Supporting Information "**A**" for

# Long-term phosphorus mass-balance of Lake Erie (Canada-USA) reveals a major contribution of in-lake phosphorus loading

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**Contents of this file** (32 pages with 10 Tables, 1 Figure, and 7 Appendices):

**Table A1**. Literature derived estimates of mean annual internal phosphorus load in metric tons (MT) in lakes St. Clair and Erie scaled up to the basin or whole lake areas. ID #1-5 indicate annual fluxes of gross internal load, while ID #6-8 indicate annual net internal load.

| ID # | Study Period | Lake St. Clair (LSC) | Lake Erie (LE) | | | | LES (LSC +LE) | Data Sources | Method |
|---|---|---|---|---|---|---|---|---|---|
| | | | Western Basin | Central Basin | Eastern Basin | Whole Lake Erie | | | |
| 1 | P1 | - | 378 to 808 | - | - | - | - | S1 | M1, M2 |
| 2 | P2 | - | - | 300 to 1250 | - | - | - | S2 | M2 |
| 3 | P3 | - | - | 2400<br>range: 0 to 4229<br>mean: 1872 | - | - | - | S3 | M3 |
| 4 | P4 | - | 415* | - | - | - | - | S4 | M1 |
| 5 | P5 | - | - | 10599 | 359 | - | - | S5 | M4 |
| 6 | P6 | - | - | 10000-11000 | - | - | - | S6 | M5 |
| 7 | P7 | 220 | 495 | 795 | 2273 | 3563 | 3783 | S7 | M6 (this study) |
| 8 | P7 | 219 | 479 | 763 | 2288 | 3530 | 3749 | S7 | M7 (this study) |
| 9 | P7 | 220 | 480 | 706 | 2268 | 3454 | 3674 | S7 | M8 (this study) |

* Just for a four-day episode only (neither annul nor seasonal load).

Study period: P1, 2013: Jun 24 & Jul 6, 2014: Jun 27 & Aug 4; P2, spring and summer of 2010 and 2012; P3, August-September of 1985 to 2012; P4, four-day episode in summer 2014; P5, long-term: analysis of *in-situ* collected sediment cores (30 cm long); P6, continuous in-situ nutrient analyzer measurements at 2 sites in summer 2019 and then scaling up to the basin size; P7, Oct 1, 2003 to Sep 30, 2016.

Data sources: S1, Matisoff et al. (2016); S2, Paytan et al. (2017); S3, Nürnberg et al. (2019); S4, Gibbons & Bridgeman (2020); S5, Wang et al. (2021); S6, Anderson et al. (2021); S7, this study.

Methods: M1, laboratory incubations scaled up to the basin size; M2, laboratory incubations of sediment cores scaled up to the basin size; M3, as a product of the estimated annual hypoxic factor (HF) values and a phosphorus release rate; M4, *in-situ* core profile analysis; M5, continuous *in-situ* nutrient analyzer measurements at 2 sites and then scaling up to the basin size; M6, Net Mass Balance Approach (NMBA) using steady state formulations (no change in basin concentrations between 2003 and 2016); M7, NMBA using non-steady state formulations (change in basin concentration ≠ 0 and based on averages between April and August concentrations with difference determined between 2003 and 2016); M8, NMBA using non-steady state formulations (change in basin concentration ≠ 0 and is based on April concentrations with the difference determined between two 3-year periods 2003-2005 and 2014-2016).

**Table A2**. Characterization of Lake Erie Quaternary Watersheds located in Canadian part of Lake Erie system.

| # | Quaternary Watersheds | | | | Tertiary Watersheds | | | |
|---|---|---|---|---|---|---|---|---|
| | ID # | Name | Drainage Area ($km^2$) | Receiving Waterbody* | ID # | Name | Latitude Longitude | Area ($km^2$) |
| 1. | 02GG-01 | Upper St Clair River | 186 | SCR | 02GG | Sydenham | 42.7658 82.0565 | 3496 |
| 2. | 02GG-02 | Lower St Clair River | 316 | SCR | | | | |
| 3. | 02GG-03 | Lower Lake St. Clair Tributaries | 88 | LSC | | | | |
| 4. | 02GG-04 | Upper Lake St. Clair Tributaries | 230 | LSC | | | | |
| 5. | 02GG-05 | East Sydenham River | 1505 | LSC | | | | |
| 6. | 02GG-06 | Lower N. Sydenham River | 550 | LSC | | | | |
| 7. | 02GG-07 | Bear Creek | 621 | LSC | | | | |
| 8. | 02GD-01 | S. Thames River | 764 | LSC | 02GD | Upper Thames | 43.2121 81.0424 | 3057 |
| 9. | 02GD-02 | Waubuno Creek | 135 | LSC | | | | |
| 10. | 02GD-03 | Middle Thames River | 294 | LSC | | | | |
| 11. | 02GD-04 | Reynolds Creek | 166 | LSC | | | | |
| 12. | 02GD-05 | N. Thames River | 734 | LSC | | | | |
| 13. | 02GD-06 | Medway River | 202 | LSC | | | | |
| 14. | 02GD-07 | Fish - Flat Creeks | 154 | LSC | | | | |
| 15. | 02GD-08 | Trout Creek | 173 | LSC | | | | |
| 16. | 02GD-09 | Avon River | 163 | LSC | | | | |
| 17. | 02GD-10 | Black Creek | 142 | LSC | | | | |
| 18. | 02GD-11 | Whirl Creek | 130 | LSC | | | | |
| 19. | 02GE-01 | Lower Thames River | 1607 | LSC | 02GE | Lower Thames | 42.5548 81.8790 | 2818 |
| 20. | 02GE-02 | Big Creek | 251 | LSC | | | | |
| 21. | 02GE-03 | Jeanette Creek | 416 | LSC | | | | |
| 22. | 02GE-04 | McGregor Creek | 285 | LSC | | | | |
| 23. | 02GE-05 | Oxbow Creek | 89 | LSC | | | | |
| 24. | 02GE-06 | Dingham Creek | 170 | LSC | | | | |

*SCR, St. Clair River; LSC, Lake St. Clair; DR, Detroit River; WB, western basin; CB, central basin; EB, eastern basin.

**Table A2**. (*Continued*)

| # | Quaternary Watersheds | | | | Tertiary Watersheds | | | |
|---|---|---|---|---|---|---|---|---|
| | ID # | Name | Drainage Area (km$^2$) | Receiving Waterbody* | ID # | Name | Latitude Longitude | Area (km$^2$) |
| 25. | 02GH-01 | Pike Crk - Puce River | 270 | LSC | 02GH | Cedar | 42.1565 82.7822 | 1765 |
| 26. | 02GH-02 | Ruscom River | 210 | LSC | | | | |
| 27. | 02GH-03 | Belle River | 150 | LSC | | | | |
| 28. | 02GH-04 | Windsor Area - Little River | 116 | DR | | | | |
| 29. | 02GH-05 | River Canard | 347 | DR | | | | |
| 30. | 02GH-06 | Lower Detroit River | 113 | WB | | | | |
| 31. | 02GH-07 | Cedar Creek - Oxley - Seacliffe | 228 | WB | | | | |
| 32. | 02GH-08 | Sturgeon Point - Point Pelee | 79 | WB | | | | |
| 33. | 02GH-09 | Hillman - Lebo Creeks | 206 | CB | | | | |
| 34. | 02GH-10 | Pelee Island | 41 | WB | | | | |
| 35. | 02GH-11 | Fighting Island | 5 | DR | | | | |
| 36. | 02GF-01 | Renwick - Erie Beach | 46 | CB | 02GF | Rondeau | 42.4741 81.7300 | 770 |
| 37. | 02GF-02 | Flat Creek - Rondeau | 115 | CB | | | | |
| 38. | 02GF-03 | Morpeth - Palmyra Beach | 99 | CB | | | | |
| 39. | 02GF-04 | Brock's Creek | 213 | CB | | | | |
| 40. | 02GF-05 | Tyconnel Beach | 101 | CB | | | | |
| 41. | 02GF-06 | Talbot Creek | 196 | CB | | | | |
| 42. | 02GC-01 | Stoney Creek | 123 | EB | 02GC | Big Creek | 42.8223 80.6028 | 3992 |
| 43. | 02GC-02 | Kettle Creek | 463 | CB | | | | |
| 44. | 02GC-03 | Catfish Creek | 417 | CB | | | | |
| 45. | 02GC-04 | Big Otter Creek | 817 | CB | | | | |
| 46. | 02GC-05 | Long Point | 220 | EB | | | | |
| 47. | 02GC-06 | South Otter - Clear Creeks | 37 | CB | | | | |
| 48. | 02GC-07 | Diedrich - Young Crks | 249 | EB | | | | |
| 49. | 02GC-08 | Big Creek | 741 | EB | | | | |

*SCR, St. Clair River; LSC, Lake St. Clair; DR, Detroit River; WB, western basin; CB, central basin; EB, eastern basin.

