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High levels of pelagic plastic pollution within the surface waters of Lakes Erie and Ontario

Sherri A. Mason [a,*], Juliette Daily [b], Ghadah Aleid [c], Rachel Ricotta [c], Morgan Smith [c], Katie Donnelly [c], Rebecca Knauff [b], William Edwards [d], Matthew J. Hoffman [b]

[a] Pennsylvania State University, The Behrend College, 4701 College Dr., Erie, PA 16563, USA
[b] Rochester Institute of Technology, School of Mathematical Sciences, 85 Lomb Memorial Dr., Rochester, NY 14623, USA
[c] State University of New York at Fredonia, Department of Chemistry and Biochemistry, 280 Central Ave., Fredonia, NY 14063, USA
[d] Niagara University, Department of Biology, Lewiston, NY 14109, USA

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ABSTRACT

During 2012 to 2014 five expeditions collected surface water samples for plastic pollution analysis representing the first data within Lake Ontario and the first multi-year dataset for Lake Erie. Lake Ontario had the highest abundances of any Great Lake to date with an average of over 230,000 particles/km^2. Though having a considerable smaller average of ~45,000 particles/km^2, Lake Erie remains second only to Lake Ontario based on studies to date and averaged across all samples and years. The high levels of pelagic plastic pollution is likely owing to their position as the last two lakes in the Laurentian Great Lakes ecosystem, as well as the prominence of population centers along their shorelines. As with previous studies, most particles were found within the smallest size classification (0.355–0.999 mm; 73%), with fragments (63%) and pellets (26%) forming the dominant morphologies. The minor contribution of fibers/lines (4%) is consistent with previous Great Lakes studies, though not with studies within other environmental compartments (e.g., sediment, fish, atmospheric). This could be due to the negative buoyancy of polymeric fibrous materials, a hypothesis consistent with the dominance of the less dense polymers polyethylene (46%) and polypropylene (43%) (based on FTIR analysis). For the first time, the multiyear Lake Erie samples were compared to modeled plastic distributions and found to fit reasonably well. Using the sample data to calibrate the model we estimate that there are 475 million plastic particles, with a total mass of 6.45 metric tons, floating on the surface of Lake Erie alone.

Introduction

While interest in the fate and impacts of plastic pollution started in the ocean (Andrady, 2011; Law, 2017; Law et al., 2010), awareness of plastic pollution in freshwater systems has increased in recent years (Ballent et al., 2016; Cable et al., 2017; Eriksen et al., 2013; Free et al., 2014; Hoffman and Hittinger, 2017; Mani et al., 2015; Mason et al., 2016). This is particularly true in the North American Great Lakes, which make up one of the world's largest freshwater ecosystems. Surface samples from the Great Lakes have found concentrations of microplastic particles that are comparable to those found in the ocean (Cable et al., 2017; Eriksen et al., 2013; Mason et al., 2016), and it has been estimated that over four metric tons of microplastic are floating in Lake Erie (Hoffman and Hittinger, 2017) alone. Around 10,000 metric tons of plastic is estimated to enter the Great Lakes every year (Hoffman and Hittinger, 2017) and has been found in surface waters, in the sediment (Ballent et al., 2016; Corcoran et al., 2015), on beaches (Driedger et al., 2015; Zbyszewski et al., 2014), and in wildlife (McNeish et al., 2018). Despite the knowledge that plastic particles permeate aquatic systems, the fate of most of the plastic that enters the Great Lakes remains unknown. Estimates of the amount of plastic in the surface waters of the Great Lakes only accounts for less than 1% of the estimated input (Hoffman and Hittinger, 2017)—a result that is consistent with similar estimates in the oceans (Eriksen et al., 2014; van Sebille et al., 2015).

One of the challenges with understanding the fate and transport of plastic pollution is that the distribution of plastic pollution is heterogeneous in space and time. Most of the field survey data that exists is of the surface waters, but these samples are expensive and labor-intensive and therefore have limited spatial and temporal coverage. Furthermore, these samples can exhibit a high variance, with trawls taken from the same location an hour apart yielding

* Corresponding author.
E-mail address: sam7201@psu.edu (S.A. Mason).

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concentrations that differ by as much as 300% (Cable et al., 2017). Additionally, little to no field survey data is available in the benthic or sedimentary regions (Ballent et al., 2016; Dean et al., 2018). This paper helps address the lack of surface water samples in the Great Lakes by presenting the first numbers for Lake Ontario, as well as a three-year surface sample data set on Lake Erie. This is the first multi-year set on the Great Lakes and allows for more analysis of interannual distribution and variability than existing data.

Even with increased data, however, there is still not sufficient resolution to capture the basin-wide distribution from samples alone. Lagrangian particle transport models can help understand the spatial distribution of plastic pollution by providing spatial and temporal resolution of the basin-scale which is impossible from in situ samples. Models have been used in the global ocean to identify major garbage patches (Lebreton et al., 2012; Lebreton and Borrero, 2013; Maximenko et al., 2012; van Sebille et al., 2012), predict transport of debris in the Mediterranean, and look at the distribution in the Great Lakes (Cable et al., 2017; Hoffman and Hittinger, 2017). While models are adept at producing spatial patterns, turning model output into environmental concentrations requires comparison with sample data. In this paper we compare results from a Lagrangian transport model of Lake Erie with a previously unpublished multi-year surface sample set to explore the potential to compare models and samples directly and refine floating mass estimates of microplastic in Lake Erie.

Materials and methods

Sample collection

During the summer of 2013, a total of 39 open-water samples were obtained within Lakes Erie (17 samples) and Ontario (22 samples). An additional 8 samples each were collected from Lake Erie during July 2012 (Eriksen et al., 2013) and 2014, yielding a combined total of 55 samples from both lakes during 2012–2014 (Figs. 1 and 2). All samples were collected with the use of a manta trawl, which consists of an aluminum frame with a rectangular opening 16 cm high by 61 cm wide attached to a 3 m long, 333 μm-mesh net with a 30 cm long, 10 cm diameter, collecting bag. The trawl was towed along the lake surface and was positioned with the towline sitting outside of the ship's wake. The sea state for all sample sites was fairly calm with values between 1 and 3 on the Beaufort Wind Scale. While the sample sites were not equidistant from one another nor were the transects of equal length, all samples were collected for 30 min (timed using a stopwatch) and the tow speed was kept under 2.0 knots. The tow length was determined in triplicate (utilizing GPS coordinates, a flowmeter suspended within the mouth of the trawl net and via an onboard knotmeter) and averaged. The surface area sampled could then be calculated by multiplying the tow length by the width of the trawl mouth opening. All samples were rinsed from the collection bag into a sample container and immediately preserved in 70% isopropyl alcohol for later laboratory processing and analysis.

Sample processing

Open-water samples were processed in a laboratory for isolation of plastic debris using a modified National Oceanic and Atmospheric Administration marine debris protocol (Masura et al., 2015), briefly described here. Each sample is filtered through a series of 8-inch diameter Tyler sieves of 4.75 mm, 1.00 mm and 0.355 mm stainless steel mesh, which separates the solid material into 3 size classifications (0.355–0.999 mm, 1.00–4.749 mm and ≥4.75 mm). Solids within the largest size fraction (≥4.75 mm) were manually sorted to remove visible plastic debris from organic material. In order to collect any microplastics initially stuck to the organic material, all were rinsed with DI water with rinse water running through the stacked sieves. The solids in the smaller two size classifications (0.355–0.999 mm and 1.00–4.749 mm) were combined and subjected to a wet peroxide oxidation (WPO), which digests labile organic material using 30% hydrogen peroxide in the presence of an iron (II) catalyst.

