

**Appendix to “An Analysis of Microlitter and Microplastics from Lake Superior Beach Sand and Surface-Water”**

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Table A1: Beach sand sample sites along with their site labels, names and coordinates in Degrees Minutes and Seconds.

Site Label and Name	Coordinates (DMS)	Date Sampled (MM/DD/YYYY)
Site 1: York	46°58'35.7"N 90°51'34.0"W	05/23/2018, 07/26/2018
Site 2: Rocky Island	47°01'12.0"N 90°40'42.4"W	05/23/2018
Site 3: Outer Island	46°59'42.9"N 90°28'02.9"W	07/26/2018
Site 4: Stockton-North (Julian Bay)	46°55'12.0"N 90°32'49.2"W	05/23/2018
Site 5: Stockton-South (Presque Isle Bay)	46°55'11.9"N 90°33'17.7"W	05/23/2018
Site 6: Long Island NE	46°42'15.6"N 90°45'44.1"W	05/23/2018
Site 7: Meyers Beach	46°53'02.2"N 91°02'53.7"W	05/23/2018, 07/25/2018
Site 8: Brule River Beach	46°44'31.8"N 91°36'21.0"W	05/23/2018
Site 9: Long Island SW	46°42'34.2"N 90°46'01.1"W	05/23/2018
Site 10: Knife River Beach	46°56'46.7"N 91°46'51.7"W	05/25/2018
Site 11: Park Point	46°43'48.5"N 92°02'56.2"W	05/25/2018
Site 12: Ontonagon Beach	46°53'03.1"N 89°18' 6.0"W	05/21/2018

Table A2. Water sample names, locations, and sampling dates

Sample name	Location (DMS): beginning of net tow	Location (DMS): end of net tow	Date sampled. (MM/DD/YR)
MB	46°53'11.2"N, 91°03'20.8"W 46°53'0.5"N, 91°03'28.5"W	46°53'33.4"N, 91°02'56.6"W 46°53'13.9"N, 91°03'14.2"W	05/21/18 07/25/18
LI	46°44'46.9"N, 90°47'56.4"W 46°44'48.8"N, 90°47'48.9"W	46°44'41.5"N, 90°47'13.0"W 46°45'0.7"N, 90°48'10.6"W	05/21/18 7/25/18
ST	46°57'38.9"N, 90°37'17.7"W	46°57'24.2"N, 90°37'35.2"W	7/25/18

Table A3. Plastic and nonplastic particles found within field replicates and blanks for the May 2018 sampling campaign. In terms of morphology in this table, fibers includes both individual fibers and fibrous bundles. Note that these counts are not blank corrected. \*indicates that sample was oxidized prior to counting, # indicates processing with unfiltered salt solution.

	Plastics/ 200g dry weight sample	Plastic morphology and color	Nonplastic microlitter/2 00g dry weight sample	Non plastic morphology and color
Site 1: York C(replicate 1)#	0		128	125 fibers (100 translucent, 4 transparent, 6 blue, 14 black, 1 white); 2 fragments (2 white), 1 film (1 translucent)
C(replicate 2)*	0		4	4 fibers (1 transparent, 2 blue, 1 black)
E#	0		138	135 fibers (104 translucent, 4 blue, 9 brown, 18 black), 3 fragments (2 translucent, 1 yellow)
Site 2: Rocky Island A#	2	1 fiber (translucent), 1 fragment (transparent)	182	175 fibers (125 translucent, 11 transparent, 5 red, 5 brown, 28 black, 1 green), 7 fragments (2 transparent, 2 yellow, 3 white)
C#	1	1 fragment (yellow)	147	145 fibers (128 translucent, 1 blue, 1 yellow, 4 brown, 11 black), 2 fragments (1 translucent, 1 yellow).
E#	1	1 fiber (red)	210	209 fibers (192 translucent, 1 red, 7 blue, 9 black), 1 film (1 translucent)
Field blank#	4	3 fibers (transparent), 1 fragment (yellow)	54	54 fibers (39 translucent, 3 transparent, 4 blue, 7 black, 1 green)
Site 4: Stockton-North (Julian Bay) C#	1	1 fiber (blue)	144	130 fibers (118 translucent, 1 transparent, 4 blue, 3 yellow, 2 black, 2 white), 11 fragments (8 translucent, 2 transparent, 1 white); 3 films (1 translucent, 2 transparent)
E#	3	1 fiber (translucent), 2 fragments (blue)	72	72 fibers (61 translucent, 1 red, 5 blue, 1 yellow, 2 brown, 2 black)
Site 5: Stockton-South (Presque Isle Bay) A#	7	2 fibers (1	225	213 fibers (168 translucent, 3 transparent,

