A deposition baseline for microplastic particle distribution in an estuary

Matthew LH. Cheng
Department of Biological Sciences, University of New Hampshire, 38 Academic Way, Durham, NH 03824 USA
lhc1005@wildcats.unh.edu
@MLCheng3

Thomas C. Lippmann
University of New Hampshire, Department of Earth Sciences, Jere Chase Ocean Engineering Lab, 24 Colovos Road, Durham, NH 03284 USA
T.Lippmann@unh.edu
@LippmannThomas

Jennifer Dijkstra
Center for Coastal and Ocean Mapping, Chase Ocean Engineering Lab, 24 Colovos Road, Durham, NH 03824 USA
Jennifer.Dijkstra@unh.edu
@DijkstraLab

Gabriela Bradt
University of New Hampshire Cooperative Extension, 59 College Road, Durham, NH 03824 USA
Gabriela.Bradt@unh.edu
@DrSquidgNH

Salme Cook
University of New Hampshire, Department of Earth Sciences, Jere Chase Ocean Engineering Lab, 24 Colovos Road, Durham, NH 03284 USA
salme.cook@gmail.com

Jang-Geun Choi
University of New Hampshire, Department of Earth Sciences, Jere Chase Ocean Engineering Lab, 24 Colovos Road, Durham, NH 03284 USA
jc1404@wildcats.unh.edu

Bonnie L. Brown
University of New Hampshire, Department of Biological Sciences, 38 Academic Way, Durham, NH 03824 USA
Bonnie.Brown@unh.edu
@B3EcoGenetics

Corresponding author: Bonnie.Brown@unh.edu, University of New Hampshire, Dept. of Biological Sciences, 38 Academic Way, Durham, NH 03824 USA

Abstract

Microplastics (MP) known to be ubiquitous in the plastisphere, have yet to be quantified within Great Bay Estuary (GBE) in the Gulf of Maine region of North America. We extracted and analyzed MP in archived sediment cores obtained from representative transects across GBE. Results indicated that MP are distributed in GBE sediments, 0-30 cm, at an average occurrence of 100±50 particles g^{-1} and that morphology varies by site and depth. Analysis by sediment depth and age class indicated that MP deposition increased over several decades but recently (5-10 years) has likely begun to decrease. Hydrodynamic and particle transport modeling indicated that bed characteristics are a more controlling factor in MP distribution than MP properties and that the highest deposition likely occurs in regions with weaker hydrodynamic flows and lower bed shear stress e.g., eelgrass meadows and along the fringes of the Bay. These results provide a baseline and predictive understanding of the occurrence, morphology, and sedimentation of MP in the estuary.

Keywords: microplastics, sedimentation, Great Bay Estuary

1. Introduction

Plastics, in particular microplastics (MP: predominantly degraded plastic particles that are <5 mm), are the fastest growing pollutant in US coastal waters (Law 2017). It is generally agreed that ~90% of MP that enter the water column eventually sink to the bottom (Kaiser et al. 2017) and that benthic sediments are a long-term sink for MP (vanCauwenberghe et al. 2015). Few studies have examined the natural dispersion and deposition of MPs in estuaries, nor do we have information on the disposition of these particles over space and time. Temporal concentrations of MPs in estuaries likely have changed as different types of plastics are introduced by society. Further, water treatment facilities have been upgraded and newly constructed facilities may capture and filter plastics better. Spatial concentrations of MPs likely are affected by the presence of oyster reefs and eelgrass beds where structure and filter-feeding can trap or slow the transport of particles. Establishing temporal and spatial baselines of ambient MP levels and predicted deposition hotspots are necessary to determine whether MP concentrations have undergone temporal changes and to craft effective seafood cultivation strategies.

Our effort was to generate baseline data for the historical complement of MP in a hydrodynamically active estuary and to enable broad-level modeling of particle distributions that will inform where there are potential locations of high MP concentrations. To accomplish this, we recovered MP particles from preserved cores collected in 2016 at strategic locations in GBE, including sites that correspond to an ongoing EPA-NCCA program led by other investigators. Suspended MP particles were seeded in a numerical particle model (Choi et al. 2018) with velocity fields and bed shear stresses obtained from a verified hydrodynamic model of the GBE system (Cook et al. 2019; Cook et al. 2021) for determination of dispersion and MP deposition hotspots.