**Table A2**. (*Continued*)

| # | Quaternary Watersheds | | | | Tertiary Watersheds | | | |
|---|---|---|---|---|---|---|---|---|
| | ID # | Name | Drainage Area ($km^2$) | Receiving Waterbody* | ID # | Name | Latitude Longitude | Area ($km^2$) |
| 50. | 02GC-09 | Lynn River | 301 | EB | | | | |
| 51. | 02GC-11 | Nanticoke Creek | 216 | EB | | | | |
| 52. | 02GC-12 | Sandusk Creek | 173 | EB | | | | |
| 53. | 02GC-13 | Welland River | 143 | EB | | | | |
| 54. | 02GA-01 | Upper Grand River | 1903 | EB | 02GA | Upper Grand | 43.6352 80.4700 | 4745 |
| 55. | 02GA-02 | Nith River | 1131 | EB | | | | |
| 56. | 02GA-03 | Galt Creek | 108 | EB | | | | |
| 57. | 02GA-04 | Speed River | 505 | EB | | | | |
| 58. | 02GA-05 | Eramosa River | 274 | EB | | | | |
| 59. | 02GA-06 | Conestogo River | 824 | EB | | | | |
| 60. | 02GB-01 | Lower Grad River | 684 | EB | 02GB | Lower Grand | 43.1122 80.1908 | 2030 |
| 61. | 02GB-02 | McKenzie River | 371 | EB | | | | |
| 62. | 02GB-03 | Big Creek | 166 | EB | | | | |
| 63. | 02GB-04 | Fairchield Creek | 412 | EB | | | | |
| 64. | 02GB-05 | Horner Creek | 397 | EB | | | | |
| 65. | 02HA-05 | Upper Welland Canal | 99 | EB | 02HA | Niagara River | 43.0581 79.4324 | 1407 |
| 65. | 02HA-07 | Welland River | 1118 | NR | | | | |
| 67. | 02HA-08 | Fort Erie Creeks | 190 | NR, EB | | | | |
| | | TOTAL for St. Clair River | 502 | | | | | |
| | | TOTAL for Lake St. Clair | 9499 | | | | | |
| | | TOTAL for Detroit River | 468 | | | | | |
| | | TOTAL for Lake Erie West Basin | 431 | | | | | |
| | | TOTAL for Lake Erie Central Basin | 2710 | | | | | |
| | | TOTAL for Lake Erie East Basin | 9033 | | | | | |
| | | GRAND TOTAL | 1407 | | | | | |

*SCR, St. Clair River; LSC, Lake St. Clair; DR, Detroit River; WB, western basin; CB, central basin; EB, eastern basin.

**Table A3**. Characterization of Lake Erie system watersheds located in United States and based on the 6-digit and 8-digit hydrological unit codes (HUC6 and HUC8).

| # | Hydrologic Unit Code 8-digit (HUC8) | | | | Hydrologic Unit Code 6-digit (HUC6) | | |
|---|---|---|---|---|---|---|---|
| | Code | Name | State | Watershed Area ($km^2$) | Code | Name | Watershed Area ($km^2$) |
| 1. | 04090001 | St. Clair | MI | 2997.0 | 040900 | St Clair - Detroit | |
| 2. | 04090002 | Lake St. Clair | MI | 662.8 | | | |
| 3. | 04090003 | Clinton | MI | 2064.4 | | | |
| 4. | 04090004 | Detroit | MI | 1518.3 | | | |
| 5. | 04090005 | Huron | MI | 2378.3 | | | |
| 6. | 04100001 | Ottawa-Stony | MI | 1806.3 | 041100 | Western Lake Erie | |
| 7. | 04100002 | Raisin | MI | 2753.3 | | | |
| 8. | 04100003 | St. Joseph | OH | 2832.3 | | | |
| 9. | 04100004 | St. Marys | OH | 2054.8 | | | |
| 10. | 04100005 | Upper Maumee | OH | 1003.3 | | | |
| 11. | 04100006 | Tiffin | OH | 2014.0 | | | |
| 12. | 04100007 | Auglaize | OH | 4316.1 | | | |
| 13. | 04100008 | Blanchard | OH | 1999.4 | | | |
| 14. | 04100009 | Lower Maumee | OH | 2798.6 | | | |
| 15. | 04100010 | Cedar-Portage | OH | 2510.3 | | | |
| 16. | 04100011 | Sandusky | OH | 4732.6 | | | |
| 17. | 04100012 | Huron-Vermilion | OH | 1979.8 | | | |

**Table A3**. (*Continued*)

| # | Hydrologic Unit Code 8-digit (HUC8) | | | | Hydrologic Unit Code 6-digit (HUC6) | | |
|---|---|---|---|---|---|---|---|
| | Code | Name | State | Watershed Area ($km^2$) | Code | Name | Watershed Area ($km^2$) |
| 18. | 04110001 | Black-Rocky | OH | 2324.7 | 041000 | Southern Lake Erie | |
| 19. | 04110002 | Cuyahoga | OH | 2101.5 | | | |
| 20. | 04110003 | Ashtabula-Chagrin | OH | 1641.5 | | | |
| 21. | 04110003 | Ashtabula-Chagrin | PA | 156.6 | | | |
| 22. | 04110004 | Grand | OH | 1827.5 | | | |
| 23. | 04120101 | Chautauqua-Conneaut | OH | 350.9 | 041201 | Eastern Lake Erie | |
| 24. | 04120101 | Chautauqua-Conneaut | PA | 1571.5 | | | |
| 25. | 04120101 | Chautauqua-Conneaut | NY | 868.0 | | | |
| 26. | 04120102 | Cattaraugus | NY | 1449.5 | | | |
| 27. | 04120103 | Buffalo-Eighteenmile | NY | 1856.1 | | | |
| 28. | 04120104 | Niagara | NY | 2069.9 | | | |

**Table A4**. Characterization of the water gauging stations in Canadian part of the Lake Erie system.

| # | WSC* Gauge Number | Gauge Name | Geographic Coordinates | | Area ($km^2$) | Tertiary Watershed Codes |
|---|---|---|---|---|---|---|
| | | | Latitude | Longitude | | |
| 1. | 02GG002 | Sydenham River Near Alvinston | 42.8306 | 81.8517 | 701 | 02GG |
| 2. | 02GG003 | Sydenham River At Florence | 42.6506 | 82.0083 | 1150 | |
| 3. | 02GG006 | Bear Creek Near Petrolia | 42.9058 | 82.1189 | 249 | |
| 4. | 02GG009 | Bear Creek Below Brigden | 42.8119 | 82.2983 | 536 | |
| 5. | 02GG010 | St. Clair River At Point Edward | 42.9803 | 82.4022 | n/a | |
| 6. | 02GG011 | St. Clair River At Port Lambton | 42.6572 | 82.5069 | n/a | |
| 7. | 02GG013 | Black Creek Near Bradshaw | 42.7622 | 82.2592 | 213 | |
| 8. | 02GE002 | Thames River At Byron | 42.9625 | 81.3317 | 3080 | 02GE |
| 9. | 02GE003 | Thames River At Thamesville | 42.5447 | 81.9672 | 4370 | |
| 10. | 02GE007 | McGregor Creek Near Chatham | 42.3833 | 82.0950 | 204 | |
| 11. | 02GH002 | Ruscom River Near Ruscom Station | 42.2114 | 82.6289 | 95 | 02GH |
| 12. | 02GH003 | Canard River Near Lukerville | 42.1589 | 83.0189 | 159 | |
| 13. | 02GH005 | Lake St. Clair At Belle River | 42.2961 | 82.7108 | n/a | |
| 14. | 02GH011 | Little River At Windsor | 42.3097 | 82.9283 | 55.3 | |
| 15. | 02GC002 | KETTLE CREEK AT ST. THOMAS | 42.7775 | 81.2139 | 331 | 02GC |
| 16. | 02GC007 | BIG CREEK NEAR WALSINGHAM | 42.6856 | 80.5383 | 567 | |
| 17. | 02GC008 | LYNN RIVER AT SIMCOE | 42.8233 | 80.2894 | 144 | |
| 18. | 02GC010 | BIG OTTER CREEK AT TILLSONBURG | 42.8572 | 80.7233 | 354 | |
| 19. | 02GC014 | YOUNG CREEK NEAR VITTORIA | 42.7656 | 80.2944 | 65.8 | |
| 20. | 02GC018 | CATFISH CREEK NEAR SPARTA | 42.7458 | 81.0569 | 295 | |
| 21. | 02GC021 | VENISON CREEK NEAR WALSINGHAM | 42.6533 | 80.5483 | 68.4 | |
| 22. | 02GC022 | NANTICOKE CREEK AT NANTICOKE | 42.8097 | 80.0761 | 177 | |

