

19 **Abstract**

20 Discharge of groundwater-derived pollutants to inland and marine coastal waters is
21 influenced by the transport and reactive processes occurring in nearshore aquifers. The
22 effect of shoreline change on these processes and subsequent discharge of pollutants to
23 coastal waters is unclear. The objective of this study was to evaluate the impact of shoreline
24 recession (landward movement of the mean shoreline) on the transport of nitrogen [N] and
25 phosphorus [P] in a nearshore aquifer and their discharge to coastal waters. Field
26 investigations were conducted on a permeable unconfined nearshore aquifer on Lake
27 Huron, Canada, in years coinciding with historically low and high lake water levels. At the
28 site, a septic system-derived nutrient-rich (N and P) groundwater plume is moving towards
29 the lake and the mean shoreline position moved ~30 m landward between sampling years
30 due coastal erosion and mean lake water level increase. Data indicate PO₄-P fluxes to the
31 lake were higher following shoreline recession due to shortened travel pathways. In
32 contrast, NO₃-N fluxes were governed by the specific geochemical conditions near the
33 sediment-water interface, which are not only a function of the shoreline position. Further,
34 findings show shoreline recession may modify mineral phases that tend to sequester
35 pollutants (e.g., iron oxides) near the sediment-water interface and this may possibly
36 mediate release of sediment-bound pollutants. The findings provide new insights into
37 potential impacts of shoreline change on chemical discharge to coastal waters as needed to
38 inform long-term water quality predictions and management.

39 **Keywords:** climate change, shoreline erosion, coastal water level change, nutrients, iron curtain,
40 septic system

41 **1 Introduction**

42 Coastal zones are the most populated and developed areas worldwide. Currently over half
43 of the global population resides in coastal areas and this is expected to increase in the future
44 based on current trends of coastal migration and population growth (Small and Nicholls,
45 2003). Sandy beaches, which occur along marine coastlines and along inland coastlines
46 that are adjacent to large inland lakes, make up more than one-third of the ice-free global
47 coastline (Luijendijk et al., 2018). Many sandy coastlines worldwide are threatened by the
48 landward movement of the shoreline, known as shoreline recession. Shoreline recession,
49 which is caused by increases in coastal water levels (i.e. sea level rise or lake level changes)
50 combined with shoreline erosion, is expected to increase as the climate changes (e.g.,
51 higher frequency and more intense storm events) and in response to anthropogenic
52 development (Bird, 2008). Luijendijk et al. (2018) found from satellite image analysis that
53 24% of the world's sandy beaches are eroding at rates exceeding 0.5 m/yr, with only 48%
54 of sandy beaches considered stable. For the Laurentian Great Lakes, which have more than
55 6000 km of permeable shoreline (Government of Canada - Environment and Climate
56 Change Canada and U.S. EPA, 2009), shoreline recession caused by interannual lake water
57 level fluctuations combined with erosive storm events is an increasing challenge. Lake
58 water level fluctuations up to one to two meters occur along large lacustrine shorelines,
59 such as the Great Lakes, approximately once a decade (Argyilan and Forman, 2003;
60 Sellinger et al., 2008).

61 It is well established that shoreline recession along marine and inland coastlines has severe
62 economic, social and environmental consequences due to impacts including land and
63 habitat loss, property damage, and reduced revenue potential from tourism and industry

64 (Mukhopadhyay et al., 2018; Theuerkauf et al., 2019). As such, this is an active area of
65 research, with studies generally focused on monitoring shoreline changes (e.g. Burningham
66 and French, 2017; García-Rubio et al., 2015; Hagenaaars et al., 2018), fundamental
67 understanding of underlying geomorphological and hydrological processes (e.g. Brooks et
68 al., 2017; Swenson et al., 2006), and techniques for forecasting shoreline change over short
69 and long time scales (e.g. Davidson et al., 2017; Mukhopadhyay et al., 2018). Few studies
70 have examined the potential impacts of shoreline recession, including sediment erosion and
71 changes in coastal water levels, on the flux of land-derived pollutants to coastal waters.
72 Two prior studies examined on the impact of shoreline erosion on nutrient loading to Lake
73 Tahoe, United States (Adams and Minor, 2002) and Lake Diefenbaker Reservoir, Canada
74 (Hewlett et al., 2015). Both studies found that nutrient loading caused by shoreline erosion
75 (due to release of sediment-bound nutrients) only contributed a fraction of the total nutrient
76 load into the lakes (< 5% of total nutrient input). More recently, beach sand erosion was
77 found to be the main mechanism of transferring *E.coli* to nearshore coastal waters during
78 intensified wave conditions at a sandy beaches (Vogel et al., 2016). Finally, focused on a
79 coastal wetlands rather than sandy shorelines, Braun et al. (2019) showed that shoreline
80 recession may reduce the carbon storage potential of a coastal wetland and thereby impact
81 carbon dynamics within the wetland. The above-mentioned studies all focus on sediment-
82 bound pollutants that are released to coastal waters as sediment near the shoreline is eroded
83 and released to the coastal waters. While this can be an important transport mechanism,
84 shoreline recession may also affect the delivery of dissolved pollutants into coastal waters,
85 particularly land-derived groundwater pollutants, whose delivery to coastal waters strongly
86 depends on the specific subsurface transport pathways between the land source and coastal

