Furthermore, particles identified as POM (n = 63) were excluded from the water column data set to eliminate the possibility of contamination of the sample caused by the sampling device (pump head of the WTS-LV).

In the sediment control samples (n = 2, 40 g dry sediment each), a total of 67 plastic items were detected, containing PP (46.3%, n = 21 and 10) and PE (53.7%, n = 17 and 19), which were subtracted from the number of plastic items in the samples, matching the total amount of dry sediment (80.0 g) for both blank and samples.

Vertical Distribution in the Water Column. A total of 2006 plastic items were identified across 12 water column samples. Plastic items above blank levels were present in all water column samples with concentrations ranging from 8 items m^{-3}, recorded at 130 m depth at the NPSG station, to 2603 items m^{-3}, found at 5000 m depth at the NPSG station (Figure 2, middle). The 2500 m depth sample collected at the open ocean station showed concentrations of 3 items m^{-3}. This result must be viewed with caution as the lowest estimate because the filter fraction >35 μm was lost during the cleanup procedure. The other concentrations were calculated using the sum of both filter fractions. The number of detected particles was comparable or higher in the smaller size fraction (factors 0.95−15.0, mean 4.9 ± 4.1) except for one station, where it corresponds to about one-fourth of the particle numbers on the larger size fraction (factor 0.24, NPSG at 300 m depth). This finding suggests that the total concentration of plastic items in the 2500 m depth sample from the open ocean station may not have been substantially higher than 3 items m^{-3}. A total of 720 surface-floating plastic items >330 μm were identified in the respective surface water samples in our previous study using a neuston catamaran, resulting in plastic item concentrations ranging from 102,700 items km^{-2} at the open ocean station to 452,800 items km^{-2} at the NPSG station (Figure 2, left).[10]

The lowest concentrations of plastic items in the water column samples, detected at depths of 130 and 2500 m at the NPSG station and 300 and 2500 m at the open ocean station, were calculated based on particle counts of less than 10 particles after blank correction and therefore require cautious interpretation. The mean concentrations (±standard deviations) of plastic items across the four depths in the water column at the sampling locations were found to be 1110 ± 354 items m^{-3} (NPSG), 66 ± 69 items m^{-3} (open ocean), and 637 ± 354 items m^{-3} (PMNM), exhibiting distinct distribution patterns along the depth profile (Figure 2).

While the highest concentrations of plastic items at the NPSG and the open ocean stations were detected near the seafloor at depths of 5000 and 5400 m, respectively, the PMNM site showed the highest plastic item concentrations at a moderate depth of 2500 m, with comparable concentrations at depths of 300 m and near the seafloor. This pattern suggests a more homogeneous distribution of plastic items across the water column at the PMNM station compared with the other stations. In contrast to a recent meta-analysis by Zhao et al., we did not observe a systematic decrease of plastic concentrations with increasing depths, but a high within-depth variability of concentrations between stations (e.g., 300 m: 9−674 items m^{-3}; 2500 m: 3−1056 m^{-3}).[37] This high variability in point measurements across the water column, spanning multiple orders of magnitude, was previously reported in the NPSG area, as well as on a global scale, and may be attributed to the contribution of the biological carbon pump to the vertical transport of plastics.[14,37] These findings indicate that the distribution of plastic items in the water column may be heterogeneous across the Pacific Ocean and is influenced by the particle properties and a wide range of different factors that are specific to the locations, such as biological interaction, large- and submesoscale open ocean processes, Langmuir circulation, internal tides, vertical mixing, and water column stratification.[27,38,39]

A correlation analysis between the concentrations of plastic items at the sea surface and the mean concentrations throughout the water column of the three sampling sites demonstrated a near-perfect Pearson correlation (r = 0.999, p = 0.02) and a perfect Spearman rank correlation (ρ = 1, p =

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Table 1. Concentrations of Plastic Items Observed across the Water Column in Different Areas across the Globe[a]

study	sampling method (size cutoff)	location/time	depths [m]	concentration [items m^{-3}]
this study	WTS-LV (>11 μm)	NPSG/June 2019	130−5000	1106 ± 354
		North Pacific Ocean/June 2019	110−5400	66 ± 69
		PMNM/June 2019	110−5300	637 ± 354
Zhao et al.[14]	WTS-LV (>25 μm)	outside NPSG/Nov−Dec 2018	75−2000	389 ± 109
		outer NPSG/Nov−Dec 2018	120−2000	283 ± 54
		inner NPSG/Nov−Dec 2018	30−3700	332 ± 140
Zhao et al.[15]	WTS-LV (>20 μm)	South Atlantic Subtropical Gyre/Jan 2019	10−5200	42 ± 54
Tekman et al.[12]	WTS-LV (>11 μm)	Arctic Ocean/summer 2016	1−5350	161 ± 293
Egger et al.[40]	MOCNESS (500 μm−5 mm)	NPSG/Nov−Dec 2018	2−2002	0.004−0.0325

[a] WTS-LV, Large-Volume Water Transfer System; MOCNESS, Multiple Opening and Closing Net with Environmental Sensing Systems.