*The Water Survey of Canada (WSC)

**Table A4**. (*Continued*)

| # | WSC* Gauge Number | Gauge Name | Geographic Coordinates Latitude | Geographic Coordinates Longitude | Area ($km^2$) | Tertiary Watershed Codes |
|---|---|---|---|---|---|---|
| 22. | 02GC026 | BIG OTTER CREEK NEAR CALTON | 42.7106 | 80.8406 | 665 | 02GC |
| 23. | 02GC029 | KETTLE CREEK ABOVE ST. THOMAS | 42.8350 | 81.1347 | 134 | |
| 24. | 02GC030 | CATFISH CREEK AT AYLMER | 42.7736 | 80.9825 | 127 | |
| 25. | 02GC031 | DODD CREEK BELOW PAYNES MILLS | 42.7872 | 81.2675 | 99.6 | |
| 26. | 02GC036 | SILVER CREEK NEAR GROVESEND | 42.6758 | 80.9533 | 40.3 | |
| 27. | 02GB010 | MCKENZIE CREEK NEAR CALEDONIA | 43.0339 | 79.9497 | 173 | 02GB |
| 28. | 02GA003 | GRAND RIVER AT GALT | 43.3531 | 80.3156 | 3520 | 02GA |
| 29. | 14827 | Grand River at York | 43.0217 | 79.8913 | 6044 | |
| 30. | 02HA019 | WELLAND CANAL DIVERSION FROM LAKE ERIE | 42.9500 | 79.2167 | | 02HA |
| 31. | 02HA003 | NIAGARA RIVER AT QUEENSTON | 43.1569 | 79.0472 | 686000 | |
| 32. | 02HA007 | Welland River Below Caistor Corners | 43.0217 | 79.6178 | 223 | |
| 33. | 02HA013 | NIAGARA RIVER AT FORT ERIE | 42.9303 | 78.9142 | 683000 | |
| 34. | 02HA024 | Oswego Creek At Canboro | 42.9911 | 79.6781 | 83.2 | |

*The Water Survey of Canada (WSC)

**Table A5**. Characterization of water gauging stations in US part of the Lake Erie system.

| # | USGS Gauge Number | Gauge Name (State) | Geographic Coordinates | | Area ($km^2$) | Hydrologic Unit |
|---|---|---|---|---|---|---|
| | | | Latitude | Longitude | | |
| 1. | 04159130 | St. Clair River At Port Huron (MI) | 42°59'13" | 82°25'29" | 576014* | 4090001 |
| 2. | 04159492 | Black River Near Jeddo (MI) | 43°09'03" | 82°37'29" | 1202 | 4090001 |
| 3. | 04159900 | Mill Creek Near Avoca (MI) | 43°03'16" | 82°44'05" | 438 | 4090001 |
| 4. | 04160600 | Belle River At Memphis (MI) | 42°54'03" | 82°46'09" | 391 | 4090001 |
| 5. | 04165500 | Clinton River At Moravian Drive At Mt. Clemens (MI) | 42°35'45" | 82°54'32" | 1901 | 4090003 |
| 6. | 04165710 | Detroit River At Fort Wayne At Detroit (MI) | 42°17'53" | 83°05'34" | 592590* | 4090004 |
| 7. | 04166500 | River Rouge At Detroit (MI) | 42°22'23" | 83°15'17" | 484 | 4090004 |
| 8. | 04167000 | Middle River Rouge Near Garden City (MI) | 42°20'53" | 83°18'42" | 259 | 4090004 |
| 9. | 04168400 | Lower River Rouge At Dearborn (MI) | 42°18'30" | 83°15'10" | 236 | 4090004 |
| 10. | 04168580 | Ecorse River At Dearborn Heights (MI) | 42°16'10" | 83°17'23" | 26 | 4090004 |
| 11. | 04174500 | Huron River At Ann Arbor (MI) | 42°17'13" | 83°44'02" | 1888 | 4090005 |
| 12. | 04174518 | Malletts Creek At Ann Arbor (MI) | 42°15'53" | 83°41'18" | 28 | 4090005 |
| 13. | 04176500 | River Raisin Near Monroe (MI) | 41°57'38" | 83°31'52" | 2699 | 4100002 |
| 14. | 04177000 | Ottawa River At University of Toledo (OH) | 41°39'35" | 83°36'45" | 388 | 4100001 |
| 15. | 04193500 | Maumee River At Waterville (OH) | 41°30'00" | 83°42'46" | 16395 | 4100009 |
| 16. | 04193999 | Wolf Creek At Holland (OH) | 41°36'34" | 83°41'03" | 64 | 4100009 |
| 17. | 04195820 | Portage River Near Elmore (OH) | 41°29'28" | 83°13'29" | 1279 | 4100010 |
| 18. | 04198000 | Sandusky River Near Fremont (OH) | 41°18'28" | 83°09'32 | 3240 | 4100011 |

* including the upper Laurentian Great Lakes.

**Table A5**. (*Continued*)

| # | USGS Gauge Number | Gauge Name (State) | Geographic Coordinates | | Area ($km^2$) | Hydrologic Unit |
|---|---|---|---|---|---|---|
| | | | Latitude | Longitude | | |
| 19. | 04199000 | Huron River At Milan (OH) | 41°18'03" | 82°36'30" | 961 | 4100012 |
| 20. | 04199155 | Old Woman Creek At Berlin Rd Near Huron (OH) | 41°20'54" | 82°30'50" | 57 | 4100012 |
| 21. | 04199500 | Vermilion River Near Vermilion (OH) | 41°22'55" | 82°19'01" | 679 | 4100012 |
| 22. | 04200500 | Black River At Elyria (OH) | 41°22'49" | 82°06'17" | 1026 | 4110001 |
| 23. | 04201500 | Rocky River Near Berea (OH) | 41°24'27" | 81°52'58" | 692 | 4110001 |
| 24. | 04201526 | Abram Creek At Kolthoff Drive At Brook Park (OH ) | 41°23'35" | 81°51'01" | 21 | 4110002 |
| 25. | 04208504 | Cuyahoga River Near Newburgh Heights (OH) | 41°27'45" | 81°40'52" | 2041 | 4110002 |
| 26. | 04208700 | Euclid Creek At Cleveland (OH) | 41°34'56" | 81°33'32" | 60 | 4110003 |
| 27. | 04209000 | Chagrin River At Willoughby (OH) | 41°37'51" | 81°24'13" | 637 | 4110003 |
| 28. | 04212100 | Grand River Near Painesville (OH) | 41°43'08" | 81°13'41" | 1774 | 4110004 |
| 29. | 04213000 | Conneaut Creek At Conneaut (OH) | 41°55'37" | 80°36'15" | 453 | 4120101 |
| 30. | 04213075 | Brandy Run Near Girard (PA) | 41°59'31" | 80°17'29" | 12 | 4120101 |
| 31. | 04213152 | Walnut Creek Upstream Pool Near Erie (PA) | 42°04'26" | 80°14'05" | 97 | 4120101 |
| 32. | 04213500 | Cattaraugus Creek At Gowanda (NY) | 42°27'48" | 78°56'03" | 1137 | 4120102 |
| 33. | 04214500 | Buffalo Creek At Gardenville (NY) | 42°51'17" | 78°45'18" | 368 | 4120103 |
| 34. | 04215000 | Cayuga Creek Near Lancaster (NY) | 42°53'24" | 78°38'42" | 250 | 4120103 |
| 35. | 04215500 | Cazenovia Creek At Ebenezer (NY) | 42°49'47" | 78°46'30" | 350 | 4120103 |
| 36. | 04216000 | Niagara River At Buffalo (NY) | 42°52'40" | 78°54'59" | 682980* | 4120200 |
| 37. | 04216220 | Niagara River At Black Rock Lock At Buffalo (NY) | 42°56'02" | 78°54'17" | 682980* | 4120104 |
| 38. | 04218000 | Tonawanda Creek At Rapids (NY) | 43°05'35" | 78°38'10" | 904 | 4120104 |
| 39. | 04218518 | Ellicott Creek Below Williamsville (NY) | 42°58'40" | 78°45'49" | 211 | 4120104 |

* including the upper Laurentian Great Lakes.