2. Methods

2.1 *Study area and relevance*

Great Bay Estuary (43°04'0.60"N, 70°52'4.19"W) is a well-mixed body of water located in coastal New Hampshire. The strong currents of Piscataqua River along the Maine-New Hampshire border introduce oceanic water into the Bay and mix with numerous (typically) low-discharge rivers that provide freshwater sources (Short 1992). Pollution has become an issue forcing change, particularly from the permitted waste-water treatment facilities located in 42 communities in New Hampshire and 10 communities in Maine (NHDES, 2016). These facilities discharge directly into the tributaries of GBE and from that discharge micro- and nano-plastics (particles ≤100 μm) likely are introduced. There are no prior data regarding the concentrations of MP in GBE waters or sediments and because the flow of water within GBE is complex and river input varies both spatially and temporally (Short 1992), the extent to which rivers are the primary source of MPs is unclear. We also have no knowledge of where MPs concentrate once they are introduced. If particles concentrate at certain times of the year or in some areas but not others (e.g., wetlands, mud flats, oyster reefs, eelgrass beds, deep channels), then better knowledge of the dispersal and settling of MP would provide harvesters, aquaculturists, and resource managers with the necessary information to evaluate contaminant risks, choose low MP sites for their activities, and provide information on the time of year with the highest or lowest MP concentrations.

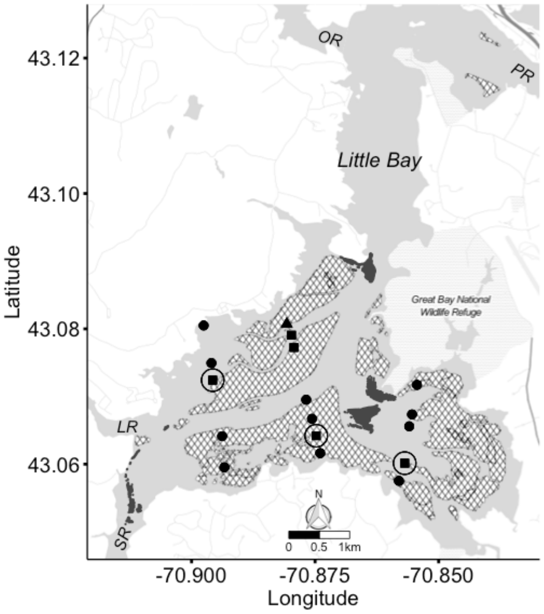

Figure 1. Map of Great Bay Estuary in New Hampshire showing the location of previously archived sediment cores obtained in 2015-2016 that were analyzed in this study using Nile Red MP detection (dots), sediments reanalyzed for micro-FTIR MP detection (triangles), cores submitted for ^{210}Pb dating (encircled squares). Hatched areas indicate eel grass meadow. Dark gray areas show oyster reef. LR: Lamprey River mouth, OR: Oyster River, PR: Piscataqua River, and SR: Squamscott River are shown for reference.

2.2 *Sediment sampling and dating*

We examined a total of 25 subsections across 17 cores that had previously been divided into 2-cm subsections (Fig 1) for MP concentration. Morphology and sediment samples from the three deepest cores (0-30 cm) were subjected to radiometric depositional age determination. All core samples were collected in 2015-2016 (similar to Lucking et al. 2017), already had been divided into 2 cm sections, analyzed for grain size and for wet and dry bulk density to 20-30 cm depth, and subsequently were stored dry. Radiometric dating was accomplished at Flett Research, Ltd. with ^{210}Pb and ^{226}Ra, a method commonly used to obtain age by depth of estuarine and coastal sediments (Eakins and Morrison 1978; Appleby and Oldfield 1978; Mathiew et al. 1988). Selected upper and lower sections also were subsampled for MP by two rounds of density separation using a combination of $ZnCl_2$ settling (Hitchcock and Mitrovic 2019; Coppock et al. 2017; Lusher et al. 2017), filtration onto PTFE membrane, KOH-NaOCl digestion to remove biogenic matter (Enders et al. 2016), followed by Nile Red staining and MP visualization using confocal microscopy (Nikon A1R HD) (Erni-Cassola et al. 2017) to initially quantify putative MP.