It is important to note that the WPO method is stated (Masura et al., 2015; Tagg et al., 2017) to have negligible impact upon the most common plastic particulates. Nevertheless as demonstrated by Munno et al. (2018), the elevated temperatures that occur as a result of the WPO method, even in the absence of added heat, can lead to the melting and loss of some spherical plastic, namely those 'microbeads' which have wax character. As the temperature of the reaction was not monitored throughout the process, but showed characteristic boiling behavior of temperatures in excess of 75 °C, it is likely that some sphericals/microbeads/pellets were 'lost' via this chemical processing and thus the numbers reported here should be considered conservative.

After oxidative processing, samples were once again filtered through a stacked sieve set (1.00 mm and 0.355 mm) and all remaining particulates were transferred to individual glass Petri dishes using deionized water (DI) for visual analysis. Using a dissection microscope (Leica EZ4 HD, 40x), all microplastic particles within each size classification (0.355–0.999 mm and 1.00–

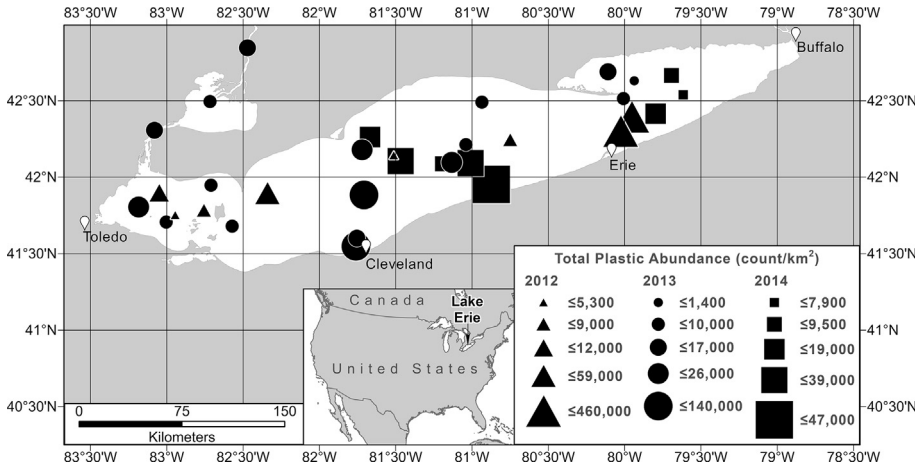

Fig. 1. Distribution of plastic abundance (particles/km^2) for the 33 open-water surface samples obtained across Lake Erie during the summers 2012–2014.

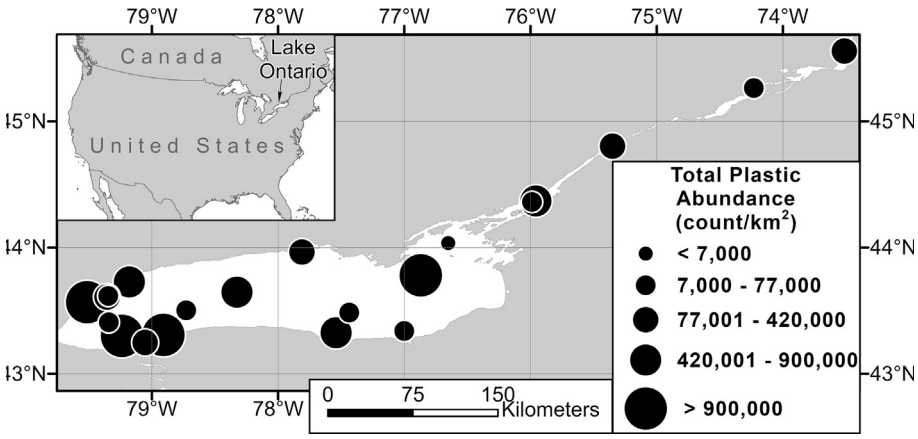

Fig. 2. Distribution of plastic abundance (particles/km^2) for the 22 open-water surface samples obtained across Lake Ontario during the summer 2013.

4.749 mm) were removed, enumerated and categorized by morphology as fragment, pellet, line/fiber, film, or foam (Mason et al., 2016; GESAMP, 2019) and archived for later spectroscopic analysis.

Quality assurance & quality control

Six blank samples in which DI water was stored within sample containers for periods of 1–14 days were processed concurrently with the open-water samples and only one was found to have any microplastic particulate (4 fibers) indicating that the risk of sample contamination from the containers, lab or processing was negligible. More recent studies (e.g., Baldwin et al., 2016) have established that there is a potential for particles, especially fibers, to be retained on the sampling net between samples and thus "carried over" to future samples. While lab blank samples were utilized during this study to account for potential laboratory/chemical contamination, no field blanks were utilized to account for this possible carry over effect. Further, no samples were 'spiked' in an attempt to account for any losses or recoveries. Seven of the samples were counted by two different research team members to verify overall counts and morphological categorization. While there was some variation in the morphology designations of the microplastics (given their subjective nature (GESAMP, 2019)), overall counts varied by less than 5%.

Spectroscopic analysis

A scanning electron microscope with an elemental detection system (SEM/EDS) was used to analyze the pelagic plastic particles within the smallest size fraction (i.e., 0.355–0.999 mm) in order to distinguish between organic and inorganic materials owing to the more reflective nature of minerals as compared to carbon-based materials (Eriksen et al., 2013). Based upon availabilities at the time of analysis, particles within this size range were not amenable to other spectroscopic techniques (namely FTIR discussed below). Additionally, the SEM can provide detailed images of the particle surface, which are not available via other analytical techniques. Of the 55 samples, 53 contained particles within this smallest size classification. Of these 53 sites, a 25% random selection of sites (13 sites total) were chosen for analysis. For these 13 sites, all particles within the smallest size classification were analyzed using the SEM/EDS system. Samples were prepared for SEM/EDS analysis by securing the particles onto double-sided carbon tape prior to imaging using a Phenom ProX desktop SEM/EDS system operating at 15 keV in backscatter mode.

Fourier Transform Infrared (FTIR) analysis was used to determine the polymeric composition of retrieved pelagic particles whose diameter (or longest particle edge) was greater than or equal to 4.75 mm. Of the 55 sampled sites, 47 sites contained a total of 263 particles within this largest size classification. Approximately half of these sites (23 sites total) were randomly selected for analysis and a total of 126 particles (48% of all particles $\geq$ 4.75 mm from all sites) were analyzed. For this analysis, particles were dissolved in a small amount of dichlorobenzene (~1 mL), which usually necessitated the use of some heat (100 °C). A small amount of this solution was then transferred and allowed to dry on a Real Crystal (NaCl) IR Card (International Crystal Laboratories). Samples were analyzed using a Matteson Polaris FTIR operating at 32 scans and 4 cm^{-1} resolution. Background scans using a Real Crystal IR Card with only dichlorobenzene dried to its surface were obtained every fifth spectra or each new day, whichever came first. Background spectra are automatically subtracted from each raw spectra to yield sample spectra. A library of spectra from known polymers was created by using in-house polymer samples or from common consumer products, such as plastic bottles, containers and cigarette filters. Sample spectra were compared to this library for polymeric composition identification to support visual interpretation.

