Supplementary Material

Full Materials and Methods

Plastic numerical and mass concentration values included all available georeferenced data in the peer-reviewed literature as well as publicly available data from the Algalita Marine Research Foundation [Table S1]. Methods for the peer-reviewed data can be found in the publication in which they appeared. Because raw data from Algalita Marine Research Foundation were not made public, we used median values for each bin. When data were combined, we were able to compare changes in microplastic abundance between 1972-87 and 1999-2010.

Methods for new data, which includes samples from 1972-3 and 2009-10, are as follows. Samples from the early 1970s were collected on three cruises. Ten samples from 7205 (spring 1972) and 21 samples from 7210 (fall 1972) were collected using an ovoid 1-meter plankton net with 505 µm mesh towed for 20 minutes at the sea surface with a velocity of 0.7-1 m s^{-1} [S1]. A flowmeter was not used, but the deployers estimated the water flow through the net at 250-300 m^3. We used a value of 275 m^3 to calculate density from these tows. Fourteen samples from Southtow 13 (winter 1973) were collected using a rectangular neuston net (1.0 x 0.5 meters) with 505 µm mesh. These nets were towed at 2 m s^{-1} for varying amounts of time ranging from 20 minutes to 4 hours. Water flow through the net was calculated using the starting and ending coordinates. All samples were preserved in 1.8% formaldehyde buffered with sodium borate and archived in the Scripps Institution of Oceanography Pelagic Invertebrates Collection.

The 2009 samples (N=119 samples, 45 of which were also sampled for *Halobates*) were collected on the SEAPLEX cruise on the *R/V New Horizon* in August 2009. The 2010 samples (N=28) were collected on the EX1006 cruise on the R/V *Okeanos Explorer* in October 2010. Both sets of samples were collected using a standard manta net (0.86 x 0.2 m) with 333 µm mesh [S2], towed for 15 minutes at 0.7-1 m s^{-1}. Water through the net was measured with a General Oceanics analog flowmeter. The SEAPLEX samples were preserved in 1.8% formaldehyde buffered with sodium borate, and the EX1006 samples in 95% ethanol.

Each sample was sorted at 6-12x magnification under a Wild M-5 dissecting microscope, and plastic particles and *H. sericeus* removed for further analysis. Plastic particles were soaked in deionized water to remove salts, dried at 60°C, and stored in a desiccator. Dry mass was measured on an analytical balance. Particles were then digitally imaged with a Zooscan digital scanner [S3]. The total number of particles as well as two-dimensional surface area and maximum diameter of each particle were measured using NIH ImageJ-based tools in the Zooprocess software, calibrated against manual measurements [10, S3].

The diameter of plastic particles as measured by Zooscan was used to determine that there was no significant effect of the change from 505 µm plankton net mesh to 333 µm mesh. There was no significant difference in particle size spectra from fall 1972 to fall 2010 (Kolmogorov-Smirnov test, p=0.2228), and 98.5% of the particles caught in the 333 µm net would also have been caught in a 505 µm net.

H. sericeus samples were enumerated and classified into 5 categories: juvenile, adult male, adult female, newly molted, and molted exoskeletons. *H. sericeus* eggs, both those attached to plastic and those that had become detached during the collection process, were also enumerated. To determine the size range of particles utilized by *H. sericeus* for oviposition, the two-dimensional surface area and maximum diameter of a subset of plastic particles with attached eggs (N=207) were measured using NIH ImageJ, and compared to the size of all plastic particles collected in 2009-10 (N=32,090). Only plastic particles in the size range known to be utilized by *H. sericeus* were used to calculate plastic concentrations in Fig. 2 (N=26,045).

Dry mass of zooplankton was obtained from preserved manta tow samples (N=46, [S4]). After fixation in 1.8% formaldehyde for 24 months, samples were split in a Folsom splitter, filtered onto 202 µm Nitex mesh pads and rinsed with isotonic ammonium formate. Filters were dried for 24 hours at 60°C and placed in a dessicator until weighing. Filters were weighed to the nearest 0.0001 gram on the same analytical balance as the plastic samples. A 20% correction factor was applied in order to compensate for the biomass lost by preservation [S4, S5]. A similar method was used for dry mass of *H. sericeus* eggs (N=52), except that they were dried in pre-weighed aluminum crucibles and weighed on an electrobalance to the nearest microgram.

Maximum length of *H. sericeus* eggs ranges from 0.8-1.2 mm, and a single microplastic particle commonly has between 1 to 3 eggs attached. To compare the relative biomass of *H. sericeus* eggs and zooplankton, we divided the median egg dry mass (0.059 mg egg^{-1}) by 1 m^2 of the median daytime zooplankton dry mass (0.642 mg m^{-2}). Using this estimation, one egg was equivalent to 9.2% of the daytime zooplankton biomass in the top 20 cm of 1 m^2 of surface water, and three eggs (0.059 mg egg^{-1} x 3) equivalent to 27.6% of the median daytime zooplankton biomass. Daytime values were used because the conspicuous size and color of *H. sericeus* eggs make them likely prey for diurnal visual predators such as omnivorous fishes or seabirds.