2.3 *MP morphology*

Elemental composition of MPs in selected sediment sections was assessed by washing particles from the original PTFE filters, transfer to stainless steel filters, then analyzed by Fourier Transform Infrared microspectrometry (micro-FTIR) on ThermoScientific Nicolet iN10MX and Shimadzu AIM9000 microscopes. Several sediment samples that were analyzed using Nile Red also were replicate extracted from the same original sediments, and following biogenic digestion, MP were transferred to AlOx filters (Whatman Anodisc) and analyzed using Bruker Lumos II micro-FTIR. Finally, MP of four samples previously analyzed by micro-FTIR were transferred from the AlOx filters into a 70% EtOH slurry, deposited onto Kevley slides (Kevley Technologies, MIRRIR Low-e glass slides, NC0733469), and allowed to dry prior to analysis using Agilent 8700 LDIR. Statistical analyses, performed in R (v 3.6.3, R Core Team 2020), included tests for normality, equal variance, and Wilcoxon and Kruskal-Wallis tests for mean differences. Quantitative data including MP quantity, density, type g^{-1} dry weight of sediment, were used in subsequent models and statistical determinations.

2.4 *Simulations*

Microplastic type and density g^{-1} dry weight of sediment were used in numerical modeling experiments to estimate spatial distributions of MP concentrations in surficial seafloor sediments, and to assess the sensitivity of particle settling and bed erosion parameters to the spatial distribution of deposited particles throughout the GBE. Two sets of simulations were conducted using velocity fields and bed shear stresses spanning the GBE estimated by Cook et al. (2019) using the Regional Ocean Modeling System (ROMS; Shchepetkin and McWilliams 2005; Haidvodel et al., 2008) coupled with an offline transport model (Choi et al. 2018) that predicts the spatial and temporal concentration of particles in the water column and settling of MP on the seafloor. Initial 5-day model simulations were initiated with a pile of unconsolidated MP located on the seafloor within the center of GBE. MP properties included particle density and fall velocity allowing sediments to be differentially suspended (uniformly) into the water column when flow velocities produce bed shear stresses that exceed a defined threshold (estimated from laboratory Eromes chamber experiments of field cores obtained from GBE, Wengrove et al. 2015). Suspended particles were dispersed horizontally by advection processes. This enabled particles to settle out based on their properties and where bed shear stresses are weak, and then potentially re-suspended again on the next ebb or flood tidal cycle. Subsequent simulations evaluated how MP might be deposited in GBE both with and without the presence of aquatic vegetation (eel grass meadows) that change the properties of the bed shear stress (as in Cook et al. 2021). Observations of eelgrass meadows used for this study were taken from Short (2017) and simulations of particle flow were parameterized by a bed erodibility function. Simulations also were conducted that assessed the sensitivity of deposited MP distributions to particle settling and resuspension parameters that grossly represent basic physical processes of settling and resuspension.

3. Results

3.1 *MP occurrence and distribution in GBE*

Sediment age determined by radiometric dating was found to increase in a linear fashion with depth. Deposition rate was quite similar between two of the three cores aged; for those two cores, the top 6 cm of sediment indicated deposition occurred over the past 10 years; the other dated core contained sediments that were aged >50 years at that depth. Numbers of particles detected by all three methods (staining, micro-FTIR, and LDIS) were within an order of magnitude of one another and MP (and fragments of other materials, most notably rubber but also chitin and plant fibers that escaped biodegradation) were found in most sediment subsamples aged ≤ 50 years. Across the 25 sediment sections analyzed, the range of MP detected was 0–1151 MP g^{-1} sediment dry weight, with a median occurrence of 10 MP g^{-1}, and a mean of 100 ± 50 MP g^{-1}. There was no significant difference in the number of particles detected in these upper segments (p=0.98) nor was there a significant difference in the number of particles detected across transects (p=0.30). Microplastics were more abundant in intermediate-aged sediments (170±65 MP) than they were in older (118±8) and newly deposited (42±25) sediment sections, albeit not to a significant degree (p=0.34). Morphology of MP was variable and differed significantly across cores in terms of length (p<0.001) and plastic type (p<0.001). Some sediment sections contained complex mixtures of copolymers, low and high density (polypropylene and polyvinylchloride, respectively), and biogenic (e.g., rubber, chitin, and plant fiber) particles (Fig 2), whereas other core segments had relatively homogenous MP populations, predominantly polyvinylchloride or polypropylene.