As the instrumental equipment available at the time of analysis are both 'destructive' techniques (e.g., dissolution to form films for FTIR and bound to carbon tape for SEM/EDS), retrieved pelagic plastic particles within the middle size classification (1.00–4.749 mm) were not analyzed by either technique. Rather these particles were archived for future research endeavors.

Hydrodynamic modeling

The existing modeling work that has been done on plastic transport in the Great Lakes is based on Lagrangian transport models. Hoffman and Hittinger (2017) used a 2D model with flow between lakes to derive a first pass mass estimate of plastic in the surface waters of Lake Erie. Cable et al. (2017) used a similar model in Lake Erie to model the behavior of both positively buoyant and neutrally buoyant particles and compared model results to samples. In this work we use the same basic modeling framework as Hoffman and Hittinger (2017), but with currents from a different hydrodynamic model. Here the model used to hindcast currents in Lake Erie is the Finite Volume Community Ocean Model (FVCOM) (Chen et al., 2006).

The horizontal velocities were obtained by running the FVCOM model for the years 2012–2014. FVCOM is a hydrodynamic model used by the NOAA Great Lakes Environmental Research Laboratory (GLERL) to operationally predict currents, temperature, and water levels in Lake Erie, Lake Michigan, and Lake Huron. It replaced

the Princeton Ocean Model that was previously used by NOAA GLERL. FVCOM utilizes an unstructured, triangular grid in the horizontal direction, which has the advantage of smoothly fitting to the shoreline. In the vertical direction, FVCOM uses terrain following sigma coordinates to account for the bathymetry (Chen et al., 2006). The FVCOM model of Lake Erie has been used to model various phenomenon such as algal transport or plankton dynamics (Jiang et al., 2015; Rowe et al., 2016). FVCOM code was obtained from NOAA GLERL along with wind and boundary condition forcing and the model was run on the NSF funded XSEDE Comet cluster (Towns et al., 2014).

The Lagrangian transport model here assumes that advection is the driving force of movement, with the effects of diffusion on the movement of the particles ignored. Our advection model is based on the methods used in other Lagrangian studies (Mendoza and Mancho, 2012) and has the same underlying MATLAB code as previous work for two dimensional model of plastic transport in the Great Lakes (Hoffman and Hittinger, 2017). For efficiency, and to follow previous work, we include an intermediate step of interpolating the FVCOM output to a uniform 2 km rectangular grid using linear interpolation. This regular interpolated output was then used as the input to the particle transport model. In the horizontal direction, the particle positions, $x(t)$ and $y(t)$, are progressed forward in time using the dynamical system,

$$\frac{dx}{dt} = u(x, y, t) \tag{1}$$

$$\frac{dy}{dt} = v(x, y, t) \tag{2}$$

where $u(x, y, t)$, and $v(x, y, t)$, are the interpolated horizontal x-direction and y-direction velocities, respectively, from FVCOM. The velocities are interpolated in time using third-order Lagrange interpolation and in space, to particle locations, using cubic interpolation. The system is then solved using the Runge-Kutta 4th-order numerical scheme (RK4) with timesteps of one hour.

To compare the model to data in each year (2012–2014), the transport model is run for each year with no particles in the lake initially. Every 12 h of the simulation, particles are released from each grid point along the shore of the lake, for a total of 492 particles per 12 h. This is different from previous plastic pollution modeling where particles were released with a frequency that corresponded to the relative population density at the shore grid point. The choice to release particles at every grid point is made to ensure that enough particles have entered the lake by the time the comparison is made to the samples. When releases were made proportionally to population it was observed that some areas of the lake had essentially no particles in them because they were close to regions with low populations. To take population into account here, the near shore population size at the release point of the particle is recorded as a property of the particle and used in the analysis for weighting. Population size here is used to serve as a proxy of the amount of pollution released from a location. Population is calculated in the same way as in Hoffman and Hittinger (2017): we collect population information from 33,120 zip codes and 56,203 Dissemination Areas around the Great Lakes and population impacting each shoreline grid point is computed by summing the effects of all zip codes and Dissemination Areas within 100 km assuming that the influence decays according to a normal distribution with standard deviation of 10 km. For a more detailed description of the derivation of population estimates for nearshore grid points see Hoffman and Hittinger (2017).

Particles are not carried over from the 2012 simulation to 2013 because that would mean that the 2013 simulation would have more particles than the 2012 simulation, which would bias the regression. This decision does mean that there is a chance that some plastic that has been in the lake for years is missing from the simulation. The loss of pelagic plastic via biofouling, which acts to increase the density of particles causing them to sink, is also not represented here.

At each 3 h time interval, the particles within a 2 km radius of the sample locations, along with the population at their origin, are saved. To assess the fit of the model to the samples, we perform a linear regression between the observed count of particles at each sample point at a given time, x_o^s (where $s = 1, \cdots, n$ and n is the number of samples for that year) to the approximation of model particles at the same sample site by counting all modeled particles within a radius R of the observation location, x_m^s (Fig. 3). In order to reduce the impact of small temporal variations we compute model averages before performing the regression. We experimented with averages of one, two, and four weeks and obtained similar results so we elected to complete the comparison using the one week average (Fig. 3). This regression between the model and sample data is also used to derive a conversion factor between model output and in-lake particle abundance, which allows for an estimated total number of particles in the surface of Lake Erie.

The sum of the near-shore populations of particles is also computed and denoted as the population-weighted model count, $x_{mpop}^s = \sum_i^{particles \in R} P_i$ where P_i is the near shore population at the origin of the particle. The nearshore population is used for weighting because particles are released at the same rate at every shore location.

To derive a mass estimate, the number of particles can be multiplied by an average mass per particle. While mass concentrations were not reported in the Great Lakes samples, we can derive crude estimates by using values from the ocean literature. One study estimated between 93 and 236 thousand metric tons floating in the ocean by using three separate values to convert from particle concentrations to mass concentrations (van Sebille et al., 2015). Here we take the middle number used in that study—1.36×10^{-2} g/particle—and use that as our conversion factor (Morét-Ferguson et al., 2010).

Results and discussion

During the summers of 2012–2014, a total of 55 open-water samples were obtained from the surface of Lakes Erie (Fig. 1) and Ontario (Fig. 2). Plastic particles extracted from the samples were categorized and counted (Table 1). Using these counts and the surface area sampled (tow length × trawl net width), the plastic abundance (no. of particles per square kilometer, particles/km^2) for each

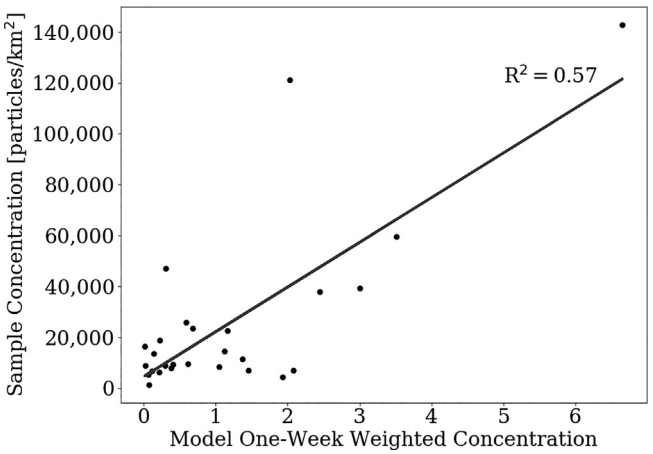

Fig. 3. Regression of normalized model and sample concentrations after the two 2012 eastern Lake Erie samples are removed.