		transparent, 1 black); 5 fragments (5 blue)		1 red, 4 blue, 7 brown, 30 black); 12 fragments (10 blue, 2 white).
C#	1	1 fiber (1 transparent)	454	441 fibers (413 translucent, 5 transparent, 2 red, 7 blue, 1 brown, 13 black), 2 fragments (1 blue, 1 white), 5 films (5 translucent), 5 other (“fibery films”, 5 translucent).
E#	0		247	246 fibers (240 translucent, 1 red, 3 blue, 2 black), 1 fragment (1 black)
Field blank#	9	3 fibers (2 transparent, 1 black), 6 fragments (6 blue)	70	69 fibers (48 translucent, 4 transparent, 6 red, 4 blue, 8 black), 1 fragment (translucent)
Site 6: Long Island NE A#	2	3 fibers (1 translucent, 2 transparent)	117	114 fibers (95 translucent, 8 transparent, 1 blue, 1 red, 1 white, 3 black, 2 grey, 1 brown), 3 fragments (1 translucent, 1 white, 1 brown)
C#	2	2 fibers (1 transparent, 1 black)	192	192 fibers (181 translucent, 2 transparent, 1 red, 3 blue, 1 brown, 4 white)
Field blank	0		14	14 fibers (14 translucent)
Site 7: Meyers Beach C#	11	10 fibers (10 translucent); 1 film (1 translucent)	234	225 fibers (215 translucent, 1 transparent, 2 red, 7 black); 7 fragments (7 translucent); 2 films (1 transparent, 1 blue)
E(replicate 1)#	3	2 fibers (1 yellow, 1 black), 1 fragment (1 translucent)	341	341 fibers (315 translucent, 14 transparent, 1 red, 6 blue, 4 black, 1 white)
E(replicate 2)#	2	2 fibers (1 transparent, 1 blue)	176	176 fibers (159 translucent, 8 blue, 4 brown, 5 black)
E(replicate 3)*	1	1 fiber (1 translucent)	58	58 fibers (54 translucent, 2 blue, 1 brown, 1 black)
Site 8: Brule River Beach				
C#	0		121	121 fibers (114 translucent, 3 red, 3 blue, 1 yellow)
E#	1	1 fiber (1 translucent)	88	88 fibers (87 translucent, 1 transparent)

Site 9: Long Island SW A#	0		115	115 fibers (106 translucent, 5 blue, 2 brown, 2 black)
C	2	2 fibers (2 translucent)	129	128 fibers (118 translucent, 3 blue, 7 black), 1 fragment (1 blue)
E	0		52	52 fibers (46 translucent, 1 red, 2 blue, 2 brown, 1 black)
Field blank	0		10	10 fibers (8 translucent, 1 blue, 1 black)
Site 10: Knife River Beach A	0		298	298 fibers (294 translucent, 2 blue, 2 black)
C	0		247	247 fibers (247 translucent)
E	0		382	382 fibers (380 translucent, 1 blue, 1 black)
Field blank	0		8	8 fibers (4 translucent, 1 red, 1 blue, 2 black)
Site 11: Park Point A	1	1 fiber (1 transparent)	379	379 fibers (376 translucent, 3 blue)
C	1	1 fiber (1 white)	526	526 fibers (518 translucent, 2 red, 1 blue, 5 black)
E	1	1 fiber (blue)	371	370 fibers (363 translucent, 3 red, 1 blue, 2 black), 1 fragment (1 translucent)
Field blank	0		18	18 fibers (14 translucent, 2 blue, 2 black)
Site 12: Ontonagon A	0		60	60 fibers (57 translucent, 1 red, 1 blue, 1 black)
C	0		70	69 fibers (65 translucent, 1 blue, 1 red, 1 black, 1 brown), 1 fragment (1 red)
E	0		60	60 fibers (57 translucent, 1 red, 1 blue, 1 brown)

Table A4. Plastic and nonplastic particles found within field replicates, field blanks, and counting blanks for the July 2018 sampling campaign. In terms of morphology in this table, fibers includes both individual fibers and fibrous bundles. Field data are not blank corrected..

Note: \*oxidized prior to microscopy; #replicate 200 g aliquots from the same sample jar.

	Plastics/200g dry weight sample	Plastic morphology and color	Nonplastic microlitter/ 200g dry weight sample	Non plastic morphology and color
Site 1: York 1(replicate 1)*	0		5	5 fibers (2 translucent, 3 black)
1(replicate 2)*	2	2 fibers (2 transparent)	8	8 fibers (7 translucent, 1 black)
Site 3: Outer Island 1*	1	1 fiber (blue)	19	18 fibers (12 translucent, 2 transparent, 2 blue, 1 brown, 1 green), 1 fragment (1 yellow)
A	0		135	135 fibers (130 translucent, 5 transparent)
C*	1	1 other (blue)	96	91 fibers (89 translucent, 2 blue), 3 fragments (1 translucent, 1 blue, 1 green), 2 films (1 transparent, 1 green)
E	0		54	53 fibers (43 translucent, 3 transparent, 2 blue, 5 black), 1 fragment (1 translucent)
4*	4	4 fragments (3 blue, 1 yellow)	69	68 fibers (64 translucent, 2 blue, 1 brown, 1 black), 1 fragment (1 yellow)
Field blank	0		15	15 fibers (13 translucent, 2 black)
Site 7: Meyers Beach 1*	0		9	8 fibers (6 translucent, 1 blue, 1 black), 1 fragment (1 black)

A	1	1 bead (1 transparent)	69	68 fibers (63 translucent, 2 blue, 1 red, 2 black), 1 film (1 white)
C(replicate 1)	1	1 fiber (1 black)	69	69 fibers (61 translucent, 1 transparent, 5 blue, 1 yellow, 1 brown)
C(replicate 2)*	2	2 fragments (1 yellow, 1 translucent)	40	36 fibers (31 translucent, 3 blue, 2 black), 4 fragments (2 blue, 2 white)
E	5	4 fibers (2 black, 2 green), 1 fragment (1 blue)	225	225 fibers (212 translucent, 11 blue, 1 red, 1 black)
4(replicate 1)*	7	7 fibers (1 black, 1 yellow, 1 red, 3 translucent, 1 blue)	85	85 fibers (82 translucent, 3 blue)
4(replicate 2)*	1	1 fiber (1 yellow)	12	10 fibers (1 red, 2 yellow, 2 translucent, 2 transparent, 1 black, 1 white, 1 blue), 1 foam (white), 1 film (yellow)
Field blank	0		5	5 fibers (4 translucent, 1 black)
Counting blanks (n=10) Average counting blank	0.3		5	
Maximum	1		14	
Minimum	0		0	
Method blank*	0		10	7 fibers (2 transparent, 3 red, 2 blue), 3 films (white)

Table A5. The abundance, morphology, and color of microplastic and non-plastic low-density particles from surface water net tows in the Apostle Islands. \* indicates oxidized prior to microscopy; # indicates collected on Nylon 250 µm filter; na-not applicable. For comparison the ranges of microplastics found at non-APIS sites (Eriksen et al., 2013; Hendrickson et al., 2018a) are also included.