**Table A6**. List of permanent water level gauges located within the study region, and operated by the Canada's Department of Fisheries and Oceans (DFO) and the United States National Oceanic and Atmospheric Administration (NOAA).

| # | Gauge # | Name (Location) | Latitude | Longitude | Other known ID # |
|---|---|---|---|---|---|
| I. | St. Clair River: | | | | |
| 1 | 9014098 | Fort Gratiot (US, MI) | 43° 0.40 N | 82° 25.3 W | |
| 2 | 9014096 | Dunn Paper (US, MI) | 43° 0.20 N | 82° 25.3 W | |
| 3 | 11940 | Point Edward (CA, ON) | 42° 59.5" N | 82° 25.3 W | 02GG010 |
| 4 | 9014090 | Mouth Of The Black River (US, MI) | 42° 58.5 N | 82° 25.1 W | |
| 5 | 9014087 | Dry Dock (US, MI) | 42° 56.7 N | 82° 26.6 W | |
| 6 | 9014080 | St Clair State Police (US, MI) | 42° 48.7 N | 82° 29.1 W | |
| 7 | 11950 | Port Lambton (CA, ON) | 42° 39.4 N | 82° 30.4 W | 02GG011 |
| 8 | 9014070 | Algonac (US, MI) | 42° 37.3 N | 82° 31.6 W | |
| II. | Lake St. Clair: | | | | |
| 9 | 11965 | Belle River (CA, ON) | 42° 17.8 N | 82° 42.7 W | 02GH005 |
| 10 | 9034052 | St Clair Shores (US, MI) | 42° 28.4 N | 82° 52.8 W | |
| III. | Detroit River: | | | | |
| 11 | 9044049 | Windmill Point (US, MI) | 42° 21.4 N | 82° 55.8 W | |
| 12 | 9044035 | Fort Wayne (US, MI) | 42° 17.9 N | 83° 05.6 W | |
| 13 | 9044030 | Wyandotte (US, MI) | 42° 12.1 N | 83° 08.8 W | |
| 14 | 11995 | Amherstburg (CA, ON) | 42° 08.7 N | 83° 06.8 W | 02GH008 |
| 15 | 9044020 | Gibraltar (US, MI) | 42° 05.5 N | 83° 11.2 W | |
| 16 | 12005 | Bar Point (US, MI) | 42° 03.7 N | 83° 06.9 W | 02GH009 |
| IV. | Lake Erie: | | | | |
| 17 | 12065 | Kingsville (CA, ON) | 42° 01.6 N | 82° 44.1 W | 02GH010 |
| 18 | 12250 | Erieau (CA, ON) | 42° 15.6 N | 81° 54.9 W | 02GF002 |
| 19 | 12400 | Port Stanley (CA, ON) | 42° 39.5 N | 81° 12.8 W | 02GC027 |
| 20 | 12710 | Port Dover (CA, ON) | 42° 46.9 N | 80° 12.1 W | 02GC028 |
| 21 | 12865 | Port Colborne (CA, ON) | 42° 52.5 N | 79° 15.2 W | 02HA017 |
| 22 | 9044036 | Fort Wayne (US, MI) | 42° 17.9 N | 83° 05.6 W | |
| 23 | 9063090 | Fermi Power Plant (US, MI) | 41° 57.6 N | 83° 15.4 W | |
| 24 | 9063085 | Toledo (US, OH) | 41° 41.6 N | 83° 28.3 W | |
| 25 | 9063079 | Marblehead (US, OH) | 41° 32.6 N | 82° 43.9 W | |
| 26 | 9063063 | Cleveland (US, OH) | 41° 32.5 N | 81° 38.1 W | |
| 27 | 9063053 | Fairport (US, OH) | 41° 45.6 N | 81° 16.9 W | |
| 28 | 9063038 | Erie (US, PA) | 42° 09.2 N | 80° 05.6 W | |
| 29 | 9063028 | Sturgeon Point (US, NY) | 42° 41.5 N | 79° 02.8 W | |
| 30 | 9063020 | Buffalo (US, NY) | 42° 52.6 N | 78° 53.4 W | |

Abbreviations: CA, Canada; US, United States; ON, Ontario; MI, Michigan; OH, Ohio; PA, Pennsylvania; NY, New-York.

**Table A7**. Water quality stations used in deriving TP Loadings for 2003-2016. Abbreviations for the water quality stations are explained in Table A8.

| System Segment | Tributary Name | Water Quality Station ID |
|---|---|---|
| SCR | St. Clair River Upstream | ECCC station at Point Edward (43.0048°N, −82.4155°W) |
| | St. Clair River Upstream | station at Port Huron (MIDEQ 740376; USEPA #3740376) |
| | Black River-MI | MIDEQ 740267; 21MICH_WQX-740267; USGS-04160075 |
| | Belle-Pine Complex | UAL* from Clinton River |
| | St. Clair River Downstream | ECCC station at Port Lambton (42.6589°N, −82.5068°W) |
| | St. Clair River Downstream | Station at Algonac (MIDEQ 740016; USEPA #740016) |
| LSC | Clinton River | MIDEQ 500233 |
| | Ruscom River | PWQMN 04001000302 |
| | Thames River | PWQMN 04001308202 & WQMSD |
| | Sydenham River | PWQMN 04002701702 & WQMSD |
| DR | Detroit River Upstream | Station at Fort Wayne (21MICH-820414; USEPA #820017) |
| | Canard River | PWQMN 10000200202 |
| | Turkey Creek | PWQMN 10000100302 |
| | Rouge River | MIDEQ 820070 |
| | Detroit River Downstream | GLWAP & 21MICH-820017 |
| WB | Huron River-MI | OHEPA 580364 |
| | Swan Creek | UAL* from Raisin River |
| | Raisin River | NCWQR |
| | Ottawa River | UAL* from Raisin River |
| | Maumee River | NCWQR |
| | Portage River | OHEPA 500510/NCWQR |
| CB | Sandusky River | NCWQR |
| | Huron River-OH | OHEPA 501030 |
| | Vermilion River | NCWQR/OHEPA 501260/USGS 04199500 |
| | Black River-OH | OHEPA 501510/USGS 04200500 |
| | Rocky River | OHEPA 501790 |
| | Cuyahoga River | NCWQR |
| | Chagrin River | OHEPA 502400 |
| | Grand River-OH | NCWQR/502530 |
| | Ashtabula-Conneaut Complex | USGS 04212945 (or UAL* from Grand River Ohio) |
| | Catfish Creek | PWQMN 16009700802/1600150104 & WQMSD |
| | Kettle Creek | PWQMN 16008701002/1600150103 & WQMSD |
| EB | Big Otter Creek | PWQMN 16010900802/1600150106 & WQMSD |
| | Erie-Chautaugua Complex | UAL* from 1. Cattaraugus Creek or 2. Grand River |
| | Cattaraugus Creek | NYDEC 01041302B/USGS 04213500 |
| | Eighteenmile Complex | UAL* from 1. Cattaraugus Creek or 2. Grand River |
| | Big Creek | PWQMN 16012401102 |
| | Lynn River | PWQMN 16015900302 & WQMSD |
| | Grand River-ON | PWQMN 16018403502 & WQMSD |
| | Nanticoke Creek | PWQMN 16016400102 & WQMSD |
| NR | Niagara River at Fort Erie | ECCC station at Fort Erie (station #ON02HA0045) |

*UAL, the unit area load (UAL) extrapolation approach used in estimates of loads from unmonitored areas.

**Table A8**. Abbreviations for the water quality monitoring agencies listed in Table A7.

| Acronym | Description | Agency/Institution |
|---|---|---|
| ECCC | Upstream/Downstream Monitoring Program for rivers St. Clair and Niagara | Environment and Climate Change Canada |
| MIDEQ | Water Resources Division | Michigan Department of Environmental Quality |
| NCWQR | National Center for Water Quality Research | Heidelberg University |
| NYDEC | Division of Water | New York Department of Environmental Conservation |
| OHEPA | Division of Surface Water | Ohio Environmental Protection Agency |
| PWQMN | Provincial Water Quality Monitoring Network | Ontario Ministry of Environment, Conservation and Parks |
| STORET/NWIS | Water Quality Portal | United States Environmental Protection Agency (USEPA), USGS |
| USEPA | - | United States Environmental Protection Agency |
| USGS | Water Resources Division - NWIS | United States Geological Survey |
| WQMSD | Water Quality Monitoring and Surveillance Division | Environment and Climate Change Canada (ECCC) |
| WSC | Water Survey of Canada | Environment and Climate Change Canada (ECCC) |

**Table A9.** Name, location, and characteristics of Canadian ground water monitoring wells* with reported concentrations of TP in the ground water and located within 8 km wide band of shoreline surrounding the lake boundaries.