Table 1
Locations, counts and abundances of pelagic plastic for all 55 Lakes Erie and Ontario samples.

				Particle Counts						Tow Length	Abundance
Sample ID	Date	Latitude	Longitude	Fragments	Pellets	Fibers/Lines	Films	Foams	Total	(km)	(count/km^2)
Lake Erie											
GL12 #14	24-Jul-2012	41.90	−83.05	20	5	–	–	1	26	3.70	11,510
GL12 #15	25-Jul-2012	41.75	−82.95	4	1	–	1	3	9	3.36	4385
GL12 #16	28-Jul-2012	41.78	−82.76	12	4	–	–	4	20	3.86	8487
GL12 #17	28-Jul-2012	41.90	−82.34	7	–	–	–	124	131	3.61	59,450
GL12 #18	29-Jul-2012	42.14	−81.51	11	1	–	–	1	13	4.03	5294
GL12 #19	29-Jul-2012	42.24	−80.75	12	–	1	–	8	21	3.84	8956
GL12 #20	29-Jul-2012	42.39	−79.95	399	695	2	–	5	1101	3.89	463,423
GL12 #21	29-Jul-2012	42.30	−80.03	349	295	2	7	4	657	3.65	294,998
GL13 #1	24-May-2013	42.69	−80.11	17	2	3	–	–	22	2.64	13,660
GL13 #2	24-May-2013	42.63	−79.94	2	–	–	–	–	2	2.31	1422
GL13 #3	25-May-2013	42.51	−80.01	12	–	–	–	–	12	2.93	6715
GL13 #4	25-May-2013	42.21	−81.04	7	–	1	–	–	8	2.20	6332
GL13 #5	25-May-2013	42.18	−81.72	25	–	5	2	–	32	2.22	23,577
GL13 #6	26-May-2013	41.68	−82.57	11	–	3	1	1	16	2.78	9425
GL13 #7	27-May-2013	41.71	−83.00	7	2	–	–	–	9	2.13	6917
GL13 #8	27-May-2013	41.81	−83.18	29	2	2	–	–	33	2.41	22,467
GL13 #9	27-May-2013	42.31	−83.08	18	–	7	–	–	25	3.12	13,125
GL13 #10	27-May-2013	42.85	−82.47	11	1	4	–	1	17	1.65	16,860
GL13 #11	2-Jun-2013	42.49	−82.72	13	1	–	–	–	14	2.22	10,327
GL13 #12	2-Jun-2013	41.95	−82.71	12	–	2	–	–	14	2.59	8872
GL13 #13	5-Jun-2013	41.60	−81.75	10	–	11	–	–	21	2.38	14,449
GL13 #14	5-Jun-2013	42.49	−80.94	10	–	1	–	–	11	2.59	6955
GL13 #96	3-Aug-2013	41.55	−81.76	66	73	–	5	26	170	1.95	142,874
GL13 #100	6-Sep-2013	41.88	−81.71	60	66	–	–	12	138	1.87	121,073
GL13 #101	6-Sep-2013	42.10	−81.13	10	7	–	1	17	35	2.22	25,893
GL14 ER#63	8-Jul-2014	42.42	−79.80	12	10	4	–	–	26	2.26	18,819
GL14 ER#09	8-Jul-2014	42.54	−79.62	4	–	1	–	3	8	1.67	7853
GL14 ER#10	8-Jul-2014	42.66	−79.69	3	1	2	2	3	11	1.90	9499
GL14 NER#32	10-Jul-2014	41.95	−80.87	15	45	4	6	3	73	2.54	47,106
GL14 ER#32	10-Jul-2014	42.09	−81.01	18	38	–	4	1	61	2.54	39,335
GL14 ER#30	10-Jul-2014	42.91	−81.19	5	3	4	–	–	12	2.09	9405
GL14 ER#38	10-Jul-2014	42.26	−81.67	9	10	1	2	–	22	2.20	16,383
GL14 ER#36	10-Jul-2014	42.11	−81.47	14	21	6	4	8	53	2.29	37,889
Lake Ontario											
GL13 #15	9-Jun-2013	43.34	−77.00	11	4	1	–	–	16	1.75	14,957
GL13 #16	12-Jun-2013	43.33	−77.54	456	21	1	2	149	629	4.29	240,120
GL13 #17	15-Jun-2013	43.31	−78.91	859	109	20	41	23	1052	2.79	618,844
GL13 #18	16-Jun-2013	43.25	−79.05	66	2	3	6	26	103	1.30	130,247
GL13 #19	17-Jun-2013	43.30	−79.24	455	6	5	15	8	489	1.25	643,406
GL13 #20	20-Jun-2013	43.57	−79.51	1148	30	22	5	2	1207	2.54	778,859
GL13 #21	21-Jun-2013	43.73	−79.18	433	20	27	18	30	528	4.25	203,465
GL13 #22	23-Jun-2013	43.96	−77.81	101	15	5	2	–	123	2.04	98,855
GL13 #30	13-Jul-2013	45.56	−73.51	94	14	22	9	1	140	1.48	154,906
GL13 #31	14-Jul-2013	45.26	−74.23	9	3	2	–	–	14	0.93	24,785
GL13 #32	15-Jul-2013	44.80	−75.35	27	3	–	1	–	31	0.46	109,762
GL13 #33	16-Jul-2013	44.37	−75.96	103	15	97	18	1	234	1.86	206,256
GL13 #34	17-Jul-2013	44.04	−76.65	6	–	3	–	–	9	2.42	6104
GL13 #35	17-Jul-2013	43.78	−76.87	1483	519	12	33	8	2055	2.44	1,378,327
GL13 #36	17-Jul-2013	43.48	−77.44	40	13	–	1	–	54	2.07	42,738
GL13 #37	18-Jul-2013	43.65	−78.33	56	86	39	–	40	221	1.90	190,577
GL13 #38	18-Jul-2013	43.50	−78.73	64	–	1	–	6	71	2.28	50,986
GL13 #39	19-Jul-2013	43.62	−79.35	77	2	2	3	10	94	2.81	54,823
GL13 #92	14-Jul-2013	45.27	−74.23	7	6	7	7	5	32	0.93	56,651
GL13 #93	16-Jul-2013	44.36	−75.99	7	3	1	1	–	12	1.55	12,702
GL13 #94	19-Jul-2013	43.61	−79.36	183	27	12	21	–	243	2.94	135,679
GL13 #95	22-Jul-2013	43.41	−79.34	13	2	–	2	23	40	1.65	39,733

sample was determined (Table 1; Figs. 1 and 2) in order to standardize the data due to the varying tow lengths. Across both lakes for all dates sampled plastic abundances varied from ~1400 to more than 1.3×10^6 particles/km^2.

Plastic particle abundances: Lake Erie

During the summers of 2012–2014 a total of 33 samples were collected from the surface of Lake Erie (Fig. 1), with the first 8 of these samples (acquired in 2012) having been previously reported (Eriksen et al., 2013). All samples showed some presence of plastic pollution with abundances ranging from ~1400 to over 460,000 particles/km^2.