We computed all statistics using the R statistical environment (version R-2.13.1) [S6]. Because the data did not meet parametric assumptions of normality, we used nonparametric methods with alpha values of 0.05. We used the Mann-Whitney test for abundance comparisons and the Spearman rank correlation to examine the relationship between *H. sericeus* and microplastic. R^2 values were calculated from Spearman's rho. Lines were fit to correlations using the Theil-Sen single median method as implemented in the R MBLM package [S7]. Maps were created in Surfer 8 (Golden Software) [S8] and interpolated using point kriging. Figures were created in R and formatted in Adobe Illustrator CS5.1. New data from this study are deposited with the California Current Ecosystem LTER DataZoo (http://oceaninformatics.ucsd.edu/datazoo/data/ccelter/datasets?action=group&id=1).

Control for regional and seasonal variation in microplastic abundance

In order to avoid effects of regional and seasonal variations in oceanic conditions on data analyses, we chose an area in the North Pacific Subtropical Gyre (NPSG) between Hawaii and California where comparable samples were available for the present study. A subset of 41 samples, 21 collected in October 1972, and 20 in October 2010 were analyzed. We found an increase of two orders of magnitude in microplastic, with median numerical concentration (NC) increasing from 0.003 to 0.270 particles m^{-3} (two-tailed Mann-Whitney test, $p<0.0001$) and mass concentration (MC) increasing from 0.004 to 1.158 mg m^{-3} (two-tailed Mann-Whitney test, $p<0.0001$).

Supplementary Tables

Table S1. Data sources for microplastic numerical and mass concentrations. The season/year, region, sample size, and data type are given for each data source. Samples used for the *H. sericeus* analyses are designated with an “x.” Regions are the North Pacific Subtropical Gyre (NPSG), Alaska (AK), the California Current (CA), and the Eastern Tropical Pacific (ETP). Data types are numerical concentration (C), and mass concentration (M). Data sources are given as cruise names for new data or citations for published or public data.

season/year	region	N	data type	source	*H. sericeus* analysis
Spring 1972	NPSG	10	C, M	7205 (new data)	x
Fall 1972	NPSG	21	C, M	7210 (new data)	x
Fall 1972	NPSG	31	M	Wong et al. 1974 [7]	
Winter 1973	NPSG	14	C, M	Southtow 13 (new data)	x
October 1974 & 1975	AK	71	C	Shaw 1977 [8]	
June-August 1976 & 1985	AK, NPSG	31	M	Day & Shaw 1987 [9]	
Winter 1984	CA	61	C, M	Gilfillan et al. 2009 [10]	
Fall 1987	ETP	23	C, M	STAR 8710 (new data)	
Season varied, years 1999, 2000, 2002, 2005-2008	CA, NPSG	178	C, M	Algalita Marine Research Foundation [19]	
Fall 1999	ETP	5	C, M	STAR 9910 (new data)	
Fall 2000	ETP	6	C, M	STAR 0010 (new data)	
Throughout 2006	AK, CA	234	C, M	Doyle et al. 2011 [11]	
Fall 2006	ETP	9	C, M	STAR 0610 (new data)	
Winter 2007	CA	66	C, M	Gilfillan et al. 2009 [10]	
Summer 2009	CA, NPSG	119	C, M	SEAPLEX (new data)	x
Fall 2010	CA, NPSG	28	C, M	EX1006 (new data)	x

Table S2. Microplastic particle and mass concentrations in 1972-87 and 1999-2010. Median, 5th to 95th percentiles of the data, and maximum values are given for each region. Regions are the North Pacific Subtropical Gyre (NPSG), Alaska (AK), the California Current (CA), and the Eastern Tropical Pacific (ETP).

particle concentration 1972-1987					
	median (no. m^{-3})	**5th to 95th percentile (no. m^{-3})**	**maximum (no. m^{-3})**	**N**	**data source**
all data	0.000	0.000-0.117	0.822	200	-
NPSG	0.003	0.000-0.071	0.221	45	this study
AK	0.000	0.000-0.010	0.010	71	[8]
CA	0.012	0.000-0.471	0.822	61	[10]
ETP	0.000	0.000-0.043	0.087	23	this study
particle concentration 1999-2010					
all data	0.116	0.000-4.696	32.760	645	-
NPSG	0.425	0.092-8.649	32.760	301	[19], this study
AK	0.000	0.000-0.140	0.406	22	[11]
CA	0.010	0.000-0.228	3.141	302	[10,11,19], this study
ETP	0.012	0.000-0.034	0.044	20	this study
mass concentration 1972-1987					
	median (mg m^{-3})	**5th to 95th percentile (mg m^{-3})**	**maximum (mg m^{-3})**	**N**	**data source**
all data	0.000	0.000-0.675	5.337	191	-
NPSG	0.003	0.000-0.705	3.500	87	[7,9], this study
AK	0.000	0.000-0.078	0.600	20	[9]
CA	0.003	0.000-1.001	5.337	61	[10]
ETP	0.000	0.000-0.089	0.401	23	this study
mass concentration 1999-2010					
all data	0.086	0.000-25.000	250.000	645	-
NPSG	3.000	0.000-25.000	250.000	301	[19], this study
AK	0.000	0.000-0.268	0.406	22	[11]
CA	0.001	0.000-0.713	3.000	302	[10,11,19], this study
ETP	0.003	0.000-0.385	6.581	20	this study

Supplementary References

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