Site	Length of tow (m)	Microplastics # per surface area and # per volume	Plastic morphology and color	Non-plastic microlitter # per surface area and # per volume	Non-plastic morphology and color
MB*# (May 21, 2018)	871	<b>9000/km<sup>2</sup></b> <i>0.07/m<sup>3</sup></i>	2 fragments ( 1 blue, 1 pink), 3 fibers (1 pink, 1 blue, 1 green), 1 foam (yellow), 1 other (blue)	<b>4000/km<sup>2</sup></b> <i>0.03/m<sup>3</sup></i>	2 fragments (1 white, 1 blue), 1 film (brown)
LI * (May 21, 2018)	875	<b>11,000/km<sup>2</sup></b> <i>0.08/m<sup>3</sup></i>	7 fibers (3 pink, 4 blue), 1 film (1 black)	<b>9000/km<sup>2</sup></b> <i>0.07/m<sup>3</sup></i>	6 fragments (3 tan, 2 white, 1 black), 1 other (1 brown)
MB* (July 25, 2018)	440	<b>40,000/km<sup>2</sup></b> <i>0.29/m<sup>3</sup></i>	2 fragments (1 white, 1 grey), 11 fibers (5 pink, 3 blue, 3 purple), 1 film (brown), 1 foam (white)	<b>27,000/km<sup>2</sup></b> <i>0.19/m<sup>3</sup></i>	5 fragments (1 white, 3 brown, 1 grey), 2 fibers (1 brown, 1 black) 1 film (blue), 2 spheres (1 white, 1 black)
LI*# (July 25, 2018)	388	<b>18,000/km<sup>2</sup></b> <i>0.13/m<sup>3</sup></i>	1 fragment (1 white), 4 fibers (3 blue, 1 black), 1 film (1 brown)	<b>12,000/km<sup>2</sup></b> <i>0.09/m<sup>3</sup></i>	2 fragments (1 brown, one black), 2 films (1 white/black, 1 brown)
ST* (July 25, 2018)	846	<b>19,000/km<sup>2</sup></b> <i>0.14 /m<sup>3</sup></i>	10 fibers (3 pink, 1 transparent, 2 purple, 1 white, 2 blue, 1 silver). 4 films (4 white)	<b>8000/km<sup>2</sup></b> <i>0.06/m<sup>3</sup></i>	1 fragment (white), 4 fibers (3 transparent, 1 white).
Counting blanks (5)#	na	No plastics found	na	0.2 (average per filter)	1 fragment (black)
Western Lake Superior <sup>1*</sup>	478-1916	Below detection limit to 81,000/km <sup>2</sup> (mean: <b>37,000 km<sup>2</sup></b> )	39 % fiber, 34 % fragment, 21 % film	Presence reported (cotton fibers). Total not enumerated.	Not reported
Eastern Lake Superior <sup>2</sup>	1940-3850	1300-13,000/km <sup>2</sup> (mean: <b>5400 km<sup>2</sup></b> )	Not reported for Lake Superior	Presence reported (e.g., coal ash and coal fly ash). Total not enumerated	Not reported