Table 2 details calculated particle abundances for Lake Erie only, averaged over each sampling year by morphology and size for all sampling years, as well as combined. Overall counts for 2013 and 2014 were lower as compared to 2012. For example, 2013 and 2014 yielded averages of roughly 26,000 and 23,000 particles/km^2 across all sampling sites, respectively, with maximum abundances of nearly 143,000 and 47,000 particles/km^2, respectively, while in 2012 the average was over 100,000 particles/km^2, with a high of over 460,000. Despite the among-year differences and within-year variabilities, there are some consistencies. For

Table 2
Plastic abundances (count/km^2) for Lake Erie averaged over each sampling year and combined across all years, distinguished by size and particle type.

	0.35–0.99 mm	1.00–4.74 mm	>4.75 mm	% of total
	2012 Data			
Fragment	28,401.7	14,360.3	1402.5	41%
Pellet	53,073.3	598.5	52.6	50%
Fiber/Line	0.0	214.7	56.1	0%
Film	0.0	112.3	341.5	0%
Foam	6319.4	1903.7	226.3	8%
count/km^2	87,794.4	17,189.4	2079.1	
% of total	82%	16%	2%	
	2013 Data			
Fragment	8403.2	4115.4	1634.5	53%
Pellet	7019.5	555.6	103.2	29%
Fiber/Line	804.3	619.9	121.1	6%
Film	191.8	49.4	170.8	2%
Foam	1248.0	705.3	784.2	10%
count/km^2	17,666.8	6045.6	2813.8	
% of total	67%	23%	11%	
	2014 Data			
Fragment	5004.2	1238.7	897.0	31%
Pellet	10,296.7	510.1	0.0	46%
Fiber/Line	1457.6	497.4	89.4	9%
Film	899.7	173.7	492.6	7%
Foam	923.3	716.8	89.4	7%
count/km^2	18,581.4	3136.6	1568.3	
% of total	80%	13%	7%	
	All Years Combined			
Fragment	41,809.1	19,714.3	3934.0	42%
Pellet	70,389.6	1664.2	155.8	46%
Fiber/Line	2261.8	1331.9	266.6	2%
Film	1091.5	335.4	1004.9	2%
Foam	8490.7	3325.7	1099.9	8%
count/km^2	124,042.6	26,371.6	6461.1	
% of total	79%	17%	4%	

example, for all years the majority of particles are found within the smallest size classification (0.355–1 mm), ranging from 67% to 82% (with an average of 79%) of the total retrieved plastic particles. Plastic fragments and pellets are the most common morphologies of plastic particles retrieved with combined abundances of 77–91% (average = 88%) of the total, while fibers/lines, films and foams represent a minor contribution (9–23%; average = 12%) (Table 2).

Lake Erie is the smallest Great Lake by volume and consists of a shallow (average depth 7.32 m) western basin, a deep (average depth 27.4 m) eastern basin and a relatively flat (average depth 18.3 m) central basin which is the largest by surface area. The central basin is large enough to feel the effects of the earth's rotation, especially in the summer when it becomes stratified. Large scale circulation exhibits significant variability during the stratified period May to October. It is rather weak in May, but currents pick up in speed and large-scale anticyclonic circulation develops especially strongly in July. Outside of this seasonal effect, annual mean current modeling suggests that currents in the Lake Erie push southward along the shorelines (Beletsky et al., 1999). As visualized in Fig. 1, we note that the plastic abundances generally increase as we follow the flow of water easterly across the lake, with abundances within the central and eastern basins being greater than those within the western basin. Additionally we note that the highest abundances for all sampling years occur within those samples along the southern shoreline of the lake (Fig. 1), which is consistent with the highest population centers—Cleveland, Toledo, Buffalo, and Erie which are all on the south shore—and with annual mean surface current modeling which traps particles along the southern shore and moves them eastward (Beletsky et al., 1999; Hoffman and Hittinger, 2017).

Plastic particle abundances: Lake Ontario

During June and July 2013, two separate expeditions, yielding a total of 22 samples, were conducted to investigate the prevalence of plastic pollution within the open-waters of Lake Ontario and the St. Lawrence River (Fig. 2). All samples obtained showed some presence of plastic pollution with particle abundances ranging from ~6000 to over 1.3×10^6, with an average of over 230,000 particles/km^2. These data are the first to be reported for the open waters of Lake Ontario.

Table 3 provides more detailed information of plastic particle abundance by morphology and size. As with previous Great Lakes studies (Eriksen et al., 2013; Mason et al., 2016) and with Lake Erie above, the smallest size classification dominates (68%) with regard to overall percentage, with the largest contributors by morphology being fragments (76%) followed by pellets (12%).

As visualized in Fig. 2, Lake Ontario has the highest counts of pelagic plastic pollution on the surface any Laurentian Great Lake to-date. The highest abundances appear on the western end of the lake, close to the outfall from Lake Erie (i.e., the Niagara River) and the population center of Toronto, Ontario, Canada, as well as at the mouth of the St. Lawrence River on the eastern edge of the lake. Lower relative abundances are found within the central area of Lake Ontario. As with Lake Erie above, this is consistent with annual mean surface current modeling, which notes a seasonal gyre formation on the western end of the lake, with current transport out of this circulation, along the southern shoreline to the mouth of the St. Lawrence River (Beletsky et al., 1999).

The consistently high counts (relative to the other Great Lakes) associated with our Lake Ontario samples can be attributed to two factors. First is the fact that Lake Ontario and the St. Lawrence River represent the final stage within the Great Lakes ecosystem prior to it flowing out into the North Atlantic Ocean. Thus all of the plastic particles that entered the system since its start in Lake Superior, if they are not decomposed or 'lost' somewhere along the way (via beaching (Smith and Markic 2013) or biofilm-induced sedimentation (Morét-Ferguson et al., 2010), for example), will flow into Lake Ontario on their way to the sea; adding to the contributions from those living within the Lake Ontario watershed. While the outflows

Table 3
Plastic abundances (count/km^2) for 2013 Lake Ontario averaged over all 22 samples, distinguished by size and particle type.

	0.35–0.99 mm	1.00–4.74 mm	>4.75 mm	% of total
Fragment	123,108.0	55,198.6	2218.3	76%
Pellet	25,293.3	3188.4	364.8	12%
Fiber/Line	8030.7	1784.4	704.4	4%
Film	2588.3	2945.1	1062.8	3%
Foam	2534.3	6096.4	917.9	4%
Total	161,554.6	69,212.9	5268.1	
% of total	68%	29%	2%	

Table 4
Comparison of results of this study to previous Laurentian Great Lakes study in percentages by size and particle type.

	Size Classification			Morphology				
	0.35–0.99 mm	1.00–4.74 mm	>4.75 mm	Fragment	Pellet	Fiber/Line	Film	Foam
Eriksen et al., 2013	81%	17%	2%	42%	48%	<1%	1%	8%
Mason et al., 2016	59%	32%	9%	79%	4%	14%	2%	1%
Cable et al., 2017	93%[a]	6%	1%	97%	1%[b]	<1%[c]	<1%	1%
This study	73%	24%	3%	63%	26%	4%	2%	6%

[a] The lower limit within smallest size classification in this study was 106 μm.
[b] To be consistent with previous studies, the pellet category here includes the combination of nurdles and spheres detailed in this study.
[c] Does not include fibers since, as the authors note, they could not be quantified with equal high confidence across all sizes.

of pelagic plastic pollution from Lake Superior and Lake Michigan into Lake Huron might be quite slow given their long hydraulic residence times (199 years and 99 years, respectively), the flow from Lake Erie to Lake Ontario would be much more rapid given Lake Erie's relatively short residence time of just over 2 years. Secondly, because of the nature of the sampling campaigns and boats utilized, the two Lake Ontario/ St. Lawrence River expeditions circumnavigated the lake, tending to remain closer to the shore rather than the center of the lake. Several studies (Eriksen et al., 2013; Ballent et al., 2016; Cable et al., 2017) have noted that particle counts tend to be higher closer to the shoreline likely owing to a combination of source proximity, urbanization/industrialization, and lack of dilution, combined with surface current aggregation of floating materials.