| # | Location (Lake Erie basin) | Well ID | Latitude (North) | Longitude (West) | Ground Elevation (m.a.s.l.) | Well Depth (m.b.gr.) | Aquifer Type | Lithology of Aquifer | # of TP observations |
|---|---|---|---|---|---|---|---|---|---|
| 1 | Western | W0000099-1 | 42°1'53" | 82°54'4" | 192.58 | 39.32 | Bedrock | Limestone | 13 |
| 2 | | W0000358-1 | 42°2'16" | 83°1'49" | 177.85 | 19.20 | Interface | Gravel, limestone | 13 |
| 3 | | W0000359-1 | N/A | N/A | 203.27 | 41.15 | Bedrock | Limestone | 13 |
| 4 | | W0000205-2 | 42°7'22" | 82°36'10" | 194.00 | 28.96 | Overburden | Sand | 4 |
| 5 | | W0000112-1 | 42°5'32" | 82°51'16" | 190.00 | 32.77 | Interface | Silty till, Sand, Silt | 12 |
| 6 | | W0000204-1 | 41°46'4" | 82°40'14" | 175.11 | 28.96 | Bedrock | Limestone | 13 |
| 7 | Central | W0000237-1 | N/A | N/A | 180.92 | 42.06 | Bedrock | Shale | 12 |
| 8 | | W0000236-1 | N/A | N/A | 189.81 | 50.9 | Overburden | Gravel, Sand | 1 |
| 9 | | W0000181-1 | 42°19'12" | 81°56'11" | 179.45 | 38.1 | Interface | Gravel, Shale | 11 |
| 10 | | W0000249-1 | 42°25'10" | 81°55'49" | 200.09 | 35.66 | Overburden | Gravel, Sand | 13 |
| 11 | | W0000445-1 | 42°34'13" | 81°31'41" | 199.38 | 85.34 | Bedrock | Shale | 7 |
| 12 | | W0000185-1 | N/A | N/A | 212.05 | 42.98 | Overburden | Sand | 11 |
| 13 | | W0000452-1 | 42°42'31" | 81°11'48" | 210.58 | 60.96 | Overburden | Sand, Gravel | 11 |
| 14 | | W0000335-2 | 42°43'37" | 80°53'2" | 225.75 | 10.97 | Overburden | Sand, Silty clay | 3 |
| 15 | Eastern | W0000169-1 | 42°39'31" | 80°30'38" | 208.23 | 12.5 | Overburden | Sand | 3 |
| 16 | | W0000171-2 | 42°45'2" | 80°25'12" | 231.53 | 7.62 | Overburden | Sand | 3 |
| 17 | | W0000271-1 | 42°50'4" | 80°8'57" | 205.12 | 41.15 | Bedrock | Limestone | 5 |
| 18 | | W0000178-1 | 42°56'40" | 79°56'46" | 216.365 | 25.91 | Bedrock | Limestone | 13 |
| 19 | | W0000289-1 | 42°54'4" | 79°6'56" | 186.34 | 4.5 | Bedrock | Limestone | 18 |

* The Canadian monitoring well data was obtained from the Ontario Provincial Groundwater Monitoring Network: PGMN (PGMN, 2022); the sampling frequency is one sample per year.

**Table A10.** Mean over-lake precipitation ($Y$; m$^3$ s$^{-1}$), calculated volume-weighted mean total phosphorus (TP) concentration ($p^{PR}$; mg L$^{-1}$) in precipitation, and over-lake TP wet and total deposition rates ($D^w$, $D^t$; Metric Tonnes per Annum: MTA) in Lake St. Clair and Lake Erie for each water year during the study period (2003 – 2016).

| Water Year | Lake St. Clair | | | | Lake Erie | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | $p^{PR}$ | $Y$ | $D^w$ | $D^t$ | $p^{PR'}$ | $p^{PR''}$ | $p^{PR}$ (mean) | $Y$ | $D^w$ | $D^t$ |
| 2003 | 0.0304 | 21.90 | 21.0 | 72.4 | 0.0280 | 0.0114 | 0.0197 | 580.3 | 360.5 | 1243.8 |
| 2004 | 0.0311 | 20.00 | 19.7 | 67.8 | 0.0152 | 0.0156 | 0.0154 | 564.3 | 274.9 | 948.4 |
| 2005 | 0.0321 | 22.40 | 22.7 | 78.3 | 0.0122 | 0.0090 | 0.0106 | 447.4 | 149.4 | 515.3 |
| 2006 | 0.0452 | 31.20 | 44.5 | 153.5 | 0.0090 | 0.0087 | 0.0089 | 722.3 | 201.9 | 696.5 |
| 2007 | 0.0267 | 26.40 | 22.2 | 76.7 | 0.0287 | 0.0136 | 0.0211 | 799.9 | 532.6 | 1837.6 |
| 2008 | 0.0066 | 23.00 | 4.8 | 16.6 | 0.0051 | 0.0175 | 0.0113 | 658.6 | 235.5 | 812.6 |
| 2009 | 0.0216 | 25.30 | 17.2 | 59.4 | 0.0317 | 0.0156 | 0.0237 | 577.1 | 430.4 | 1484.9 |
| 2010 | 0.0091 | 32.60 | 9.3 | 32.2 | 0.0062 | 0.0122 | 0.0092 | 825.4 | 239.2 | 825.3 |
| 2011 | 0.0067 | 27.30 | 5.8 | 19.9 | 0.0127 | 0.0167 | 0.0147 | 604.0 | 279.2 | 963.2 |
| 2012 | 0.0087 | 29.80 | 8.2 | 28.3 | 0.0080 | 0.0110 | 0.0095 | 684.3 | 205.1 | 707.5 |
| 2013 | 0.0182 | 26.30 | 15.1 | 52.1 | 0.0120 | 0.0202 | 0.0161 | 580.3 | 295.0 | 1017.6 |
| 2014 | 0.0098 | 29.10 | 9.0 | 30.9 | 0.0050 | 0.0128 | 0.0089 | 702.0 | 198.1 | 683.3 |
| 2015 | 0.0064 | 28.80 | 5.8 | 20.0 | 0.0068 | 0.0153 | 0.0110 | 656.2 | 228.6 | 788.7 |
| 2016 | 0.0067 | 30.00 | 6.3 | 21.9 | 0.0115 | 0.0238 | 0.0177 | 708.2 | 395.5 | 1364.5 |
| Average: | 0.0185 | 26.7 | 15.1 | 52.1 | 0.0137 | 0.0145 | 0.0141 | 650.7 | 287.6 | 992.1 |

Note: Two wet precipitation stations for Lake Erie, at Point Pelee and Rock Point (Fig. 1; Appendix A5), are indicated with symbols <’> and <’’>, respectively.