Table 2. Concentrations of Plastic Items Detected in Sediment Samples[a]

study	sampling method (size cutoff)	location/time	depths [m]	concentration [items/kg]
this study	MUC (>11 μm)	NPSG/June 2019	5020	1127/3189
		Pacific Ocean/June 2019	5660	3215
		PMNM/June 2019	5300	1578
		mean ± SD	5020−5660	2277 ± 938
Peng et al.[51]	ARV with lander/box corer (>73 μm)	West Pacific Ocean/Dec. 2016	4800−10,890	53−133
Abel et al.[50,52]	MUC (>11 μm)	Kuril-Kamchatka Trench/Summer 2016	5143−8255	14−209
			5740−9450	215−1596
Tsuchiya et al.[19]	push corer/core sampler (>11 μm)	NW Pacific/Aug.−Sep. 2019	855−1387	29,600 ± 23,600
			5707−5813	601,500 ± 629,400
			9218−9232	11,200 ± 6000
Tekman et al.[12]	MUC (>11 μm)	Arctic Ocean/summer 2016	272−5569	239−13,331
Cunningham et al.[16]	MUC (no information)	Antarctic Peninsula/2018	499−1246	1300 ± 510
		South Sandwich Islands/2019	1619−3342	1090 ± 220
		South Georgia/2017 + 2019	136−3633	1040 ± 390

[a] MUC, multicorer; ARV, autonomous and remotely operated vehicle.

0.33), suggesting that high concentrations observed at the sea surface co-occurred with increased plastic item concentrations in the water column. This correlation was already indicated in an earlier study in the NPSG area for larger plastic items in the size range between 0.5 and 50 mm.[40] However, the number of samples (n = 12 from 3 stations) is limited, which is a consequence of the substantial sampling effort and long sampling times at such extended water depths. Given the resulting lack of statistical power and the effect of single high values (e.g., NPSG at 5000 m depth) driving this correlation, these results should be interpreted with caution. In addition to the challenges associated with sample collection, the low concentrations of particles represent a major analytical challenge, further contributing to the need for a cautious interpretation of the results. Additionally, no correlation was found between the plastic item concentrations and water parameters (salinity, chlorophyll-a content, turbidity, temperature, density) measured by the CTD probe during descent (Figure S1).

Despite these limitations, our results support the hypothesis that plastic items found in the water column below surface accumulation zones originate as "fallout" from surface-floating plastic items.[40] This hypothesis is further supported by the role of Ekman currents, which drive the accumulation of plastic debris in the centers of subtropical gyres and weaken substantially with increasing depth, particularly within the top 100 m of the water column. As a result, the reduced convergence in deeper layers allows the surface distribution patterns of plastic items to persist throughout the water column.[41,42] This reduced accumulation at depth aligns with the fallout hypothesis, suggesting that the increased concentrations of plastic items underneath the surface accumulation zones are likely not generated by subsurface currents, but rather from sinking of initially surface-floating plastic items after weathering and the influence of biological processes, both at the sea surface and across the water column.[43,44]

The concentrations of plastic items detected in the water column samples are of a comparable magnitude to those observed in previous studies conducted in the NPSG area and other oceanic regions using similar sampling techniques but partly different analytical approaches.[12,14,15,45] An additional study focusing on plastic items >500 μm in the NPSG area demonstrated concentrations that were several orders of magnitude lower, likely due to the much larger size cutoff of that study (Table 1).[40]

Compared to the studies by Zhao et al., the detected plastic item concentrations across the water column in this study were 2 to 5 times higher, which may partly be explained by the lower size cutoff of 11 μm used in the current study, compared to the 20 and 25 μm used in the previous ones.[14,15] This difference indicates that the abundance of plastic items increases with a decreasing size, most likely as a result of fragmentation processes of plastic items.[46] This is supported by a recent study, showing the ubiquity of nanoplastics across the water column of the North Atlantic Ocean.[47] Moreover, in addition to the data set by Egger et al., our study provides additional information about the distribution of plastic items by extending both the investigated particle size range to smaller particles and the sampled water depth, showing a comparable distribution pattern with elevated concentrations inside the