87 waters, as well as the geochemical conditions along these pathways in nearshore aquifers
88 (Beldowska et al., 2022; Destouni et al., 2010). For instance, shoreline recession brings
89 the shoreline closer to areas of human development (e.g. infrastructure, landfills, and
90 contaminated sites), which decreases the distance between a groundwater pollution source
91 and the coastal water. This may in turn increase the potential for groundwater-derived
92 pollutants reaching coastal waters.

93 Submarine groundwater discharge (SGD) and lacustrine groundwater discharge (LGD) are
94 important pathways for land-derived pollutants such as nutrients, metals and organic
95 contaminants to enter marine and inland coastal waters, respectively (e.g. Beck et al., 2016;
96 Lewandowski et al., 2015; Moore, 2010). With increased urbanization, industrialization
97 and intensification of agriculture in coastal areas, groundwater pollutant concentrations can
98 be high in coastal areas and pose a risk to coastal water pollution. The importance of LGD
99 has mainly been recognized for its contribution to lake nutrient loads and lake
100 eutrophication (Robinson, 2015; Rosenberry et al., 2015). For instance, high nutrient
101 concentrations, more than ten times higher than in the adjacent lake water, were observed
102 in groundwater discharging into Lake Simcoe (Roy and Malenica, 2013). In addition to
103 nutrients, studies have also shown that LGD may play an important role in delivering other
104 chemical constituents, including metals (Lee et al., 2014; Rakhimbekova et al., 2018) and
105 road salt (Howard and Livingstone, 2000; Roy, 2019) to lakes. The transport of pollutants
106 via groundwater flow paths to coastal waters is generally slow, and transport can be
107 retarded by various physical and chemical processes (e.g. filtration, sorption,
108 precipitation). Therefore, changes in the shoreline position may impact the groundwater
109 travel times, travel distances and the specific pathways, by which pollutant are transported

110 from their land source to coastal waters. In addition, it is well established that an important
111 reaction zone can exist in nearshore aquifers close to the shoreline due to the mixing of
112 groundwater and coastal water. This zone, commonly referred to as a subterranean estuary
113 in marine environments, is known to strongly influence the discharge of groundwater-
114 derived pollutants to coastal waters due to its high reactivity (Rakhimbekova et al., 2021b,
115 2021c; Robinson et al., 2018). One important reactive phenomenon in this mixing zone is
116 the sequestration (trapping) of pollutants to sediment mineral phases, particularly iron (Fe)
117 oxides, which form under certain redox and pH conditions. This sediment trap mechanism,
118 commonly referred to as the “iron curtain”, can act as a natural barrier that limits the release
119 of a range of pollutants, including phosphorus (Charette and Sholkovitz, 2002; Spiteri et
120 al., 2008), carbon (Linkhorst et al., 2017; Sirois et al., 2018), metals (McAllister et al.,
121 2015), and trace metals (Beck et al., 2008) to coastal waters. It is possible shoreline
122 recession may cause a shift in the location of the mixing zone, thereby impacting reactivity
123 and geochemical conditions, including the function of sediment trap and its ability to
124 sequester pollutants. This may in turn modify pollutant fluxes delivered to coastal waters
125 via SGD and LGD.