3.2 *Model predictions*

The numerical simulations indicated that unconsolidated MP initially on the seafloor within the center of GBE were suspended rapidly by the tidal flows and advected and deposited over just a few tidal cycles in regions of low bed shear stress near the fringes of the GBE, especially in the absence of aquatic vegetation. In the presence of eel grass, particles also were efficiently trapped by the reduced erodibility and shear stresses within the meadows, indicating that eelgrass meadows act as effective sinks for suspended MP (as expected). Much higher concentrations of particles remained in suspension during model runs without vegetation compared with model runs that included eel grass meadows, indicating higher numbers of MP that disperse widely and may leave GBE in the absence of aquatic vegetation. Model simulations including eelgrass meadows showed that erosion rate (easily eroded versus erosion-resistant conditions) was the most important determinant of MP spatial distribution in GBE. Simulations (Fig. 3) showed that resuspended particles were repeatedly suspended, advected, deposited, and resuspended by the tidally varying flow until the MP permanently settled out in regions of low bed shear stress (i.e., weak erodibility conditions with low erosion rate) associated with the eelgrass meadows or fringes of the Bay. Far fewer MP remained in suspension during vegetated model runs compared with the non-vegetated model indicating a much higher number of particles ultimately remain within the GBE under realistic conditions. Under conditions of weak settling velocity (i.e., more buoyant particles), fewer MP will settle to the bottom, a situation that increases the likelihood that more MP in the water column will be flushed from GBE. Interestingly, model results (Fig. 3) indicated a higher dependence on bed-erodibility, k_{rs}, than on settling rate, k_{st}. The latter term is determined by MP density and size and is complicated by poorly understood flocculation processes that increase sinking rates, whereas k_{rs} is strongly determined by characteristics of the seafloor that may include the presence or absence of aquatic vegetation. Collectively, these results indicate that bed characteristics are a more controlling factor in MP distribution than the particle properties themselves.

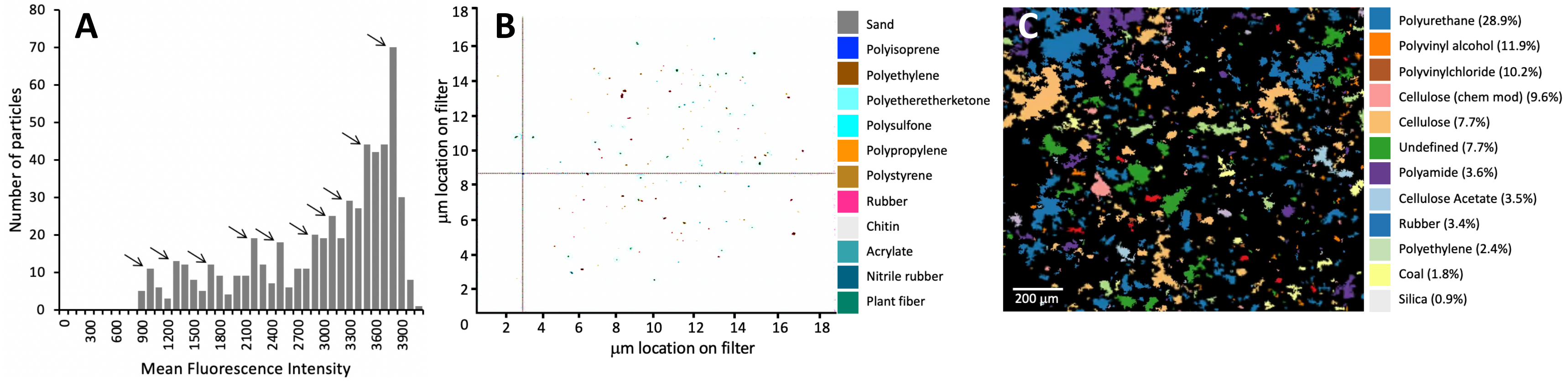

Figure 2. Three renditions of occurrence and morphology of microplastic particles found in representative sediment cores from Great Bay Estuary. A: Multiple fluorescence peaks (arrows) of Nile Red stained, confocal imaged MP from a sediment core section signify relative abundance of putatively different types of plastic particles. B: Particle map generated by micro-FTIR for a small section (300 µm^2) of an AlOx filter shows multiple (false colored) plastic particles isolated from a sediment sample but also biogenic substances that escaped digestion. C: Partial view of LDIR analysis particle map from a sediment sample shows multiple plastics and also biogenic substances (false color key sorted by frequency out of 3,283 particles).