<sup>1</sup>Hendrickson et al , 2018; note: includes fibers

<sup>2</sup>Eriksen et al., 2013; note: does not include fibers



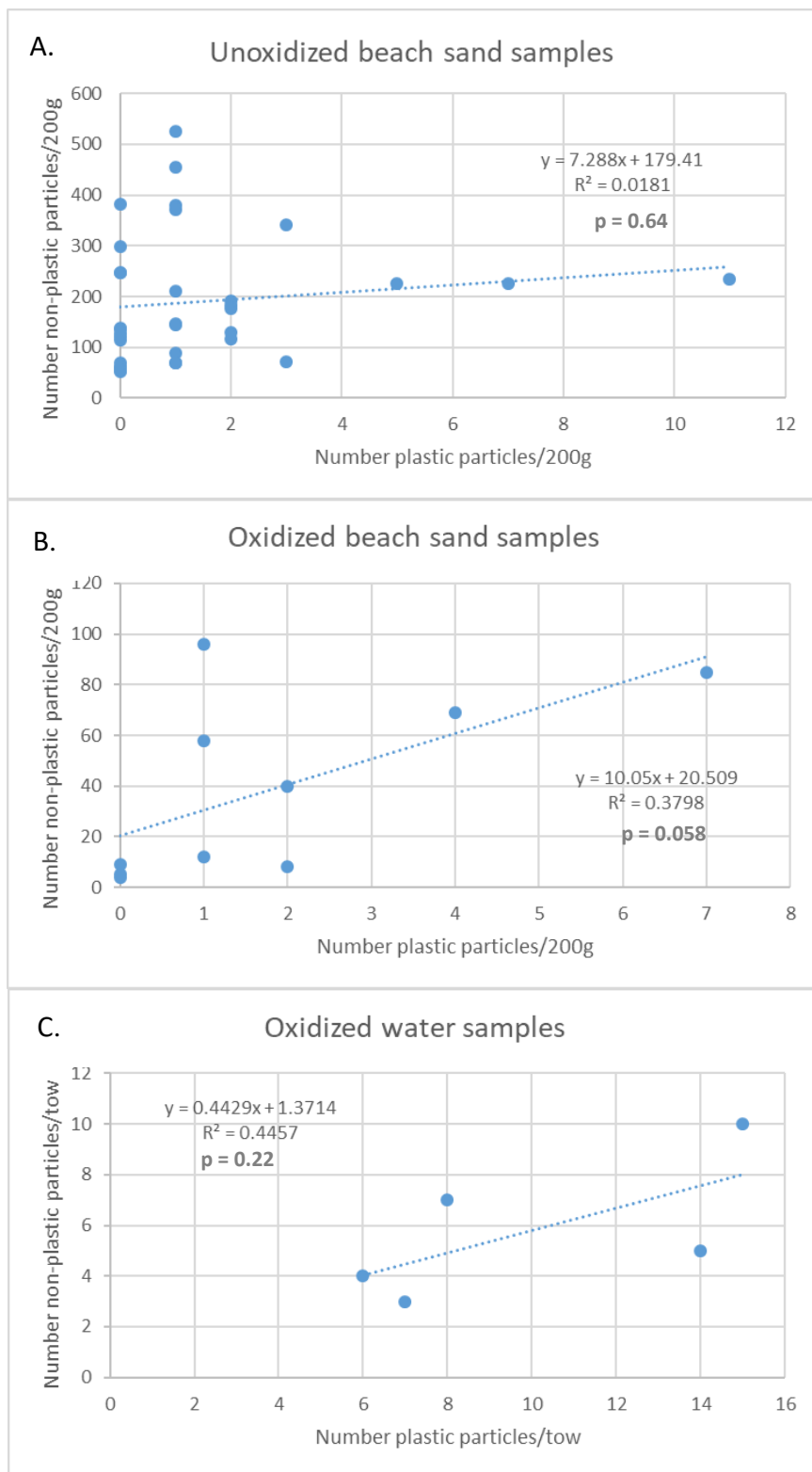