126 To the best of our knowledge, there are no studies that have explored the impact of
127 shoreline changes, including shoreline recession, on the discharge of groundwater-derived
128 pollutants to coastal waters. The objective of this study is to address this knowledge gap.
129 Field data collected downgradient of a large septic system over a six-year period is used to
130 examine the influence of shoreline recession on the subsurface transport of a P- and N-rich
131 wastewater plume and its potential discharge to the adjacent lake. In addition, the study
132 investigates the way in which shoreline recession may alter geochemical conditions near

133 the sediment-water interface with a specific focus on the abundance and distribution of
134 mineral phases that tend to sequester pollutants, including P, close to the groundwater-
135 coastal water interface. Finally, this study explores the broader possible impacts of
136 shoreline recession beyond our specific study site by discussion of potential sources of
137 anthropogenic groundwater contaminant sources along shorelines and future projections
138 on lake water level changes and expected shoreline changes. The findings of this study are
139 needed given the prevalence of groundwater contamination near marine and inland
140 coastlines, and the projected changes in coastal water levels and storm intensity and
141 frequency as the climate changes.

142 **2 Methodology**

143 **2.1 Field investigation**

144 ***2.1.1 Site description***

145 Field investigations were conducted at Ipperwash Beach on Lake Huron, Ontario, Canada
146 in 2014, 2015, and 2020 (Figure 1a). Ipperwash Beach is a sandy beach that extends over
147 20 km and has an extensive dune system behind the beach that rises several meters above
148 the lake surface (Eyles and Meulendyk, 2012). Extensive shoreline recession occurred at
149 Ipperwash Beach between 2015 and 2020 in response to an increase in the water level in
150 Lake Huron combined with beach erosion. In 2020, the water level in Lake Huron reached
151 177.45 m (IGLD 85 datum) following over a decade of low water levels during which the
152 record low water level of 175.57 m occurred in 2013 (Gronewold et al., 2021; Gronewold
153 and Stow, 2014). The extent of shoreline recession between 2015 and 2020 are illustrated
154 in aerial photographs shown in Figure 1b.

155 Ipperwash Beach was selected for this study as a public comfort station with a large septic
156 system is located in close vicinity to the beach (Figure 1b). The septic system has been
157 operational since at least 1974 and is only used during the warmer months from May to
158 September. Due to shoreline recession, the mean shoreline position was located ~120 m
159 from the septic system tile bed in 2015, but only ~90 m from the septic tile bed in 2020
160 (Figure 1b). Detailed (approximately monthly) field sampling was conducted between
161 August and October in 2014, and between May and August in 2015, to evaluate the spatial
162 and temporal variability of phosphorus (P) and nitrogen (N) transport from the septic
163 system through the nearshore aquifer to the lake. Details of this previous study are provided
164 in Rakhimbekova et al. (2021a). This current study expands upon this prior work with new
165 sampling performed in 2020 to determine the impact of shoreline recession on the transport
166 of P and N through the nearshore aquifer and their potential discharge to the lake.

167 ***2.1.2 Field monitoring approach***

168 ***2.1.2.1 Field equipment***

169 For all sampling periods in 2014-2015 and 2020, a cross-shore monitoring transect was
170 installed that aligned with the center of the septic system-derived nutrient-rich plume that
171 is moving towards the lake. The original cross-shore monitoring transect in 2014-2015
172 consisted of ten multi-level samplers (MLS) and extended from ~71 m downgradient of
173 the septic tile bed ($x = 71$ m; landward extent of the beach dunes) to ~20 m offshore ($x =$
174 140 m; Figure 1c). For the 2020 sampling, the cross-shore monitoring transect consisted of
175 six MLS, and extended from $x = 71$ m downgradient of septic tile bed ~20 m offshore ($x =$
176 110 m) (Figure 1c). The x - z coordinate system used herein is defined with $x = 0$ m located
177 at the landward edge of the septic tile bed and $z = 0$ as the elevation of a local permanent

178 benchmark. All MLS were installed up to 3 m below ground surface, with sampling ports
179 at 0.2 m depth intervals.

180 Two piezometers (PZ) were permanently installed at $x = 71$ m (PZ-1) and $x = 93$ m (PZ-2)
181 in July 2014, with additional piezometers temporarily installed along the cross-shore
182 monitoring transect at the start of each sampling event to measure the groundwater and
183 lake water levels across the nearshore aquifer and offshore. The location of monitoring
184 equipment and ground surface elevations were surveyed with a total station (Topcon GTS-
185 239W) relative to a local permanent benchmark during each sampling event (Figure 1c).