Plastic particle abundances: comparison to previous studies

Three other studies have been conducted within the open-waters of the Laurentian Great Lakes (Cable et al., 2017; Eriksen et al., 2013; Mason et al., 2016). Table 4 compares on a percentage basis our numbers with these previous studies by size and morphological classification. Consistently among all of these studies, the majority of particles are found within the smallest size classification. Notably, the smallest size class within the Cable et al. (2017) study went down to 106 μm (rather than 355 μm for the other studies) and, thus, found an even greater percentage of particles within this size class. This is consistent with the growing literature on marine plastic pollution and the photoxidative mechanisms which will continue to break plastic particulate into smaller and smaller pieces with little change in their chemical nature.

Also consistent within all of these studies is that fragments and pellets dominate the other morphological classifications (fibers/lines, films, and foams), representing a combination of 83–98% of the sampled particles. Within these dominant morphologies, generally fragments are more prominent as compared to pellets with the exception of Eriksen et al. (2013) who found a slightly higher percentage of pellets as compared to fragments though this was largely owing to two out of the 21 samples within that study. Additionally the sample processing methodology of Eriksen et al. (2013), which utilized only density separation, differed from the other two studies that utilized other biochemical methods (which could have influenced the prominence of pellets). While fibers appear to be a minor contributor (<1%–14%) to the open-water samples, studies on tributaries to the Great Lakes (Baldwin et al., 2016; McNeish et al., 2018) indicate they dominate over other morphologies. This difference may be due to the fact that fibers are typically comprised of polymers that are denser than water. While the turbulence of a riverine system may keep such particles afloat, they would be expected to sink in less turbulent systems, such as large lakes.

Plastic composition analysis

Several studies have confirmed the need to analyze retrieved particulates obtained in environmental sampling in order to confirm their identification as plastic as compared to natural materials (Eerkes-Medrano et al., 2015; Eriksen et al., 2013; Filella, 2015). Based upon SEM/EDS analysis an average of 15% of the particles within the 0.355–0.999 mm size class were found to be misidentified as plastic when they were really of mineral origin (Table 5), results similar to those from Eriksen et al. (2013) and Mason

Table 5
Summary of SEM/EDS analysis on retrieved particles in smallest size classification (0.355–0.999 mm).

	Number of Particles Analyzed	Percent of Total	
Sample ID	0.35–0.99 mm	Plastic	Mineral
GL12 #14	13	69%	31%
GL12 #16	10	100%	0%
GL12 #17	233	100%	0%
GL12 #18	11	100%	0%
GL12 #19	19	89%	11%
GL12 #20	1729	72%	28%
GL12 #21	707	73%	27%
GL13 #4	6	67%	33%
GL13 #8	9	89%	11%
GL13 #9	12	92%	8%
GL13 #13	4	100%	0%
GL13 #17	29	93%	7%
GL13 #34	7	57%	43%
Total/Average	2789	85%	15%

Table 6
Summary of FTIR results on retrieved pelagic plastic particles in largest size classification (≥4.75 mm). PE = polyethylene; PP = Polypropylene; PS = Polystyrene.

	Number of Particles Analyzed	Type of polymer			
Sample ID	>4.75 mm	PE	PP	PS	Other/Unknown
GL13 #4	1	–	1	–	–
GL13 #5	4	3	1	–	–
GL13 #6	1	–	1	–	–
GL13 #7	1	–	1	–	–
GL13 #8	4	2	2	–	–
GL13 #9	1	1	–	–	–
GL13 #12	1	1	–	–	–
GL13 #14	1	–	1	–	–
GL13 #15	2	1	1	–	–
GL13 #16	17	6	8	–	3
GL13 #17	36	18	16	–	1
GL13 #18	8	3	4	1	–
GL13 #19	5	3	1	1	–
GL13 #20	5	2	3	–	–
GL13 #21	18	10	3	–	5
GL13 #22	5	3	2	–	–
GL13 #30	3	–	2	–	1
GL13 #31	2	1	1	–	–
GL13 #32	1	–	1	–	–
GL13 #34	2	2	–	–	–
GL13 #35	4	2	1	–	–
GL13 #36	2	–	2	–	–
GL13 #39	2	–	2	–	–
Total	126	58	54	2	11
Percentage		46%	43%	2%	9%

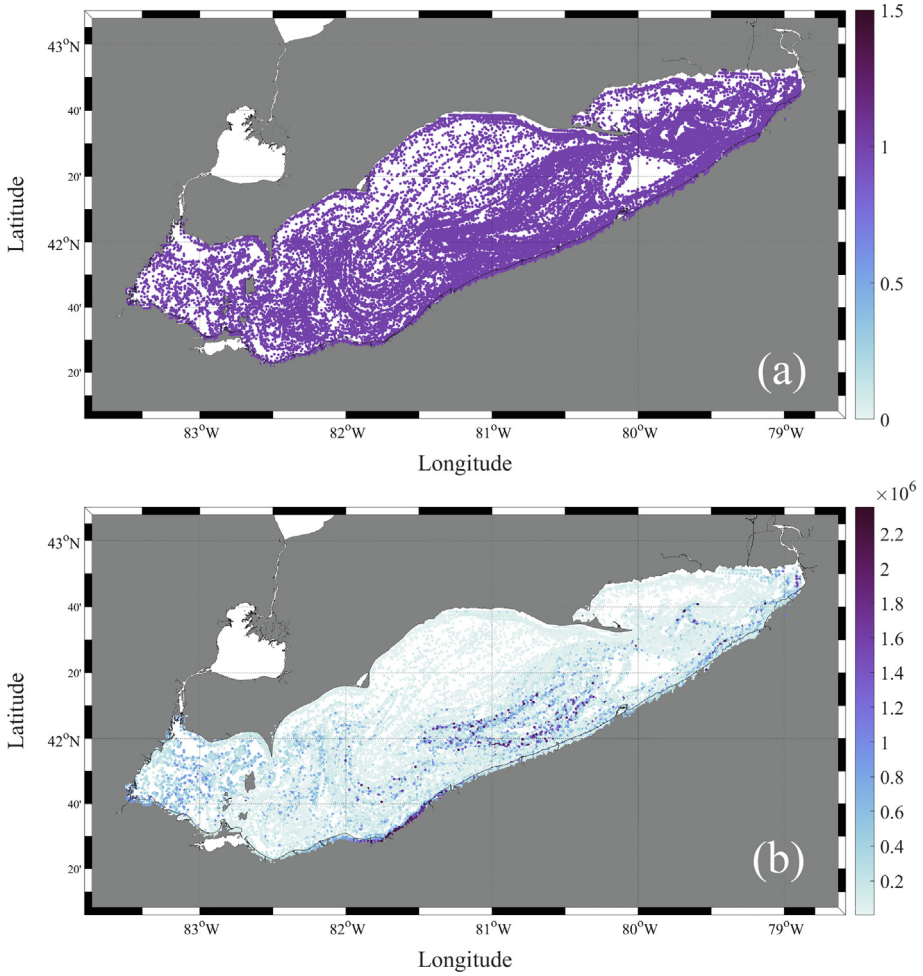

Fig. 4. Results of year-long hydrodynamic modeling simulations of Lake Erie where, due to the dynamics of the lake, uniform particle release leads to heterogeneous distributions. (a) Shows equal weighting of particles, while (b) shows particles weighted by the nearshore population of the release location to provide insight into particle origin and movement.