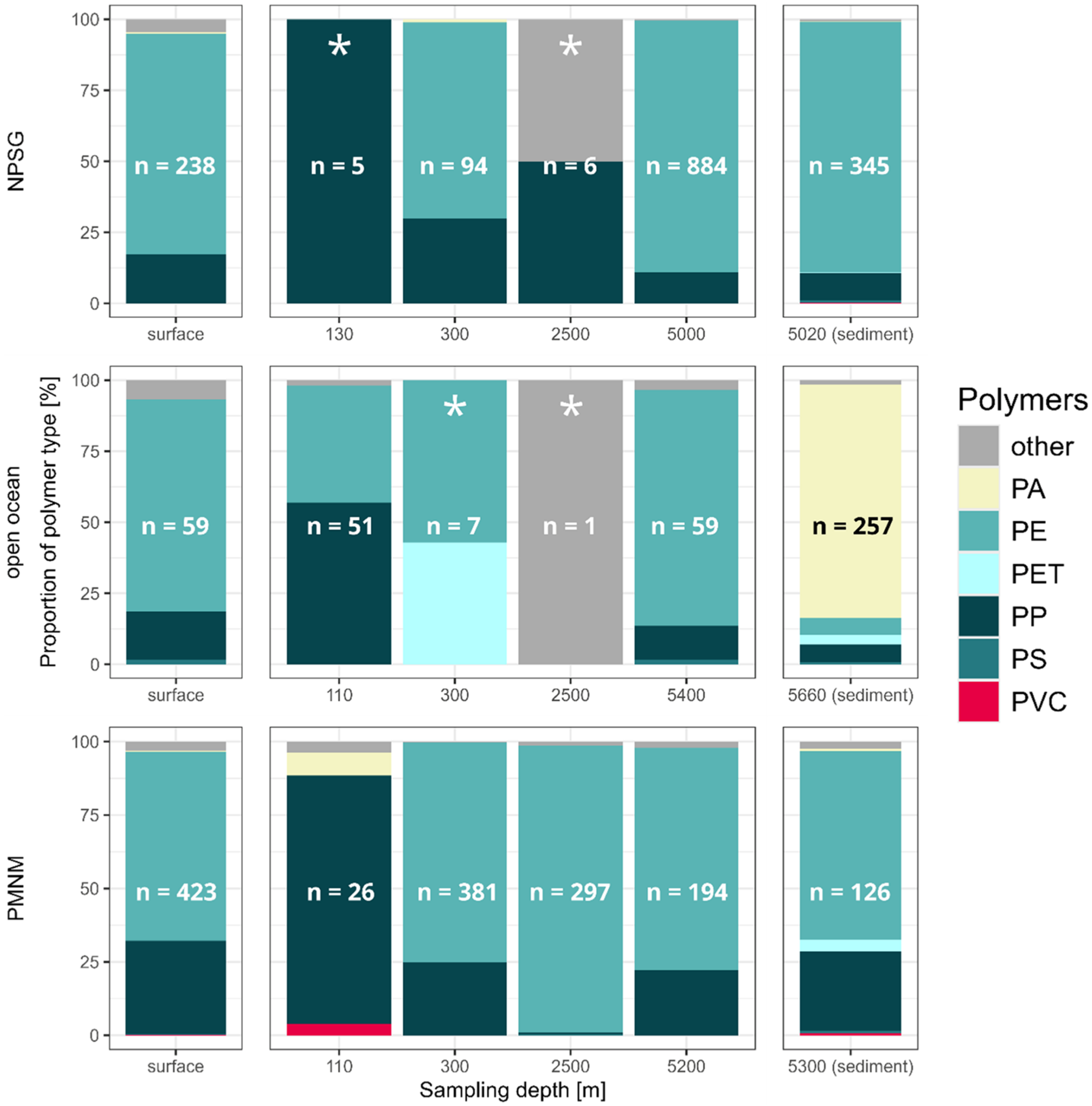

Figure 3. Polymer distribution of particles from the sea surface (>330 μm), water column (>11 μm), and deep-sea sediments (>11 μm) at the North Pacific Subtropical Gyre (NPSG), the open ocean station and in the area of the Papahānaumokuākea Marine National Monument (PMNM). Samples were collected during the RV SONNE expedition SO268/3 in 2019. Based on the small number of detected plastic items after blank correction, samples marked with asterisks (*) have to be viewed with caution. The colors represent the respective polymer (PA, polyamide; PE, polyethylene; PET, poly(ethylene terephthalate); PP, polypropylene; PS, polystyrene; PVC, poly(vinyl chloride)), and the number in the bars represent the total number of analyzed polymer particles. Polymers acrylonitrile butadiene styrene (ABS), ethylene-vinyl acetate (EVA), polyacrylonitrile (PAN), poly(butylene terephthalate) (PBT), polycarbonate (PC), polylactic acid (PLA), poly(methyl methacrylate) (PMMA), poly(tetrafluoroethylene) (PTFE), and polyurethane (PU) were clustered in the category "other" based on their scarcity of occurrence. More detailed information can be found in the SI (Figure S2 and Table S5).

NPSG area.[40] Furthermore, we observed up to three times higher concentrations of plastic items than previously detected in the Arctic Ocean.[12] This observation aligns with a commonly used distribution model for plastic items at the sea surface.[48]

Plastic Concentrations in Sediment Samples. A total of 728 plastic items were detected in four sediment samples (three stations and one in duplicate). Plastic item concentrations in the deep-sea sediment ranged from 1127 items kg^{-1} detected at the NPSG station to 3215 items kg^{-1} found at the open ocean station (Table 2). Duplicate samples from the