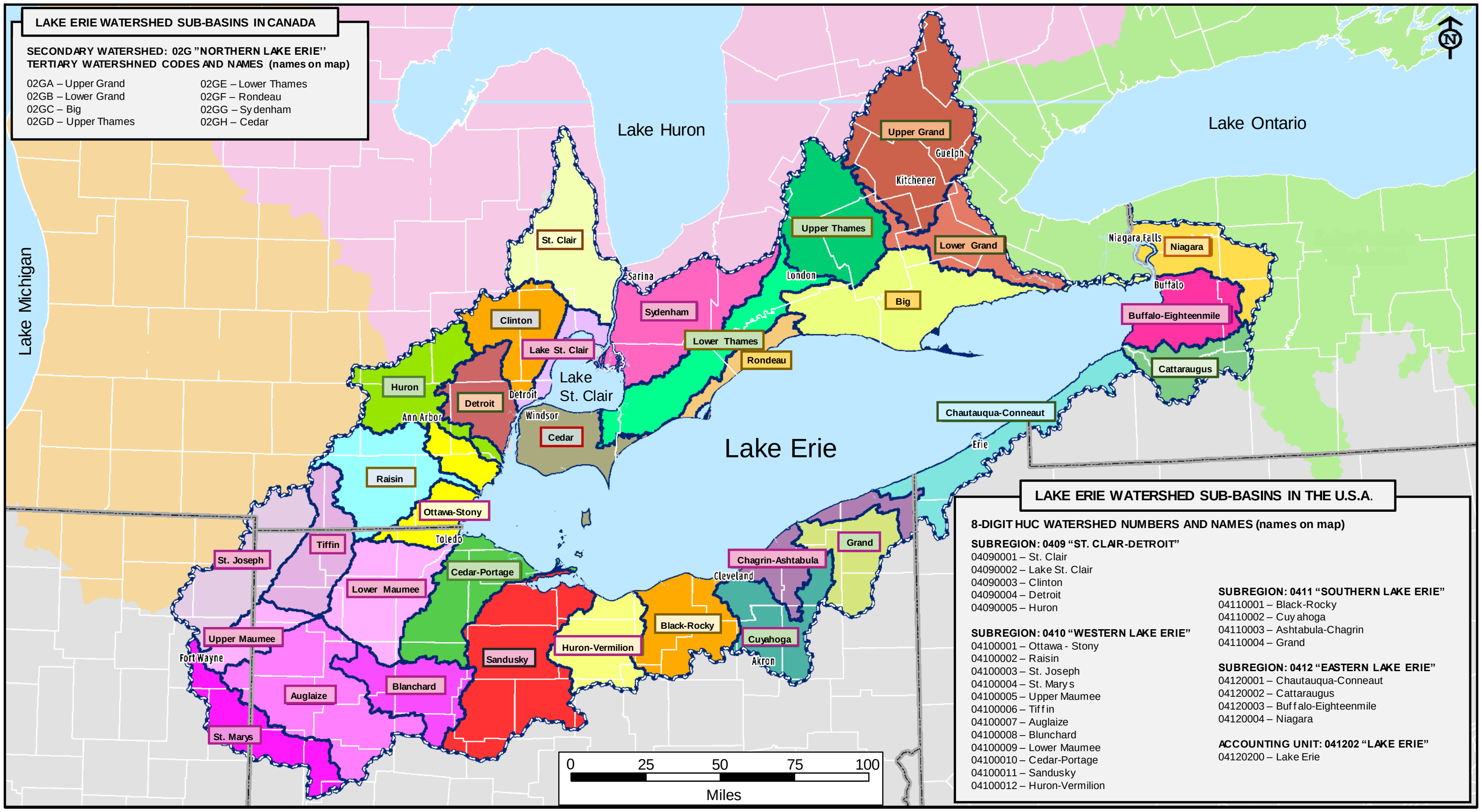


**Figure A1**. Map of Lake Erie sub-watersheds (modified from the map courtesy to the Ohio Department of Natural Resource - ODNR map library). The eastern most sub-watershed with identification code 04120004, named "Niagara" and discharging directly to the Niagara River, was not included into the water and phosphorus mass budgets.

**Appendix A1**. *Defining an appropriate temporal scale for a mass budget construction.*

To construct a mass budget based on input-output approach for a given conservative substance (e.g. water) in any system with a significant volume ($V$) compared to its flow-through rate ($Q$) and resulting a high water storage capacity or mean water residence time ($R_w$; $R_w = V/Q$), it is important to construct a budget over the period of time appropriate for the transport time scales (e.g. larger than water age for a water parcel at any location in the lake) and which is long enough to minimize the effects of initial conditions, and keep the effect of initial conditions on the results as small as possible.

$R_w$, a water storage capacity, or also known as a mean water residence time, in Lake Erie System (LES) is primarily determined by Lake Erie's $R_w$ (Bolsenga and Herdendorf, 1993: 1000 days or ~2.8 years; this study: 944 days or ~2.6 years, Table 4) because of negligible value of $R_w$ for Lake St. Clair (~ 9 days; Bocaniov et al., 2019), the St. Clair River (0.88 day; Griffiths et al., 1991) and Detroit River (0.83 day; Derecki, 1984). Assuming Lake Erie as an ideally mixed continuous steering tank reactor (CSTR) and considering a Lake Erie's $R_w$ = 2.8 years, then 4.6 times of $\tau$ or 13 years would be needed for the outflow to contain a 1% of the initial lake concentration. Therefore, in this study we decided to use a period of 14 years which is larger than 4.6 times $R_w$, so that the lake concentrations will be in full equilibrium (>99%) with the external loads. Over 14 years when the steady state is reached, the in-lake P concentrations must be in equilibrium with the loads, so the left sides in Equations 8 and 10-12 (Table 1) will be zero, so the internal net P load can be estimated for each specific segment.

**Appendix A2**. *The run-off from the Thames, Sydenham, and Grand Ontario rivers*.

The run-off from the three largest sub-watersheds (the Thames, Sydenham, and Grand Ontario rivers) was estimated using the following approach. For these sub-watersheds, the most downstream gauging stations were relatively far away from the river outlets with significant changes occurring in elevations between river outlet and monitored areas (up to 360 m; for example, the Grand Ontario River: elevations at the source and at the mouth are 525 and 174 m above sea level, respectively) which may affect along-river precipitation pattern. For these three sub-watersheds we calculated the total runoff using the exponential relationship (Eq. A2.1):

$$R = R_g \cdot K^S \tag{A2.1}$$

$$K = \frac{A}{A_g} \tag{A2.2}$$

$$S = \frac{\log_{10}\left(\frac{R_1}{R_2}\right)}{\log_{10}\left(\frac{A_1}{A_2}\right)} \tag{A2.3}$$

where, $R$ is the total runoff into the lake, $R_g$ is the runoff at the furthermost downstream gaging station in the basin, $K$ is the ratio of the total area of the drainage basin ($A$) to the drainage area above the gaging station ($A_g$), $S$ is the basin exponent, $R_1$ and $R_2$ are the gauged runoffs from downstream and upstream gauges, $A_1$ and $A_2$ are the gauged areas corresponding to $R_1$ and $R_2$.

**Appendix A3**. *Approach to close the water budget.*

The constructed budget revealed the negative residuals of 6 and 52 $m^3 s^{-1}$ for lakes St. Clair and Erie, respectively. Though, this is a small amount accounting for less than 1% of the combined flow for connecting channels and tributary, it must be balanced. It was done by correcting flows with the largest uncertainties in estimates and using them as a target for budget closure. Lake water levels as well as flows in connecting channels and water diversions are relatively accurately monitored using a network of land- and shoreline based in-situ gauge measurements. Consumptive water use is also strictly regulated and monitored (GLC, 2019). Precipitation is also measured via a network of land-based and shoreline-based rain gauges. The reported direct groundwater water discharge into the lake is small accounting for not more than 12 $m^3 s^{-1}$. The most uncertain element is evaporation which is a simulated value and not a measured one. Moreover, in a recent study, Shao et al. (2020) measured evaporation rates at two stations in the western basin from Sept. 2011 to May 2016 and reported lower values for mean annual evaporation compared to those derived from the GLERL hydrologic database (GLERL, 2020a) which could suggest that the evaporation rates in the database may slightly overestimate the open-lake evaporation results. Therefore, to close the water budget we adjusted our mean annul evaporation rates from 37 to 31 $m^3 s^{-1}$ and from 763 to 715 $m^3 s^{-1}$ in lakes St. Clair and Erie, respectively (Table 6; Fig. 2b).

**Appendix A4**. *Water budget.*

Over-lake precipitation data were obtained from the NOAA Great Lakes Environmental Research Laboratory (GLERL) Great Lakes Monthly Hydrologic Data base (GLERL, 2020a). Over-lake evaporation data were also obtained from GLERL (2020a), where it is calculated using daily simulations from NOAA-GLERL's one-dimensional Net Basin Supply model (Hunter et al., 2015).

Daily flow rates for the major tributaries were downloaded from the USGS National Water Information System (USGS, 2019) for sites in the US and from the National Water Data Archive HYDAT (HYDAT, 2020) for sites in Canada (Tables A4 & A5), scaled to the entire sub-watershed area to include the unmonitored areas (Tables A2 & A3) based on area-weighted estimates for the closest monitored areas in most cases. The approach for the three largest sub-watersheds in Canada (the Thames, Sydenham, and Grand Ontario rivers) is described in Appendix A2.

For the connecting channels, the St. Clair and Detroit rivers, we used the USGS station 04159130 at Port Huron and 04165710 at Fort Wayne, respectively (Table A5: #1 and #6) for October 2008 to 2016 and onwards. For October 1 2002 to September 2008, we calculated daily flows based on daily mean water levels and stage-fall-discharge relationships (Fay and Kerslake, 2009) similar to Scavia et al. (2019a, b). To estimate the daily flows for the outflow, the Niagara River (Fig. 1), we used USGS station 04216000 Niagara River at Buffalo NY; Table A5: #36).

Our estimate of direct groundwater inflow to Lake Erie was based on the results of the fully-integrated surface water-groundwater model (Xu et al., 2021) that estimated an annual average rate to be 11.9 $m^3$ $s^{-1}$.

The outflow through the Welland Canal was estimated as the reported data by WSC (Water Survey of Canada; WLF, 2020) for station 02HA019 (Table A4: #30). Apart from the Welland Canal diversion, there are two small diversions bringing Lake Huron water to the system, bypassing the St. Clair River: water intakes for Detroit, Michigan and London, Ontario. London diverts about 3 $m^3$ $s^{-1}$ from Lake Huron and returns it to Lake St. Clair via the Thames River (Quinn and Edstrom, 2000) as treated sewage.

The sewage outfall is upstream of the water flow gauging station 02GE003 (Table A4: #9) which means that this diverted amount of water is already accounted for by this gauging station. Detroit also withdraws water from Lake Huron via its Lake Huron WTP water intake and returns it to Lake St. Clair. The rated capacity of the Lake Huron WTP is 17.5 $m^3 s^{-1}$ (DWSD, 2019). However, due to current conditions of treatment facilities and water demand, the maximum and average daily flows of this WTP is only 12.3 $m^3 s^{-1}$ and 7.0 $m^3 s^{-1}$, respectively (2003 to 2015 data: DWSD, 2019).