186 **2.1.2.2 Electrical resistivity imaging**

187 The septic system origin of the nutrient-rich plume observed in the nearshore aquifer was
188 confirmed in 2015 by using artificial sweeteners as a chemical tracer for human wastewater
189 (Rakhimbekova et al., 2021a). However, the artificial sweetener data were limited to point
190 information obtained from samples collected at selected MLS located over 70 m
191 downgradient of the septic tile bed (i.e., samples collected from $x = 71$ m onwards). During
192 the field sampling in 2020, a non-invasive electrical resistivity tomography (ERT) survey
193 was performed to provide continuous imaging of electrical resistivity within the subsurface
194 and determine if this approach would be able to trace the nutrient-rich plume back to the
195 septic tile beds.

196 An ERT survey line consisting of 96 stainless-steel electrodes (0.3 m) with an inline
197 spacing of 1 m was deployed. The location of the line coincided with the location of the
198 cross-shore monitoring transect but it extended from the landward edge of the septic tile
199 bed ($x = 0$ m) to the shoreline ($x = 95$ m) (Figure 1b). A multi-channel Syscal Pro Switch

200 48 resistivity system (IRIS Instruments, France) was used to complete the survey. As this
201 system is limited to 48 electrodes at a time, the roll-along method was used to record the
202 survey line through three successive segments. For this, 24 electrodes were moved for each
203 segment to ensure 50% overlap (i.e., electrode numbers: 0-48, 24-72, 48-96) (Robinson et
204 al., 2022).

205 The multi-gradient electrode array was used with a total of 4183 measurements. Due to the
206 dry surface sand, saline water was added around each electrode to ensure strong ground
207 coupling (resistance: <1 kilo-ohm). The recorded apparent resistivity data was inverted
208 with DC_2DPro, an iterative least-squares smoothness-constrained inversion program
209 (Kim, 2016). The surface elevation at each electrode, which was surveyed using the total
210 station, was incorporated into the inversion, which was then run to a maximum of seven
211 iterations, showing good convergence with a low root mean square (RMS) error of 5.5%.

212 ***2.1.2.3 Porewater sampling and analysis***

213 For all sampling events, pore water samples were first collected from all MLS ports using
214 syringes attached to the PVC sampling tubes ($\phi = 1.35$ mm). Porewater samples were
215 collected from each port, filtered using 0.45 μ m cellulose acetate filters, and stored in 60
216 mL HDPE bottles. Samples were analyzed for phosphate (PO₄-P), nitrate (NO₃-N) and
217 chloride (Cl) within 48 hours of collection. After samples were collected, a peristaltic pump
218 was used to draw pore water directly into a flow cell where pH, electrical conductivity (EC)
219 and temperature were measured with an In-Situ Multi-Parameter TROLL® 9500 and YSI
220 6600 V2 Sonde. Multiple nearshore lake water samples were also collected during each
221 sampling event. Collected pore water samples were analyzed for nutrients (PO₄-P, NO₃-N)

222 using Lachat 750 Flow Injection Analysis (Lachat 750), and Cl using High Performance
223 Liquid Chromatography.

224 **2.1.2.4 Sediment sampling and analysis**

225 During MLS installation in May 2015 and August 2020, multiple sediment samples were
226 collected at depths of 0.5 m to 2 m below the ground surface for sediment chemistry
227 characterization (Figure 1c). Sediment samples were analyzed via total acid digestion using
228 method EPA 200.8 (US EPA, 1996) to determine the total elemental solid phase
229 concentrations. In addition, due to the potential importance of Fe-bearing solid phases in
230 sequestering chemical including P in nearshore aquifers, select sediment samples were
231 treated with (i) acid ammonium oxalate solution (McKeague and Day, 1966), and (ii)
232 dithionite-citrate-bicarbonate solution (Mehra, 1958) to provide characterization of the Fe-
233 bearing solid phases (e.g. degree of crystallization). Oxalate-extractable Fe (Fe_o) is
234 operationally thought to represent amorphous oxides such as ferrihydrite and small
235 amounts of organically-bound Fe (Schwertmann, 1964). Dithionite-extractable Fe (Fe_d) is
236 operationally thought to represent organically-bound oxides, amorphous oxides, and finely
237 crystalline oxides (Akinbola et al., 2013). The difference between Fe_d and Fe_o generally
238 represents the amount of solid phase Fe present in crystalline oxide forms (e.g., goethite
239 and hematite). Total dissolved phosphorus (TDP) and total iron (Fe) in extractant solutions
240 were analyzed using HACH DR 6000 UV VIS Spectrophotometer and inductively-coupled
241 plasma mass spectroscopy (ICP-MS), respectively. Additional details on sediment sample
242 analysis are provided in the Supporting Information (SI Section 1).