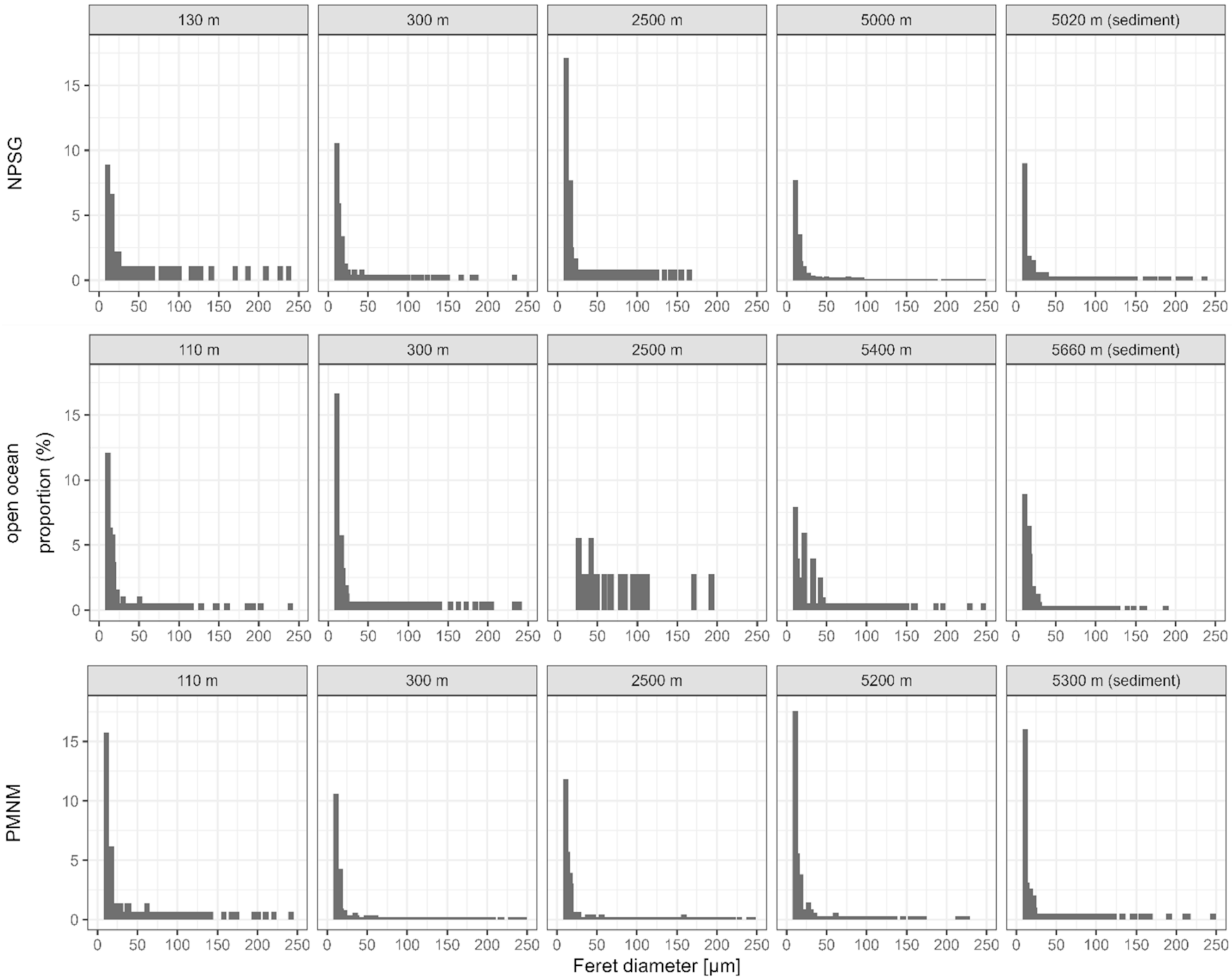

Figure 4. Particle size distributions of all particles <250 μm detected in the water column and sediment samples in the North Pacific Subtropical Gyre (NPSG), at the open ocean station and in the area of the Papahānaumokuākea Marine National Monument (PMNM). Samples were collected during the RV SONNE expedition SO268/3 in 2019. Particles larger than 250 μm only occurred occasionally and are not included here. The complete size distribution plots can be found in the SI (Figure S3).

NPSG station, collected from two of the MUCs cores, showed similar variability in concentrations (1127 and 3189 items kg^{-1}) over a small spatial scale of less than 2 m (Figure 2, right), which is consistent with a previous study conducted in the Great Australian Bight.[49] The high variation of concentrations at one station may be attributed to small-scale factors such as bioturbation and vorticity, while the large-scale distribution of plastic items may be influenced by ocean currents and regional hydrodynamic conditions, partly conserving surface water hotspots down to the sediment via fallout.[50] Due to the limited number of samples, the small sample size, and the high variability between cores from the same sampling location, no definitive statement can be made about the distribution patterns of plastic items in the deep-sea sediment. However, our data indicate that the concentrations of plastic items in deep-sea sediments may be subject to both small- and large-scale fluctuations.

Compared to previous studies, we found higher concentrations in the North Pacific Ocean than described for the West Pacific Ocean,[51] the Kuril-Kamchatka Trench,[50,52] and the Southern Ocean,[16] but approximately 300-fold lower concentrations than in the North West Pacific Ocean[19] were detected at a comparable depth, and comparable concentrations to the Arctic Ocean[12] (Table 2). Therefore, our results support the general assumption that plastic items are ubiquitous contaminants in deep-sea sediments. The comparably high amounts of plastic items in the deep-sea sediments may be a result of vertical transport of previously surface-floating plastic items. These plastic items reach the deeper regions of the ocean following incorporation into marine snow and related sinking of the aggregates, but other transport pathways like deep-sea turbidity currents may also influence the distribution of plastic items in the sediment.[43,53]