Consumptive water use was defined as the difference between the amount of water withdrawn for all purposes including but not limited to municipal supply, agricultural and industrial uses and the amount of water returned to the system on the annual base, and was estimated for Lake Erie system based on the annual report of the Great Lakes Regional Water Use Database representing water use data 2003 through 2016 and prepared by the Great Lakes Commission (GLC, 2019)

Annual net changes in storage of lakes St. Clair and Erie were calculated as the product of the measured changes in lake stage and surface area of each lake. In large shallow lakes, as our lakes St. Clair and Erie, the water levels are highly sensitive to the wind set-ups (storm surges) and surface seiches which can cause great variability in lake stage at different parts of a lake, we used available measurements of mean daily water levels at multiple locations around lakes obtained from a permanent network of water level gauges operated by the Canada’s Department of Fisheries and Oceans (DFO) and the United States National Oceanic and Atmospheric Administration (NOAA) and listed in Table A6.

**Appendix A5**. *Estimates of atmospheric total phosphorus (TP) deposition.*

Within the Great Lakes Basin, Environment and Climate Change Canada (ECCC) maintains a nine-station network of atmospheric deposition stations, named the Great Lakes Precipitation Network (GLPN). Within GLPN, Canada operates one station located on Lake St. Clair, and two stations located on Lake Erie. The St. Clair site is located on the east shore of Lake St. Clair (Fig. 1). The two stations on Lake Erie are located at Point Pelee and Rock Point. Point Pelee station is located at the southwestern end of Lake Erie near the Point Pelee, while Rock Point station is located on the northeastern shore of Lake Erie (Fig. 1). Station locations and sampling protocols are described in Chan et al. (2003). All three stations are downwind of the major industrial centers and heavily industrialized regions of the Detroit, Windsor, Sarnia and Cleveland areas. The sampling sites are located in the close proximity of lake shores and deemed to be regionally representative of each lake atmospheric conditions. These three stations measure total phosphorus concentration in wet/frozen precipitation and can be used for estimates of wet deposition.

The Volume Weighted Mean ($p_i^{PR}$) concentration in precipitation for each water year for segment *i* (where *i* can stand for either Lake St. Clair or Lake Erie) was calculated as the sum of the masses of TP measured in each sample divided by the sum of the volumes of all the samples of precipitation collected for all samples during the given year:

$$p_i^{PR} = \frac{\sum_{j=1}^{j=N} C_j \cdot V_j}{\sum_{j=1}^{j=N} V_j} \qquad \text{(A5.1)}$$

where, $C_i$ is the concentrations in the sample (mg $L^{-1}$), $V_j$ is the volume of the sample *j* (L), *j* is a rain sample, and *N* is the total number of rain samples taken within a given year.

To determine the wet deposition flux ($F_i^W$; mg $m^{-2}$ $year^{-1}$) for segment *i*, we used the annual mass of TP collected at each site, divided by the area (*A*; $m^2$) of the collection bucket (0.0314 $m^2$), corrected for the rain water collection area and difference between the actual precipitation and those captured by the rain collectors.

$$F_i^w = \frac{p_i^{PR} \cdot \sum_{j=1}^{j=N} V_j}{A} \quad \text{(A5.2)}$$

For Lake Erie, the average $F_{LE}^{w}$ was calculated as the mean of two site-specific values measured at Pelee and Rock Point sampling stations ($F_{LE}^{w'}$ and $F_{LE}^{w''}$, respectively). For Lake St. Clair, $F_{LSC}^{w}$ was taken as for the station St. Clair.

Because of one or two contaminated or missing samples within each year, the wet deposition flux was corrected for the difference between collected precipitation and actual over-lake precipitation:

$$F_i^{w,cor} = \frac{Y_i \cdot A}{\sum_{j=1}^{j=N} V_j} \quad \text{(A5.3)}$$

The annual wet deposition rate ($D^w$; MTA) over the entire lake was estimated as:

$$D_i^w = F_i^{w,cor} \cdot A_i \cdot 10^{-9} \quad \text{(A5.4)}$$

where, $A$ is a lake area ($m^2$; see Table 4: Lake St. Clair: $A$ = 1114 x $10^6$ $m^2$; Lake Erie: $A$ = 25657 x $10^6$ $m^2$), $10^{-9}$ is a coefficient to convert milligrams (mg) into metric tonnes (MT).

Based on the measurements of wet and dry TP deposition in the agricultural regions within the continental United States (Anderson and Downing, 2006), the dry deposition rates over the entire lake ($D^d$; MTA) were assumed to be 2.45 times of the $D^w$ rates. The total deposition ($D^t$; MTA) determined as the sum of $D^w$ and $D^d$, was calculated by multiplication of $D^w$ by 3.45.

**Appendix A6**. *On estimates of the TP sedimentation rates in Lake Erie.*

The current appendix provides a comparison of the sedimentation rates in Lake Erie estimated using the apparent settling velocities reported by Chapra et al. (2016) with those reported by literature.

Our sedimentation rates of TP in the western (3188 MTA) and central (5032 MTA) basins are reasonable and consistent with the previous studies. For example, Kemp et al. (1977) estimated the sediment loads to the western and central basins as 2.681 and 5.866 million Metric Tonnes per Annumn (MTA) per year, respectively. The field observations (Williams et al., 1976) suggest the average values of 1,150 and 0.900 µg of P per g dry weight of sediments in the depositional areas of the western and central basins, respectively. This will give us annual TP sedimentation rates of 3,100 and 5,300 MTA in the western and central basins, respectively.

Our TP sedimentation rate for the eastern basin maybe smaller as can be predicted from the sediment budget for this basin with the sedimentation rate of 5.8 million MTA (Kemp et al., 1977; Mortimer, 1987). Such a sedimentation rate assumes the deficit of 4 million MT (Mortimer, 1987) and sediment budget that is not closed. If someone can assume that the sedimentation rates were somehow overestimated by the amount of the deficit (4 million MTA) so it is not 5.8 million but 1.8 million MTA, then all inputs are balanced with the outputs within the eastern basin. This number (1.8 million MT) is closer to our estimate of the TP sedimentation of 1500 MTA. According to Williams et al. (1976) the average value of 1000 µg of P per g dry weight of sediments in the depositional areas of the eastern basin. This would mean a TP sedimentation flux of 1800 MTA. The difference between this and our estimate can be attributed to the arrival and establishment of mussels and their ability to filter the particulate matter and redirect fluxes of phosphorus from the particulate pool to the dissolved pool.

Our estimate TP sedimentation areal rate in the eastern basin is 0.233 gP $m^{-2}$ $yr^{-1}$ (Table 8). Bloesch (1982) reported mean summer (Jun 28 to Sept 8, 1978) deposition rates of Particulate Phosphorus (PP) for his offshore station (depth = 40 m; northern side of the basin) at two depths, 3 m above bottom (hypolimnetic fluxes) and just below epilimnion (epilimnetic fluxes), as 5.44 mgP $m^{-2}$ $d^{-1}$

and 1.3 mgP $m^{-2}$ $d^{-1}$, respectively. He observed that epilimnetic fluxes of PP were 4-20 times less than the PP fluxes measured in the hypolimnion due to effect of frequent resuspension events at the bottom of the lake. This means that the measured sedimentation rates near the bottom of the eastern basin can be significantly overestimated. This may suggest that the high sedimentation rates of 5.8 million MTA fine-graned sediments observed by Kemp et al. (1977) in the eastern basin were overestimated by a factor of 3 as they were not corrected for the bottom resuspension effects, and in this case no deficit of 4 million MT of fine-grained sediments exists in the eastern basin. Moreover, if Bloesch’ (1982) summer averaged epilimnetic sedimentation flux of PP (1.3 mgP $m^{-2}$ $d^{-1}$ = 0.0013 gP $m^{-2}$ $d^{-1}$) is assumed to be the same for the entire year (0.0013 gP $m^{-2}$ $d^{-1}$ x 365.25 d $yr^{-1}$ = 0.475 gP $m^{-2}$ $d^{-1}$) then it will equal to 0.475 gP $m^{-2}$ $d^{-1}$ that is only 2 times higher than our annual rate of 0.233 gP $m^{-2}$ $d^{-1}$ (Table 8), and it is reasonable to assume that the mean summer sedimentation rates are higher than the mean annual rates, and that the arrival of mussels (~1990) resulted in clearing of the water column and reduced standing crop of the detrital carbon and phytoplankton biomass not only nearshore but also offshore (e.g. see Bocaniov et al., 2014, for the nearshore offshore differences in the phytoplankton biomass, and for the effect of the advection of the nearshore waters with lower Chl-a on the offshore biomass of Chl-a). So, the pre-mussel (e.g. 1978 rates) sedimentation rates should be smaller than the after-mussel sedimentation rates (e.g. 2003 to 2016).