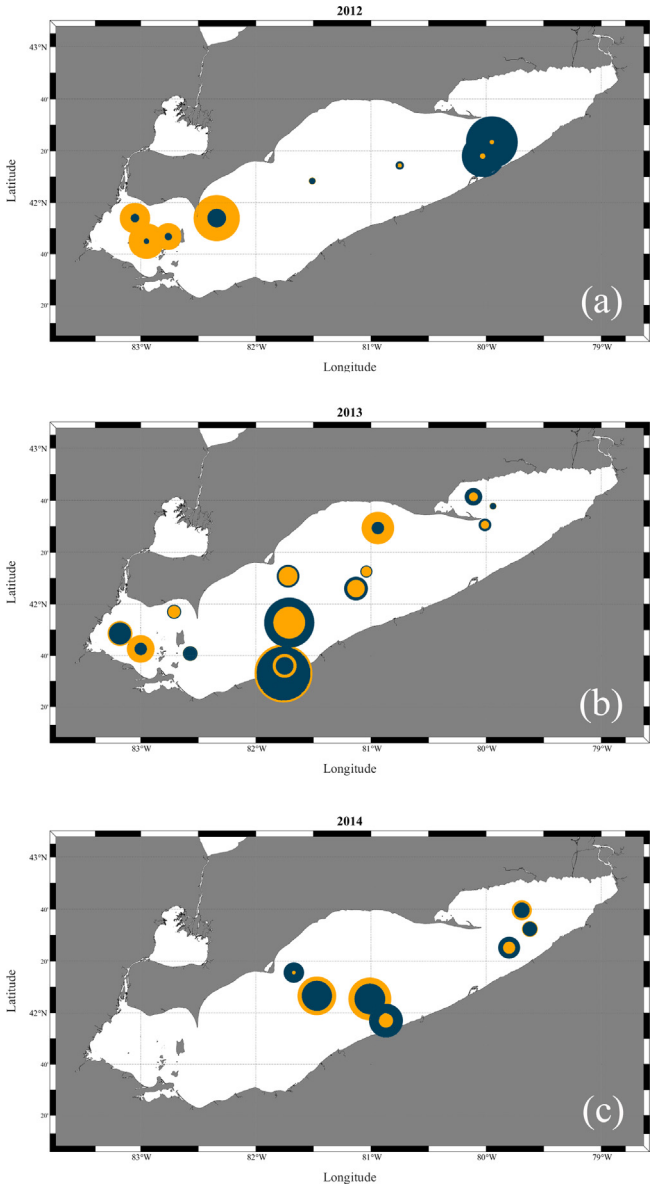

Fig. 5. Comparison of normalized model (grey) and sample (black) concentrations for (a) 2012, (b) 2013, and (c) 2014.

et al. (2016). All counts within this size class were adjusted accordingly (Table 1).

Table 6 details the FTIR results of particles within the largest size classification. Polyethylene (PE) was found to be the dominant polymeric type (46%), with high-density polyethylene being more prevalent (88%) as compared to low-density. Polypropylene (PP) was found to be the second most abundant polymeric type (43%) within these samples. These findings are consistent with global trends in the mass production of plastics, with polyethylene, followed by polypropylene, being the most widely manufactured polymers (Europe, 2017), as well as with the findings of beach surveys (Zbyszewski and Corcoran, 2011) and similar open-water studies (Mason et al., 2016).

The prominence of PE and PP could also be related to the dominant particle morphologies. Fragments and pellets have a lower surface area to volume ratio as compared to fibers. Biofouling and/or particle adherence, which acts to increase the density of particles, will influence the buoyancy of particles with a larger surface area to volume ratio to a greater extent given the increased area upon which to adsorb. Given this, fragments and pellets have more buoyant character as compared to other particle morphologies (GESAMP, 2019).

The remaining 11% of particles analyzed were found to be polystyrene (2%), polyurethane (2%), cellulose acetate (2%), copolymers (1%) or could not be identified (4%) given our instrumentation and polymer libraries (Table 6).

Comparison with modeling studies

Modeling studies indicated that, even when releasing particles uniformly from the shore, the dynamics of Lake Erie lead to a heterogeneous distribution after a year as particles converged in some locations and dispersed in others (Fig. 4a). Nearshore particle populations illustrated the movement of some particles from the Cleveland area spreading east along the southern shore and then being transported out into the Lake (Fig. 4b). These nearshore populations helped distinguish between areas in the central basin from those in the eastern basin that appeared to have high concentrations of particles (Fig. 4a). In the eastern basin the particles came from sparsely populated areas, while those in the central basin came more from the highly populated areas of Toledo (southwestern edge) and Cleveland (just west of middle of the southern shore) (Fig. 4b).

The 2012 Lake Erie samples were previously compared to a surface particle transport model in Hoffman and Hittinger (2017). That model had the same underlying Lagrangian transport code but used currents from the Princeton Ocean Model (POM) as opposed to the FCVOM currents used here. The previous study noted that the fit between the model and the samples was particularly bad in the eastern part of Lake Erie where sample concentrations of ~460,000 and ~300,000 particles/km^2 were measured but where the model predicted lower concentrations than in the western part of the lake. The same holds true in this model, with low concentrations predicted in eastern Lake Erie in 2012 as compared to the rest of the lake (Fig. 5a). There are no such issues in 2013 or 2014 as the normalized model concentrations match the normalized sample concentrations very well throughout Lake Erie (Fig. 5b and c). Based on these results, we made the same decision as in Hoffman and Hittinger (2017) to exclude those two points in 2012 from the regression (Fig. 3).

There are several potential reasons for the disagreement between the model and the samples for those two locations. There could be dynamics that are missing from the model, either horizontal diffusion or any type of vertical motion. It is also possible that debris was aggregated by currents of a smaller spatial scale than the 2 km grid model. Input could also be an issue. The model assumes uniform input with time, but some type of pulse event, such as a high rain event or a combined sewage overflow (CSO), could discharge a large amount of plastic at one time and lead to unusually large concentrations. The 2012 samples in eastern Lake Erie have an unusually large number of fragments and pellets in particular. In fact, the 695 pellets counted at one of those sites are the largest number of pellets from either lake in any year. In July 2012 there were some documented CSO events along the US shore of Lake Erie, but the transport model indicated that such a release from the southern shore was likely to stay trapped along the shore. It is possible that the inclusion of horizontal diffusion could bring some of those particles to the sample location, but preliminary simulations of advective transport backwards in time from the 2012 sample locations indicates that those particles were more likely to come from the Canadian shores. It is also possible that population centers upstream in rivers like the Grand River have a greater influence than the current Gaussian weighting allows, leading to an underestimation of input from the river mouth. Better quantification of river fluxes considering the water-