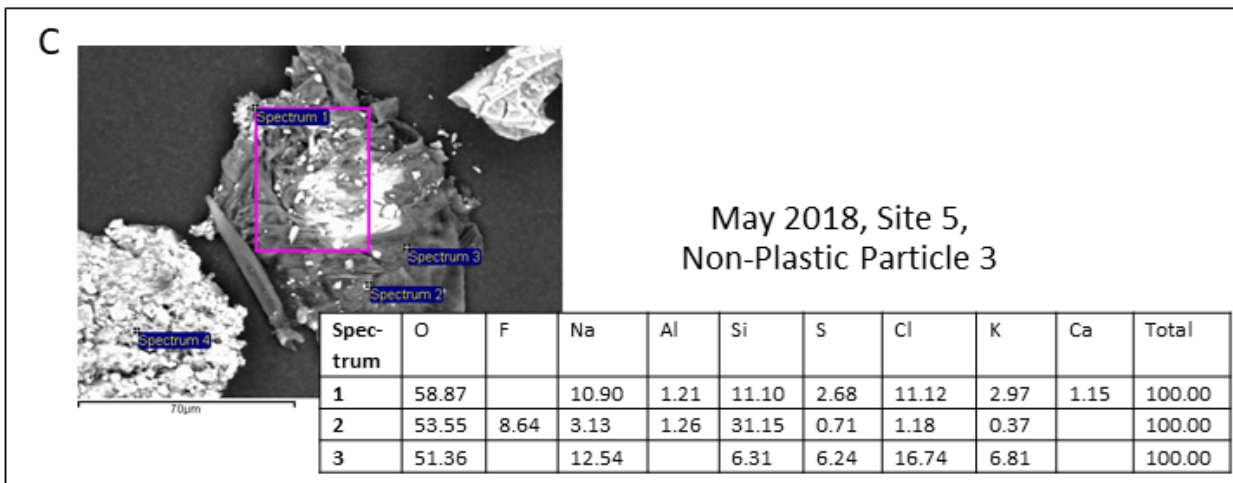
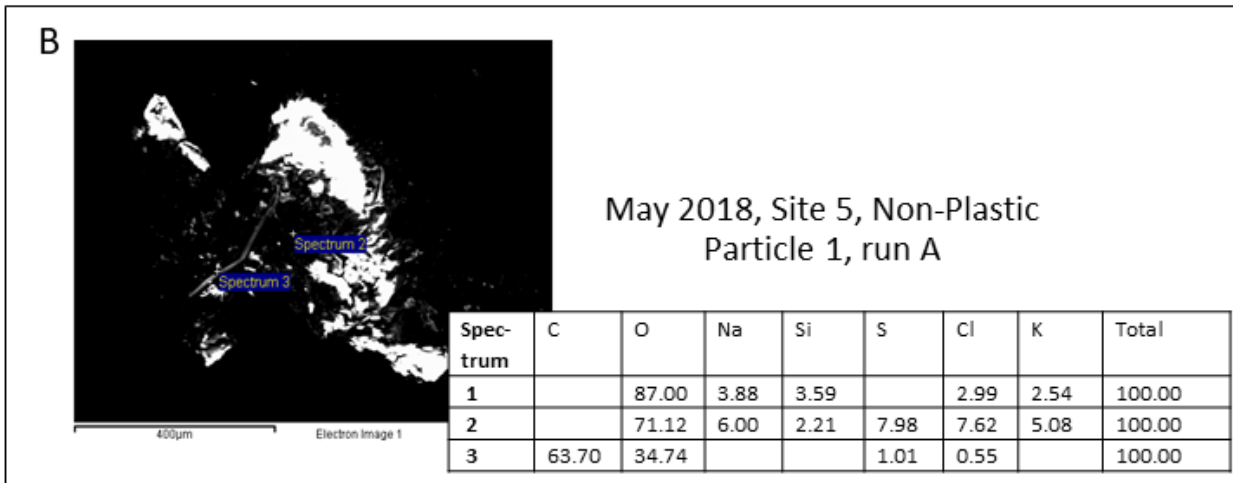
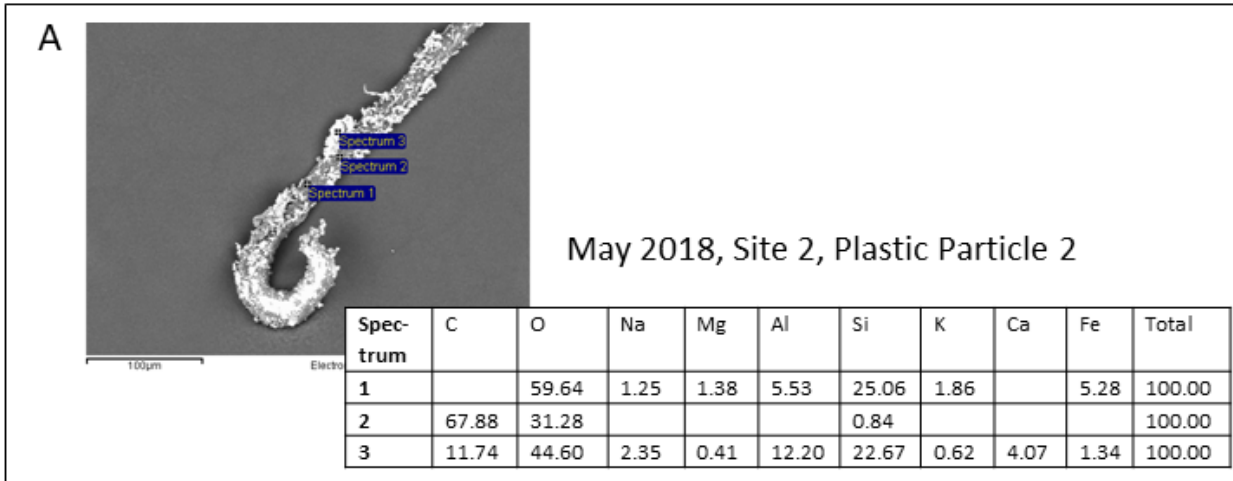
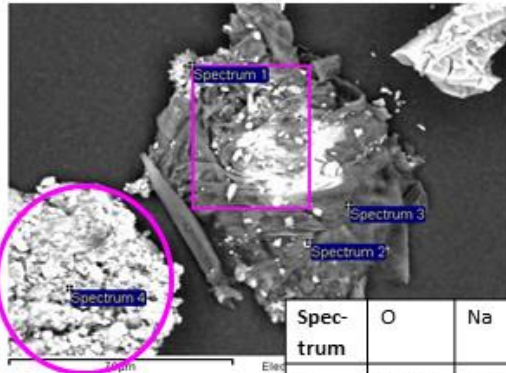