Particle Characteristics. A total of 2734 items were identified as plastics across the water column and sediment samples, which were dominated by PE (74.8%, $n = 2045$), PP (15.2%, $n = 415$) and PA (7.9%, $n = 216$). All water column samples predominantly contained low-density polymers such as PE and PP (Figures 3, S2 and Table S5).

The high share of PE and PP detected in the samples, which is consistent with previous findings, may generally be explained by their high combined market share of 40–50% and the wide application range of both polymers, especially in single-use products.[14,15,37,40,54] In alignment with a previous study conducted in the NPSG area, our data set with a lower size cutoff (11 μm vs 25 μm) is also dominated by small particles <100 μm (83.7%) and shows an increasing relative contribution of small microplastics in deeper waters. In contrast to the study by Zhao et al., buoyant polymers concentrated in the 10−75 μm size range (75.9%), while denser polymers contributed relatively more to particles ≥75 μm (47.3%).[14] Although the initial density of buoyant particles is lower than that of seawater, making them float, it may increase by weathering processes, facilitating transport into deeper regions of the ocean.[2,55] These processes include surface fouling and biofilm growth or the incorporation into natural particles, e.g., marine snow or fecal pellets, leading to elevated bulk density and increasing sinking velocity.[43,56] Furthermore, the similarities in polymer distribution from the ocean surface to deep-sea sediments support the assumption that these plastic items are fallout of surface-floating particles, consistent with a previous study conducted in the NPSG area.[40]

All samples, both from the water column and the deep-sea sediments, showed comparable size distributions with the majority of plastic items in the lower size range (<20 μm) detected by the used method and only small amounts of particles with a size of >250 μm (Figures 4 and S3).