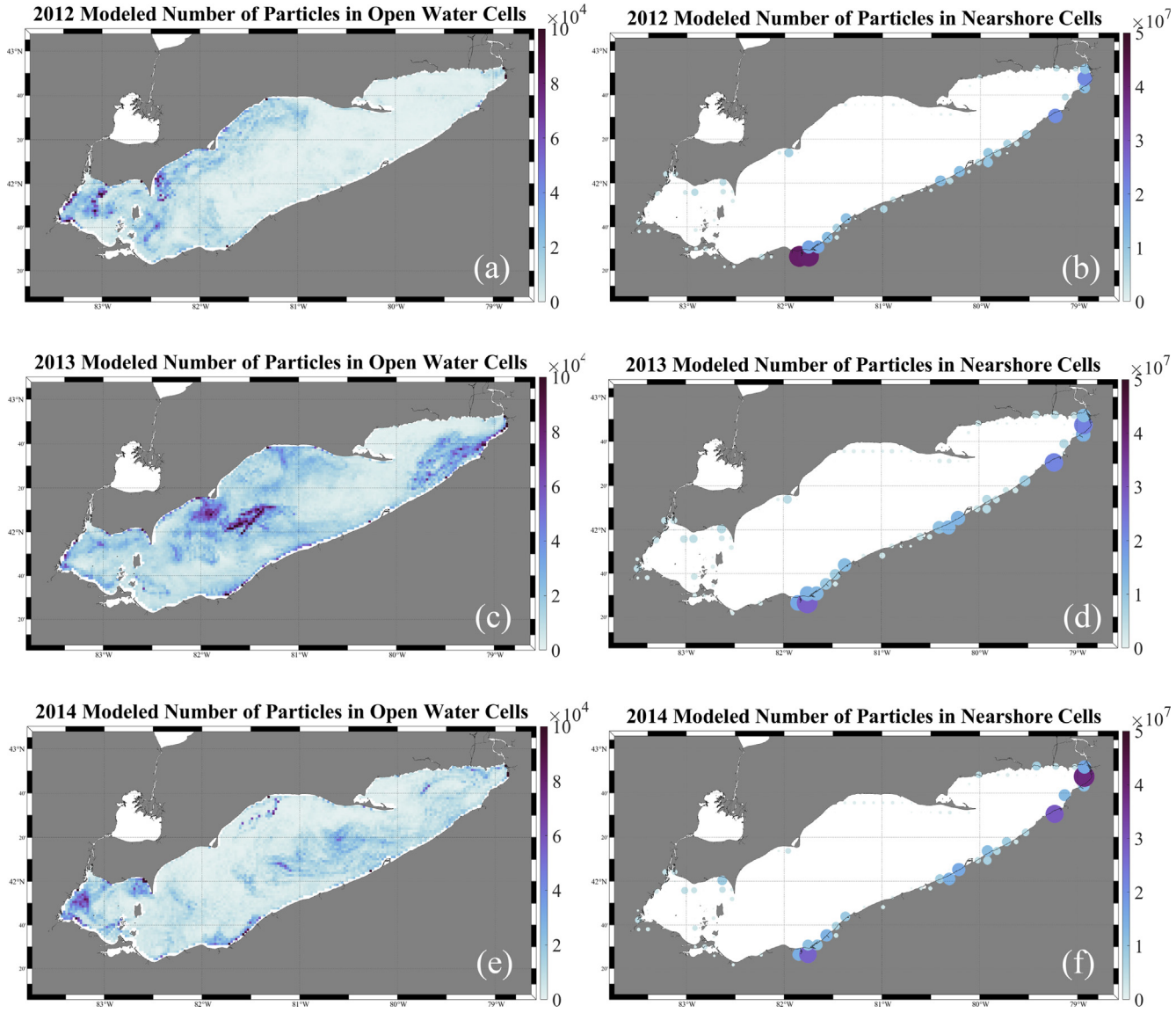

Fig. 6. Model predicted particles in 2 km × 2 km grid cells for open water (left column) and nearshore (right column) for the years 2012 (top row; a and b), 2013 (middle row; c and d), and 2014 (bottom row; e and f).

shed properties would be an important future step to improve model results.

A linear regression of the 2012–2014 data with those two points, mentioned above, removed has an R^2 value of 0.57 and gives a conversion factor of 17,561.33 to translate model concentrations to Lake Erie particle abundances (Fig. 3). Because this regression value was obtained using data from 2012 to 2014, each of the years (2012, 2013, 2014) yield a similar estimate of pelagic plastic particles of around 475 million. This quantity of particles yields a mass estimate of around 6.45 metric tons of floating microplastic, which is similar to the estimate of 4.41 metric tons from Hoffman and Hittinger (2017). However, the modeled spatial distribution of plastic in Lake Erie did vary between the years studied. There were higher modeled concentrations in the center of the lake during the 2013 sampling than in 2012 or 2014 (Fig. 6). In both 2012 and 2014 the highest concentrations were in the western part of Lake Erie and 2012 had the lowest modeled concentrations in the eastern basin (Fig. 6). This, as noted above, contrasts with the samples, which showed the highest nearshore concentrations, for all years, around Cleveland and near Buffalo. The most model particles were close to Cleveland during the sampling time in 2012 and to Buffalo in 2014, whereas 2013 has fewer particles nearshore in general (Fig. 6). These different model distributions provide insight into the variability of modeled estimates. For example, the use of separate regressions for each year yields values of: 4.85 metric tons (214 million particles) for 2012, 7.79 metric tons (573 million particles) for 2013 and 2.62 metric tons (193 million particles) for 2014. While stated by year, these numbers represent the variability in, rather than actual value of, modeled estimates.

Conclusions

Here we report the first plastic particle abundances for Lake Ontario, and the first multi-year dataset for Lake Erie. Perhaps unsurprisingly given its position as the last lake in the Laurentian Great Lakes ecosystem, Lake Ontario has the highest surface water abundances of any of the Great Lakes to-date with an average of over 230,000 particles/km^2, ranging from ~6000 to over 1.3×10^6. In comparison, the average across all Lake Erie samples,

across all years is just over 45,000 particles/km^2, with a range of 1400 to over 460,000. These studies are consistent with previously published work, showing the prominence of fragments and pellets as compared to fibers/lines, films and foams. While fibers/lines are often the most commonly identified morphology across many environmental compartments (e.g. air and aquatic species; Barrows et al., 2018), their reduced prominence in the Great Lakes samples may be owing to the negative buoyancy of polymeric fibrous materials causing them to more readily sink within the less turbulent inland seas environment.

This is the first time that modeled plastic distributions are compared to samples for specific years as well as for a multiyear period. The modeled distributions are reasonably good fit to the samples, with the largest misfit being in eastern Lake Erie in 2012. Using the sample data to calibrate the model, we estimate that 214–573 million microplastic particles are floating in the nearshore or open water of Lake Erie with a mass of 2.62–7.79 metric tons. This is in the same range as the only other published estimates of the floating mass in Lake Erie, which was computed using only 2012 data.

One particularly interesting question for future work is understanding why the 2012 fit in the eastern basin is so poor. There are several possible reasons for the misfit. The 2D model used here misses several key dynamics such as vertical movement and hydrodynamic features smaller than the 2 km grid size. There are also significant uncertainties regarding input, which is assumed to be temporally uniform when, in reality, large rainfall events can lead to significant point source events such as combined sewer outflows (CSOs) which have the potential to bring large amounts of plastic into the water. It is likely that point source input can have a significant impact, particularly on localized samples, and refining the input to include temporal variation is an important topic for future work.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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