243 **2.2 Numerical model**

244 A two-dimensional (2D) model was developed in MODFLOW-NWT (Niswonger et al.,
245 2011) to simulate steady state groundwater flows and estimate NO₃-N and PO₄-P loads
246 discharging to the lake along the vertical cross-shore section. The model domain and
247 parameterization were based on a groundwater model of the study site developed and
248 validated by Malott et al. (2016). The two-dimensional model assumes negligible
249 groundwater flow in the alongshore direction. The model was 150 m long in the cross-
250 shore direction and the aquifer thickness at the shoreline location was set to 13 m at the
251 landward boundary due to presence of a clay layer at this depth according to local well
252 records (Figure 2). The beach face and lake boundaries (AB and BC) were specified based
253 on topographic sand level surveys conducted during each sampling period. The model was
254 discretized into 71 layers and 128 columns with greater refinement around the shoreline
255 and the beach face. Grid discretization tests were performed to ensure a converged solution.
256 Particle size analysis performed on sediment samples ($n = 7$) collected across the nearshore
257 aquifer up to 2.5 m below the water table indicated that the nearshore aquifer is comprised
258 of relatively homogeneous fine sand (Coefficient of Uniformity, $C_u = 2.18$ and sand grain
259 size, $d_{50} = 0.16$ mm) with the saturated K estimated to range from 7 to 15 m/d using the
260 method of Krumbein and Monk (1943). Based on this, the simulated aquifer was
261 homogeneous and isotropic with the hydraulic conductivity (K) set to 10 m/d and effective
262 porosity $n = 0.3$.

263 The upper (AB) and bottom (ED) model boundaries were no-flow boundaries assuming
264 negligible aerial recharge and evaporation, and an impermeable aquifer basement,
265 respectively. The offshore boundary (CD) was specified as no-flow boundary and was

266 located sufficiently far from the shoreline (>50 m offshore) that it did not influence the
267 groundwater flows in the nearshore aquifer (Figure 2). The vertical landward boundary
268 (AE) was located at $x = 60$ m, which is 10 m inland from the landward extent of the beach
269 dunes (Figure 2) and was set to be a constant head boundary. Three simulations were run
270 with the boundary conditions modified to represent the mean hydrological conditions at
271 the study site during the 2014, 2015 and 2020 sampling periods. Based on the measured
272 groundwater levels, the hydraulic head at the landward boundary was set to -2.95 m, -3.05
273 m and -1.95 m, respectively, for the models simulating the groundwater flows over the
274 sampling periods in 2014, 2015 and 2020. A constant head condition was used along the
275 lake boundary (BC) with the specified head values set based on the mean measured lake
276 water level during each of the sampling periods and considering the effect of steady waves
277 acting on the boundary. To simulate the effect of steady waves, the approach of Anwar et
278 al. (2014) was adopted whereby the time-averaged effect of waves as described by wave
279 set-up was used (details are provided in SI Section 2). For this, the empirical equation of
280 Nielsen (2009) was used to estimate the wave-set up profile and calculate head values that
281 were then prescribed along the lake boundary (BC). Offshore root mean square wave
282 heights (H_{rms}) of 0.35 m, 0.25 m and 0.35 m were used in the equation of Nielsen (2009)
283 to simulate the mean wave conditions over the sampling periods in 2014, 2015 and 2020,
284 respectively. The mean H_{rms} values were calculated using monitoring data from a wave
285 buoy located ~35 km from the study site (C45149 Southern Lake Huron, Department of
286 Fisheries and Oceans Canada Buoy, 2020). Based on the mean lake water levels measured
287 at the study site during the sampling periods in 2014, 2015 and 2020, the still lake water
288 level in the models were set to -3.64 m, -3.73 m and -2.21 m, respectively.