The size distribution patterns of plastic items in the water column samples from the North Pacific Ocean largely reflect those reported in earlier studies. A study conducted in the Arctic Ocean showed a comparable size distribution with the majority in the range of 11−25 μm, and particles detected in the NPSG area also showed an increase in abundance with decreasing particle size.[12,14] In comparison with a study conducted in the Atlantic Ocean, we observed slightly smaller particles.[45] The observed trend toward even smaller particles is further confirmed in a recent study in the North Atlantic Ocean.[47] Additionally, the high share of small particles also explains the large differences (factor 200−280.000) in particle concentrations between this study and the study by Egger et al., focusing on larger plastic items >500 μm.[40]

The particle size distributions of the sediment samples were comparable to the data from the Kuril-Kamchatka trench with high shares especially of small particles.[50] These results are in accordance with a recent modeling study, showing that mainly particles <100 μm are able to reach the deep-sea sediments, based on the size-selective incorporation into marine snow aggregates.[43] The observed size distribution of the plastic items also matched the grain size distribution of the sediments, showing no sediment particles >20 μm, with median grain sizes between 3 and 5 μm (Table S2).

Implications for Future Research and the Need of Joint Mitigation Actions. The results of this study, including a uniform polymer distribution from the sea surface to the deep-sea sediments and comparable size distributions across all depths, align with the hypothesis that plastic items in the water column partly originate from "fallout" of surface-floating plastic debris.[40] This hypothesis suggests a relationship between surface-level plastic item concentrations and the concentrations in the water column and the deep-sea sediment. Additionally, this is, to the best of our knowledge, the first study in the open North Pacific Ocean to assess concentrations of plastic items throughout the water column and in the deep-sea sediment, combined with data of surface-floating plastic items, at the same sampling sites and times, focusing on small particles down to 11 μm. Comparable studies were only conducted by Tekman et al. in the Arctic Ocean, Quintana et al. in the Gulf of Cádiz, and Singh et al. in the San Pedro Bay (California), with the latter focusing on plastics >200 μm and >330 μm, respectively.[12,57,58] Consequently, our data may help refine modeling approaches and give additional information about the overall distribution of plastic items in the world's oceans, from surface layers down to deep-seawater layers and sediments.[43] In this, they add to our understanding of the fate of plastics in oceanic regions and extend our knowledge base to develop suitable and effective mitigation options. Our results also support the view that the ocean water column is a critical but understudied reservoir of microplastics, highlighting the need for expanded research efforts using harmonized methodologies.[37]

In addition, the observed size distribution patterns, with most particles being at the lower size cutoff of the used method (11 μm), indicates that established sampling and analysis approaches may not be sufficient to assess the overall plastic pollution in the marine environment, leaving a knowledge gap for still smaller micro- and nanosized plastic particles. The recent detection of plastics in the size range <1 μm in the water column of the North Atlantic Ocean further supports the necessity to include these small plastics in future research and policy making.[47] These smaller particles occurred in high abundance, increasing the likelihood of ingestion by a wide range of particularly small marine organisms and potentially leading to biomagnification within marine food webs. Additionally, they may pose an elevated risk due to their high surface-to-volume ratio, increasing the possibility and kinetics of leaching plastic-associated chemicals into the surrounding media. Consequently, they may be particularly relevant for the investigation of plastic impacts on marine life and the overall marine environment. By extending the observed size range to smaller particles, appropriate contamination controls, adaption of workflows to reduce contamination possibilities, and QA/QC considerations become increasingly important, as the small particle range is particularly susceptible to losses and introduction of contamination, e.g., from air.

Moreover, robust and reliable data on the distribution of plastic pollution in the environment are essential for identifying effective strategies to mitigate marine plastic pollution, particularly in the context of the United Nations' Plastics Treaty negotiations, which aim to establish a legally binding framework for addressing this global issue.[59,60] The ubiquity of plastics even in background marine areas and in particular in the deep sea exerts pressure to fast and efficiently limit emissions of such persistent contaminants which we currently cannot remove without disturbing the marine ecosystem.[55] Therefore, future policies should not only target emissions and the waste issue but also need to address the whole plastics life cycle, including the reduction of primary production and a switch to more benign polymers and less hazardous additives.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c11358.

Sample data and environmental parameters for all water column and sediment samplings; detailed polymer distribution data; and size distribution of plastic items from water column and sediment samples (PDF)

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Notes

The authors declare no competing financial interest.

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