Fig. A1. Type 1 linear regression of A. unoxidized sand samples; B. oxidized sand samples, and C: oxidized water samples. The number of plastic particles is the explanatory variable and the number of non-plastic microlitter particles is the dependent variable.



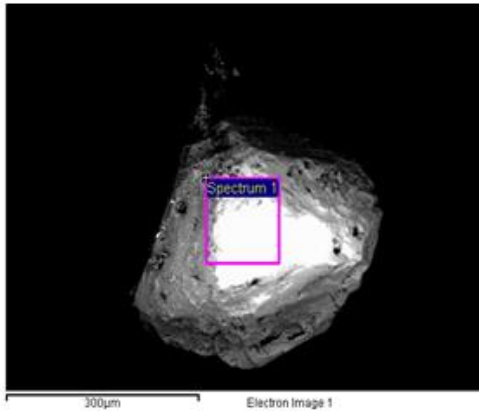
D



May 2018, Site 5,  
Non-Plastic Particle 4

Spectrum	O	Na	Mg	Al	Si	P	Cl	K	Fe	Total
4	46.72	5.66	0.55	10.48	16.51	0.58	4.65	1.07	13.77	100.00

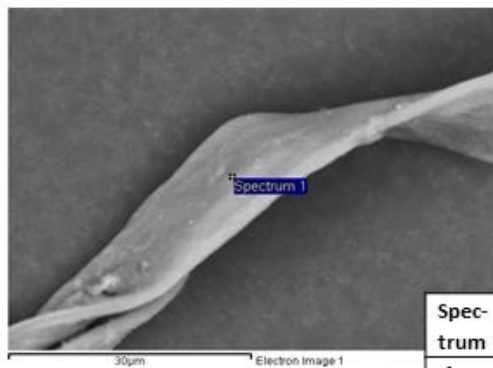
E



May 2018, Site 5,  
Non-Plastic Particle 5

Spectrum	O	Si	Total
1	64.30	35.70	100.00

F



May 2018, Site 5,  
Non-Plastic Particle 6

Spectrum	O	Na	Si	S	Cl	K	Ca	Total
1	68.46	7.50	2.02	5.54	8.85	6.03	1.60	100.00

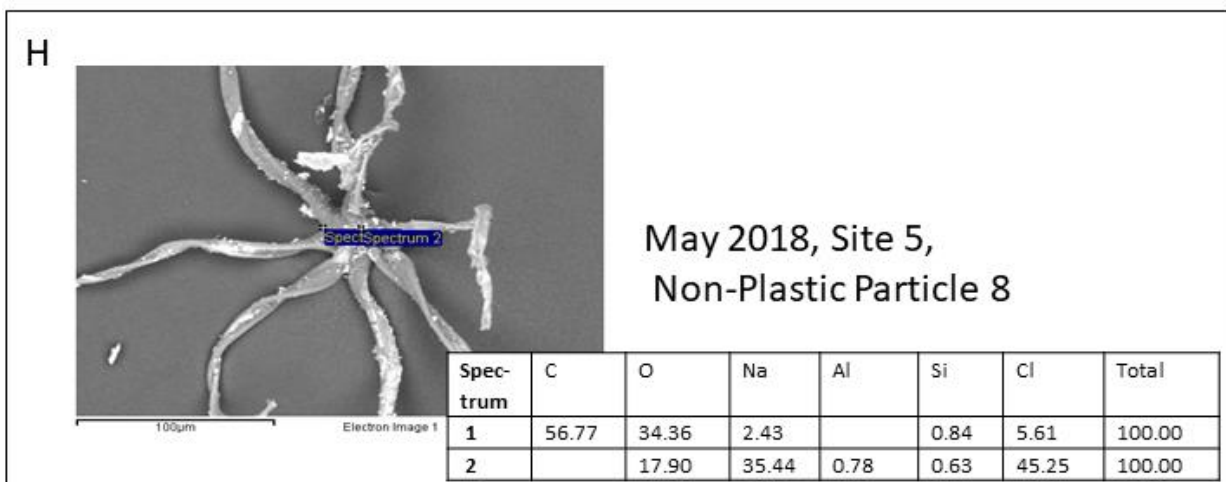
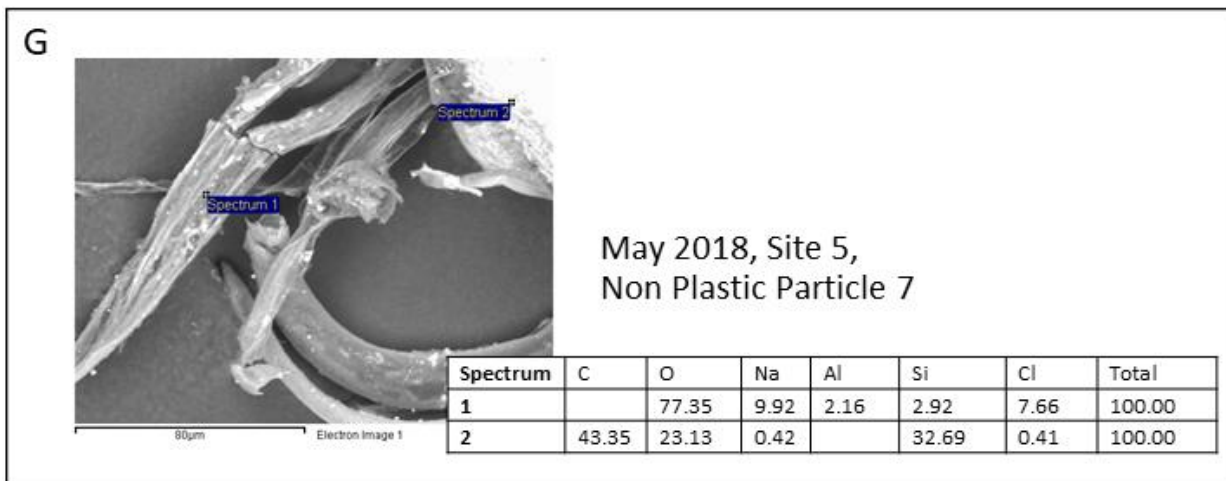


Fig A2. SEM-EDS data for (A) a particle positive for plastic via melt test and (B-H) particles deemed non-plastic via melt test. Images show the spectrum locations on each particle for the EDS spectral data in the overlaid table.