Note to Editors: this figure will require color printing

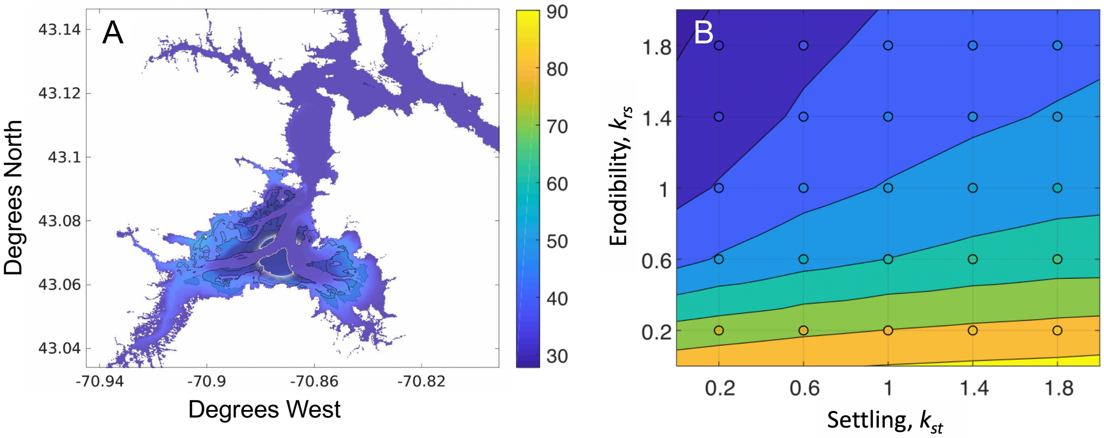

Figure 3. Spatial distribution of MP particles after 5-day model simulations. A: Single simulation showing that MP are trapped in eelgrass meadows or transported to the sides of GBE where they settle in areas along the fringes and shallow areas of the Bay (areas with low hydrodynamic velocities, low erodibility, and weak bed shear stress). B: Ensemble of all simulations showing contours of % MP deposited on the seafloor as a function of bed-erodibility, k_{rs}, and settling rates, k_{st}. Results show that bed-erodibility is a stronger determinant of MP concentration in the sediments than settling properties. Color scale in center is % of MP deposited on the seafloor and applies to both panels.

4. Discussion

There is a pressing need to determine the sources, distribution, and fate of MP particles so that interactions of MP with aquatic organisms that are harvested for human consumption can be understood. Deepening our knowledge of MP in estuarine systems is a necessary component of addressing the knowledge gap of the realistic levels of risk these particles pose to living resource populations (Lenz et al. 2016). To our knowledge, this is the first study to generate temporal and spatial patterns of MPs using preserved cores, thus providing a baseline for the historical complement of MP in an estuarine system. Concentrations of MPs have changed over time, with greatest MP concentrations at intermediate aged sediments. Variations in spatial MP concentrations are likely affected by the presence of aquatic vegetation (eelgrass meadows) where plant structure will trap or slow the transport of particles within estuaries with eelgrass, reducing erodibility and bed shear stress and promoting settling in those areas. Our study established a rough overview of ambient MP levels, types, and deposition. The coupled hydrodynamic and particle tracking model provided a mechanism for predicting MP spatial distribution patterns, understanding the behavior of MP settling, and tracing the evolution of MP concentrations in sediments. Future studies coupling MP concentration and identity found in both water and sediments will facilitate creation of illustrative models that more adequately describe the present potential for introduction, movement, and distribution of MP in estuaries. Verified model visualizations will promote data usage by harvesters, farmers, and resource managers enabling new strategies to cope with the presence of MPs, modify harvesting procedures, and hot-spot avoidance for future restoration activities.

Acknowledgements

We gratefully acknowledge Mark Townley of UNH, who assisted with optimizing confocal microsopy for Nile Red stained specimens. As demonstration of MP morphology analysis from sediments, Joe Dorsheimer (ThermoScientific), Fred Morris and Marten Seeba (Bruker), and Louis Tisinger and Keegan McHose (Agilent) provided gratis all of the infrared spectrophotometric analyses for this project. We thank Jon Hunt for assisting with the collection of sediment cores and construction of SMI devices, Kristen Mello for dive support and core collection, and Griffin Sperry for core physical analysis and preservation. We also thank volunteers Eleanor Zwart and Lily Pettit for assisting with density separation and trapping of MP from archived desiccated core sections. This work was supported by funding from the University of New Hampshire College of Life Sciences and Agriculture (2018-2022); NH Sea Grant Development Funding; and cash contributions from 3StateHomes.

References:

Arthur C, J Baker, H Bamford. 2009. Proceedings of the International Research Workshop on the Occurrence, Effects, and Fate of Microplastic Marine Debris. Group:530.