There are other indirect indications that the offshore sedimentation rates of TP are relatively small. For example, the biomass of mussels in the deep offshore areas of the eastern basin is large but the population is composed from the large-sized older individuals with the absence of smaller-size

**Appendix A7**. *Total phosphorus (TP) mass balance.*

*Tributaries and Connecting Channels* – All tributary and connecting channel nutrient loads were estimated using the Water Regressions on Time, Discharge, and Season (WRTDS) method (Hirsch et al., 2010) based on observed concentrations and flow data. To estimates P loads with three connecting channel flows (rivers St. Clair, Detroit and Niagara) where TP concentrations are not correlated with channel discharges and WRTDS method cannot be applied, similar to other previous studies (Scavia et al., 2019a, b), we used the Generalized Additive Model (Wood, 2011; Wood, 2017) to approximate daily TP concentrations as a function of time, with the smoothing parameters selected by the Restricted Maximum Likelihood (REML) method. The estimated daily TP concentration and associated standard error were multiplied by daily discharge to estimate daily loading. The daily estimates were then aggregated to estimate annual loadings.

TP concentration data were obtained from the United States Water Quality Portal (WQP, 2019), the Provincial (Stream) Water Quality Monitoring Network (PWQMN, 2019), Michigan Department of Environmental Quality (MDEQ), the Great Lakes Intake Program (GLIP, 2019) managed by the Ontario Ministry of the Environment, Conservation and Parks (MECP), and Environment and Climate Change Canada (ECCC; communication with D. Burniston and A. Dove, 2018).

For the St. Clair River, TP loads were estimated at the river source (Point Edward, Fig. 1: site #1; Port Huron, Fig. 1: site #2), and two sites close to the river mouth (Port Lambton, Fig. 1: site #3; Algonac, Fig. 1: site #4). At some stations (e.g. St. Clair River at Port Huron), similar to Scavia et al. (2019a, b), we removed extreme outliers defined here as concentrations higher than the mean plus four times the standard deviation (SD) and representing less than 0.02% of data.

Due to lower resolution of the water quality and nutrient monitoring activities at the river source (Point Edwards; Port Huron) compared to the timing and duration of the short-lived episodic events (shoreline erosion and resuspension of sediments in the nearshore due to storm surges, searches and surface waves; see Scavia et al., 2019a, b), some of the TP load is missed.  To estimate it we calculated as the difference of the TP load at the St. Clair River downstream location (average of loads calculated at both sites, Point Edwards and Port Huron) and a sum of estimated loads for the St. Clair River upstream location and for the direct river sub-watersheds. The load of the St. Clair River to Lake St. Clair was calculated as the average of loads calculated for both sides of the river, at Port Lambton (Ontario; Fig. 1: site #3) and Algonac (Michigan; Fig. 1: site #4).

For the LSC TP load to Detroit River we used station #21MICH-820414 (USEPA #820017) at the lake outflow (Table A7; Fig. 1: site #5). We estimated the total annual load from the Detroit River to the western basin of Lake Erie as a sum of this LSC load and loads from Detroit River sub-watersheds. This is because water quality measures at the downstream monitoring station (#21MICH-820017; Table A7) can be biased due towards frequent episodes of Lake Erie surface seiches and wind-driven water setups transmitting lake water upstream from the lake to the river.

*Point Sources* - Point sources identified here are only those with direct inputs downstream of the river mouth monitoring stations. Those discharging upstream of the monitoring stations were included in the tributary loads. Direct annual point TP loads from 2003 to 2016 for each sub-watershed were from Scavia et al. (2019a, b) for the St. Clair River, Lake St. Clair, and Detroit River. For the three basins of Lake Erie, direct loads were those in Maccoux et al. (2016). The latter study reported those loads for only 2003 to 2013. Because there was very little inter-annual variability, we assumed the loads for 2014 to 2016 to be the same as averaged 2003-2013 values. This assumption should not affect results or conclusions because direct TP loads are a very small compared to the total watershed loads (Maccoux et al., 2016; Scavia et al., 2019a, b).

*Groundwater* - The volume of direct groundwater to Lake Erie is small (11.9 $m^3 s^{-1}$; Xu et al., 2021) accounting for less than 1% of the combined over-lake precipitation and basin runoff or less than 0.25% of the Detroit River inflow to the lake. In terms of phosphorus concentrations, the direct ground water discharge into Lake Erie can be characterized by chemical composition of ground water in monitoring wells located within 8 km wide band surrounding the lake, because this zone, along with the lake bed, was suggested as the zone most important for ground water – lake water exchange processes

(Xu et al., 2021). The Canadian monitoring well data was obtained from the Ontario Provincial Groundwater Monitoring Network (PGMN) Program (PGMN, 2022) and the U.S monitoring well data was obtained from the USGS National Ground-Water Monitoring Network (USGS, 2022). Within such an 8 km wide band zone, on the Canadian portion of the lake, we identified 19 ground water monitoring wells reporting total phosphorus concentrations (Table A9), and there were no wells found on the USA portion of the lake with reported measurements of water chemistry. Therefore, our estimates of TP concentrations in the direct inputs of deep groundwater were based on Canadian sources. Because of the low frequency of data (one sample per year) and to extend the number of observations, we extended the period of observations to all available data, from 2002 to 2019. To reduce the importance of a few large outliers, present in the dataset, that might result from local contamination, we decided to use a pooled dataset for the entire basin and base our analysis on the most typical value that is median rather than using an average value.

*Over-lake atmospheric deposition* - Over-lake atmospheric wet deposition of TP for each water year were based on the product of volume-weighted rain water mean concentration ($p_i^{PR}$) and precipitation. $p_i^{PR}$ was calculated based on the TP concentrations in precipitation by Environment and Climate Change Canada (ECCC) and over-lake precipitation amount obtained from GLERL (2020a) (see Appendix A5 for more details). Total over-lake deposition was assumed to be 3.45 times wet deposition based on estimates of wet and dry depositions for the continental United States (Anderson and Downing, 2006; see Appendix A5 for more details).

*Outputs via the Niagara River and Welland Canal* - TP losses via the Niagara River was estimated with GAM based on the data for river outflow and nutrient concentrations collected at the ECCC upstream monitoring site at Fort Erie (#ON02HA0045; Table A7; Fig. 1: site #6). Due to close proximity of the sources of the Welland Canal and the Niagara River to each other (Fig. 1), it was assumed that the TP concentrations in the lake water outflowing through the water diversion and lake outflow are similar. Therefore, the TP loss via the water diversion was estimated as the proportion of the Niagara River TP load related to the differences in flows.

*Consumptive use* - Contribution of the water consumptive use to the loss of TP was calculated as the annual rate of water taken from the lake and not returned back multiplied by the average annual segment-specific P concentration.

*Sedimentation* - A certain fraction of a lake's TP loading is retained permanently via settling and permanent burial. While net sedimentation is a delicate balance between sedimentation and resuspension rates, over an annual cycle, it has been parametrized for each of the three Lake Erie basins including Lake St. Clair using long-term, mass balance dynamic modeling (e.g. Chapra et al., 2016). To estimate the P removal in each basin due to net sedimentation, we used the mean annual basin-specific concentrations of TP and apparent settling velocities normalized to the basin surface area for post-1990 rates as in Chapra et al. (2016; see also Tables 1, B5, B7, B9 to B11).

*Inter-basin nutrient exchange (bulk mixing)* – To characterize the inter-basin nutrient fluxes due to diffusion we used the bulk diffusion coefficients parameterized in Chapra et al. (2016; see their Table 3) and mean annual basin-specific concentrations in the adjacent basins (Tables 1, B9 to B11).

*Change in storage* - The TP losses because of changes in lakes St. Clair and Erie volumes were calculated as the product of the mean annual difference in lake volume and its average lake-wide annual concentration. For Lake Erie, we used the mean of the observed median values for all three basins from the two spring and summer lake-wide monitoring cruises representing both the isothermal (April) and stratified (August) conditions (GLNPO, 2020). The median values were chosen as a good representation of the most typical nutrient conditions as they are not inflated with potential sources of contamination, with a similar approach undertaken by Chapra et al. (2016). To estimate the mean TP concentration for Lake St. Clair over the study period we used observations of TP concentrations at the lake outflow (#21MICH-820414, USEPA #820017, Fig. 1: site #5, Table A7). Preliminary comparisons of this concentration with the lake averaged concentration determined from all lake loads and water flows, returned similar results, validating the estimated value.

*Internal P load* – It was determined as the difference between means of all annual loss and input fluxes of TP averaged over 2003 to 2016 using Equation 8 (Table 1) for Lake St. Clair and Equations 10-12 (Table 1) for the three basins of Lake Erie.

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