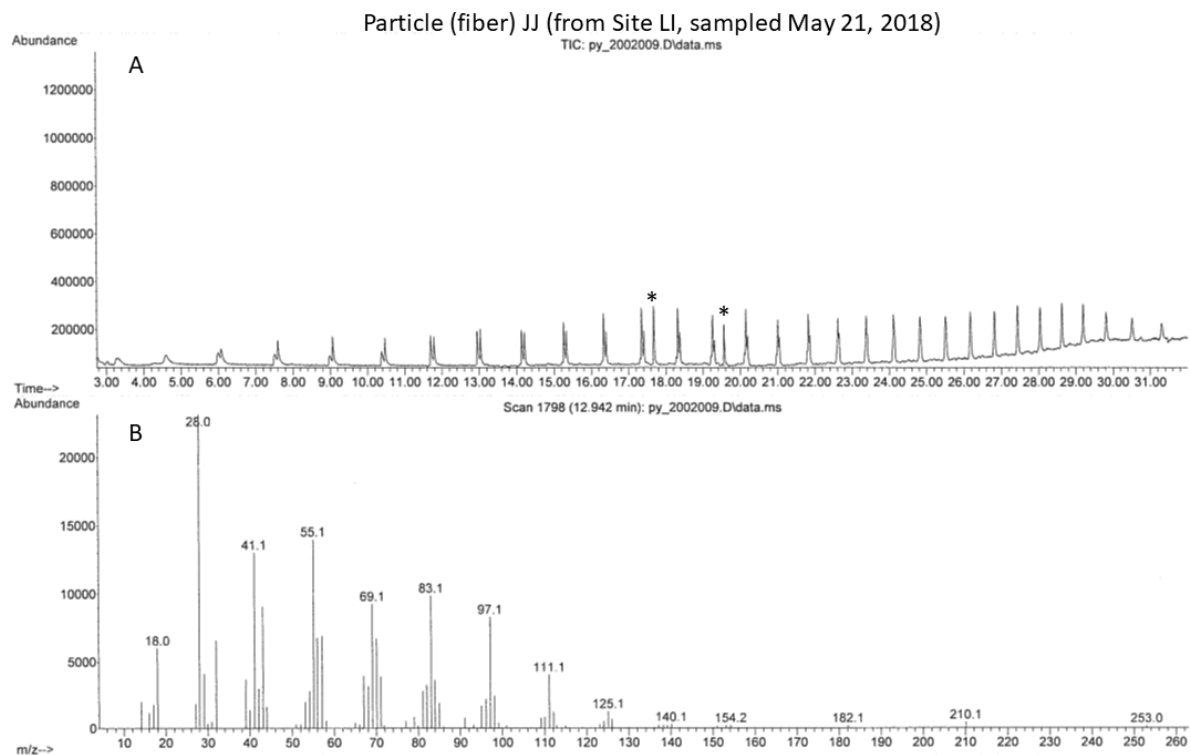


Fig. A3. PyGCMS analysis of a fiber from surface water (Site LI, sampled May 21, 2018). The fiber was identified as polyethylene. The total ion chromatogram (TIC, shown in part A) exhibits the doublet to triplet peaks described in Hendrickson et al. (2018) and individual mass spectra for peaks show the expected alkene and alkane signatures (as for the peak at 12.9 minutes in part B). Using the F-search library (Frontier Laboratories, Ltd) on the mass spectrum summed across the entire TIC leads to a match with HDPE (96% match). Note: asterisks indicate peaks from glove contamination as identified by tube blanks handled by the same mass spectrometry operator (see also Fig. A6).

Particle (fiber) MM from site LI (May 21, 2018 sampling)

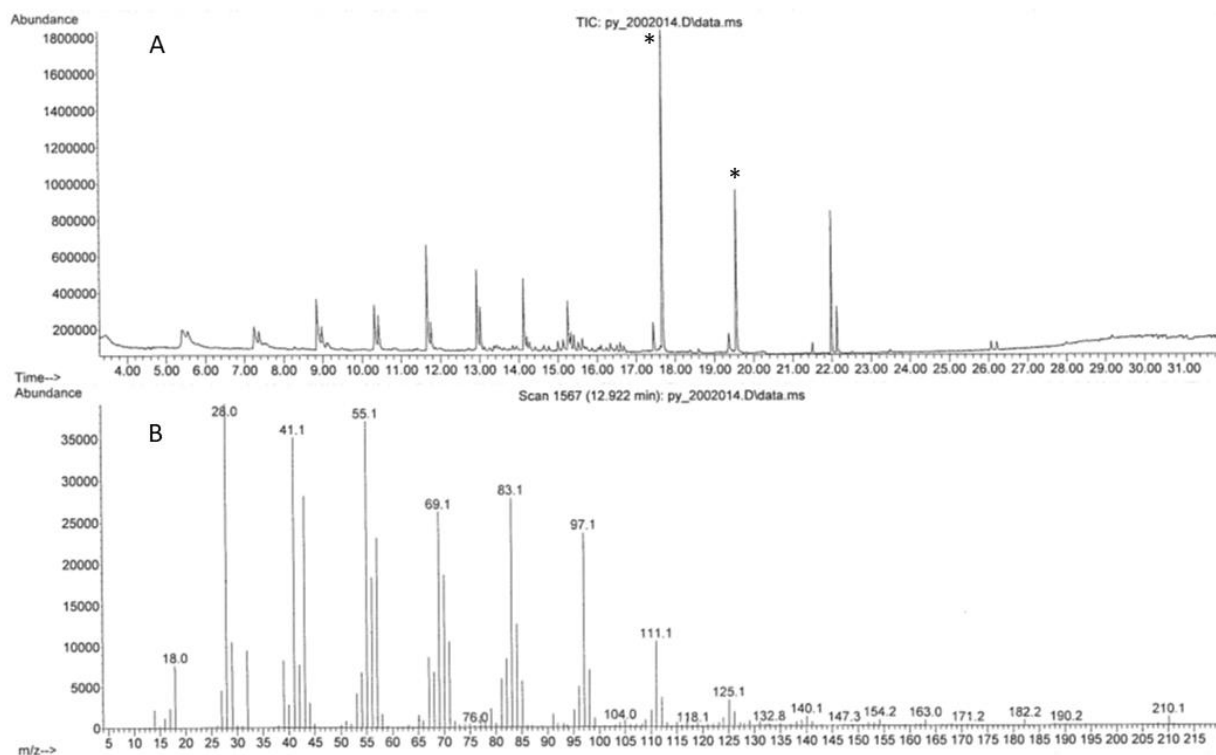


Fig. A4. PyGCMS analysis of a fiber particle from surface water at site LI (May 21, 2018 sampling). This particle was tentatively identified as polyethylene based upon doublet character in the TIC (part A) and by F-search library, where a search on the TIC range 3.3 to 17 minutes yielded a match with HDPE (91%). Part B shows the alkene mass spectrum for the peak at 12.9 minutes. Note that in addition to the polyethylene character there are two additional uncharacterized peaks at 21.97 minutes (main m/z values 301, 316, 91, 105) and 22.12 minutes (main m/z values 91, 316, 223). Note: asterisks indicate peaks from glove contamination as identified by tube blanks handled by the same mass spectrometry operator (see also Fig. A6).