Benninger LK. 1978. Pb-210 balance in Long Island Sound. *Geochimica and Cosmochimica Acta* 42:1165-1174.

Choi, JG, YH Jo, IJ Moon, J Park, DW Kim, and TC Lippmann. 2018. Physical forces determine the annual bloom intensity of the giant jellyfish *Nemopilema nomurai* off the coast of Korea, *Reg. Studies in Mar. Sci.,* 24, 55-65.

Cook SE, TC Lippmann, and JD Irish. 2019. Modeling nonlinear tidal evolution in an energetic estuary. *Ocean Modelling* 136:13-27.

Cook, SE, TC Lippmann, K Koetje, M Wengrove, and D Foster. 2021. The influence of submerged aquatic vegetation on modeled estimates of bed shear stress and nutrient resuspension in a tidally dominant estuary, *Estuarine, Coastal, and Shelf Science*, *sub judice*.

Estahbanati S, and NL Fahrenfeld. 2016. Influence of wastewater treatment plant discharges on microplastic concentration in surface water. *Chemosphere* 162:277-284.

Farmer JG. 1978. The determination of sedimentation rates in Lake Ontario using the Pb-210 dating method. *Canadian Journal of Earth Sciences* 15:431-437.

Haidvogel DB, H Arango, WP Budgell, BD Cornuelle, E Curchitser, E Di Lorenzo, K Fennel, WR Geyer, AJ Hermann, L Lanerolle, J Levin, JC McWilliams, AJ Miller, AM Moore, TM Powell, AFShchepetkin, CR Sherwood, RP Signell, JC Warner, J Wilkin. 2008. Ocean forecasting in terrain-following coordinates: formulation and skill assessment of the Regional Ocean Modeling System, *Journal of Computational Physics* 227:3595–3624.

Kaiser D, N Kowalski, JJ Waniek. 2017. Effects of biofouling on the sinking behavior of microplastics. *Environmental Research Letters* 12:124003.

Law KL. 2017. Plastics in the marine environment. *Annual Review of Marine Science* 9:205-229.

Lenz R, K Enders, TG Nielsen. 2016. Microplastic exposure studies should be environmentally realistic. *Proceedings of the National Academy of Sciences* 113:E4121-E4122.

Mason SA, D Garneau, R Sutton, Y Chu, K Ehmann, J Barnes, P Fink, D Papazissimos, D Rogers. 2016. Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. *Environmental Pollution* 218:1045-1054.

Murphy F, C Ewan, F Carbonnier, B Quinn. 2016. Wastewater treatment works (WwTW) as a source of microplastics in the aquatic environment. *Environmental Science & Technology* 50:5800-5808.

Rocha-Santos T, AC Duarte. 2015. A critical overview of the analytical approaches to the occurrence, the fate and the behavior of microplastics in the environment. *Trends in Analytical Chemistry* 65:47-53.

Shchepetkin AF, JC McWilliams. 2005. The Regional Oceanic Modeling System (ROMS): A split-explicit, free-surface, topography-following-coordinate oceanic model, *Ocean Modeling* 9 ;347–404.

Short FT. 1992. The ecology of the Great Bay Estuary, New Hampshire and Maine: An estuarine profile and bibliography. Page 222. Coastal Ocean Program Publ.

Short, FT. 2017. SeagrassNet Monitoring in Great Bay, New Hampshire, 2016. PREP Reports & Publications. 392. https://scholars.unh.edu/prep/392.

Thompson RC, Y Olsen, RP Mitchell, A Davis, SJ Rowland, AWG John, D McGonigle, AE Russell. 2004. Lost at sea: Where is all the plastic? *Science* 308:838.

vanCauwenberghe L, L Devriese, F Galgani, J Robbens, CR Janssen. 2015. Microplastics in sediments: a review of techniques, occurrence, and effects. *Marine Environment Research*. 111:5-17.

Vianello A, A Boldrin, P Guerriero, V Moschino, R Rella, A Sturaru, L Da Ros. 2013. Microplastic particles in sediments of Lagoon of Venice, Italy: First observations on occurrence, spatial patterns and identification. *Estuarine, Coastal, and Shelf Science* 130:54-61.

Wengrove, ME, DL Foster, LH Kalnejais, V Purcuoco, and TC Lippmann. 2015. Field and laboratory observations of bed stress and associated nutrient release in a tidal estuary, *Estuarine, Coastal, and Shelf Science*, <u>161</u>, 11-24.