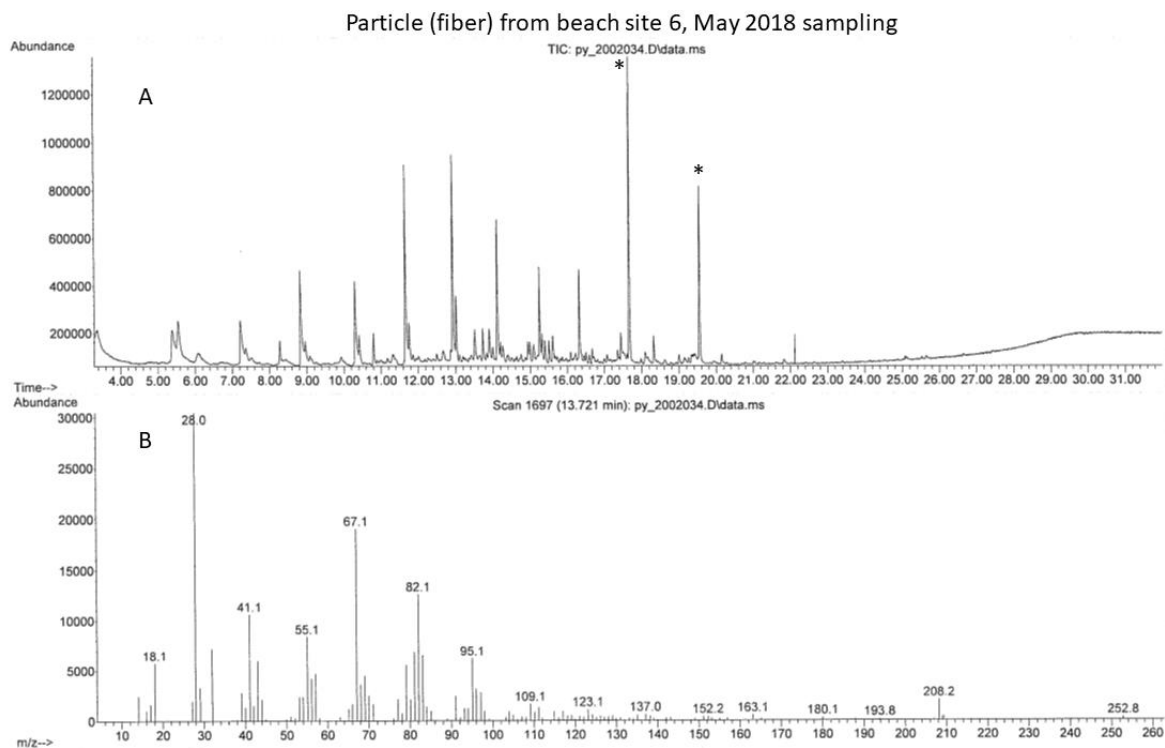


Fig. A5. PyGCMS analysis of a fiber from beach site 6 (sampled in May 2018) and exhibiting a mixed polyethylene/polypropylene character (e.g. TIC in Part A and the diene-type mass spectrum (e.g.,  $m/z$  67, 81, 95, 109, 123, 137, as described in (McLafferty and Turacek, 1993)) for the peak at 13.7 minutes in Part B).

Note: asterisks indicate peaks from glove contamination as identified by tube blanks handled by the same mass spectrometry operator (see also Fig. A6).

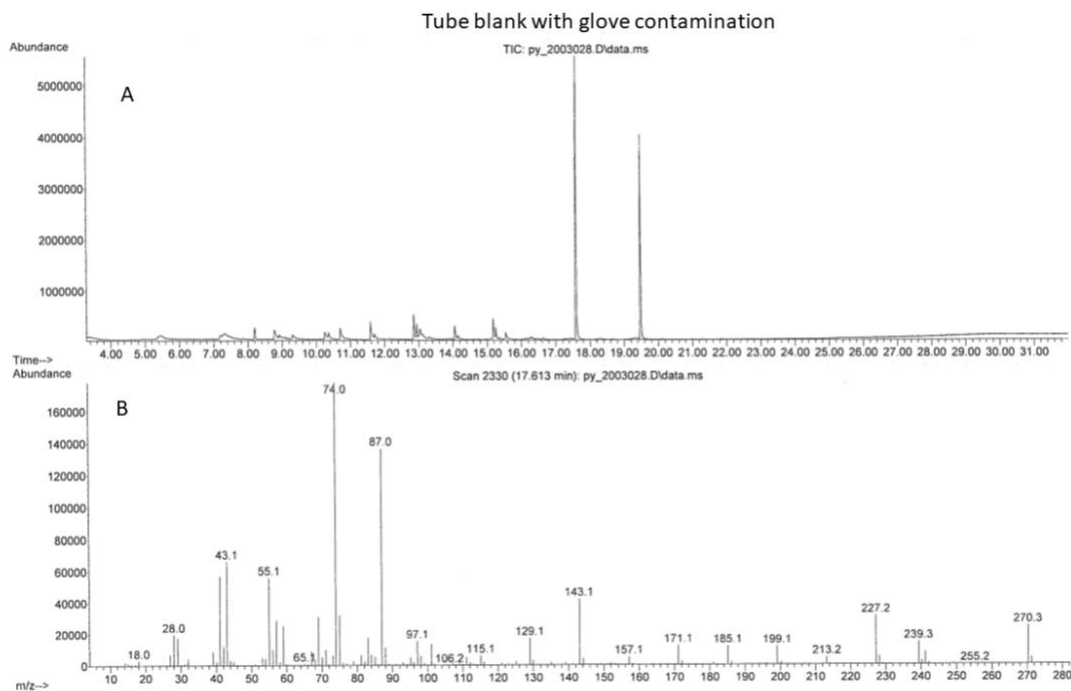


Fig. A6. PyGCMS analysis of a sampling tube blank handled with direct glove contact with purple nitrile gloves. Note that additional tube blanks without glove contact give a very low signal response with essentially no signal (and no peaks) across the chromatogram from 3 minute to 26 minutes. Part A shows the total ion chromatogram, which is dominated by C16:0 and C18:0 fatty acid methyl esters. Part B shows the mass spectrum for the peak at 17.6 minutes, which was identified as the C16:0 fatty acid methyl ester via NIST library search.

## References:

- Hendrickson, E., Minor, E.C., Schreiner, K., 2018. Microplastic abundance and composition in western Lake Superior as determined via microscopy, Pyr-GC/MS, and FTIR. *Environ. Sci. Technol.* 52, 1787–1796. <https://doi.org/10.1021/acs.est.7b05829>
- McLafferty, F.W., Turacek, F., 1993. *Interpretation of Mass Spectra*, 4th ed. University Science Books, Mill Valley, CA.