289 **3 Results and discussion**

290 **3.1 Field and modeling results**

291 **3.1.1 *Change in hydrological conditions between sampling periods***

292 The hydrological conditions at the study site including the mean lake water level, mean
293 wave conditions, and landward groundwater levels varied between the sampling periods.
294 The mean lake water level was approximately 0.7 m higher in 2020 compared to 2014-
295 2015 (Figure 3a). More intensified wave conditions were also observed in 2020 compared
296 to 2014-2015 with maximum wave height (H_{rms}) reaching 3.7 m in 2020 compared to 3.0
297 m in 2014 and 2.1 m in 2015 (mean H_{rms} was 0.35 m, 0.25 m and 0.35 m in 2020, 2015 and
298 2014, respectively; Figure 3b). The extent of shoreline recession between 2015 and 2020
299 is evident by comparing the topographical surveys performed in 2015 and 2020, along with
300 corresponding aerial photographs (Figures 1b and 1c). The shoreline was estimated to
301 move 30 m landward from $x = 120-125$ m in 2014-2015, to $x = 90$ m in 2020, in response
302 to the lake water level increase and beach sand erosion.

303 Increase in lake water levels may lead to a corresponding increase in the landward
304 groundwater table elevation, and thereby reduce the vertical separation between septic tile
305 beds and groundwater table. This can be a concern as it then limits the efficiency of the
306 septic tile bed in attenuating wastewater constituents including nutrients (Mitchell et al.,
307 2021). Based on groundwater levels measured at PZ-1 ($x = 71$ m), there was ~ 1 m increase
308 in the groundwater table between 2014-2015 and 2020 (Figure 1c). However, with high
309 sand dunes behind the beach, there is a thick (3 - 4 m) vadose zone below the septic tile
310 beds at this study site (see Section 3.1.2 for details), and therefore it is not expected that
311 the higher groundwater table elevation in 2020 compared to 2014 – 2015 would have

312 adversely impacted the function of the vadose zone below the septic tile beds in attenuating
313 wastewater constituents. The adverse impact of increasing coastal levels on the attenuation
314 of wastewater constituents below the septic tile beds is expected to be a larger concern in
315 coastal plain settings with flatter topography and shallow groundwater tables.

316 The mean hydraulic gradient across nearshore aquifer was negative in 2014-2015 ($h = -$
317 0.015) and 2020 ($h = -0.017$), indicating lakeward groundwater flow. Based on the
318 hydraulic gradient in 2014-2015 and 2020, mean hydraulic conductivity ($K = 10$ m/d) and
319 effective porosity ($n = 0.3$) for the nearshore aquifer, the mean horizontal groundwater
320 velocity towards the lake is estimated to be 0.50 m/d in 2014-2015 and 0.56 m/d in 2020.
321 Numerical simulations show the change in groundwater flow patterns (Figure 4a-c) in the
322 nearshore aquifer between the 2014-2015 and 2020 sampling periods with the groundwater
323 discharge zone shifting from $x = 120$ -125 m in 2014-2015 to $x = 85$ -90 m in 2020. Based
324 on the estimated overall mean horizontal velocities and shoreline positions, the travel time
325 for conservative species to be transported from the septic tile bed to the shoreline is
326 estimated to be around 240 days in 2014-2015 compared to only 160 days in 2020.

327 While the flow patterns simulated by the steady state groundwater models do not indicate
328 recirculation of lake water through the nearshore aquifer (Figure 4a-c), recirculating flows
329 driven mostly by changing lake water levels and wave conditions are expected to occur
330 landward of the mean shoreline position and simulated groundwater discharge zone (Malott
331 et al., 2016). With the mean shoreline position and zone of groundwater discharge shifting
332 landward in 2020 compared to 2014-2015, the recirculation zone, which is generally
333 associated with high reactivity including the presence of mineral phases (e.g., Fe oxides)
334 that tend sequester pollutants (Lee et al., 2014; Lewandowski et al., 2015; Rakhimbekova

335 et al., 2018), would also have shifted landward. It is possible that this landward shift in the
336 recirculation zone could have altered geochemical conditions in the nearshore aquifer and
337 this is explored in the following sections.

338 **3.1.2 Characterization of septic-derived nutrient plume**

339 Elevated groundwater PO₄-P and NO₃-N concentrations were observed at the study site in
340 2014, 2015 and 2020 (Figures 4d to 4i). Groundwater PO₄-P concentrations ranged from 8
341 to 166 µg/L in 2014-2015, and from 3 to 98 µg/L in 2020. Maximum PO₄-P concentrations
342 in the nearshore aquifer exceeded the upper limit of the Canadian Water Quality Guidelines
343 for the Protection of Aquatic Life for Total Phosphorus to limit eutrophic conditions (35–
344 100 µg P/L; Canadian Council of Ministers of the Environment, 2014), and were
345 considerably higher than the mean measured lake water concentrations for all sampling
346 periods (2-3 µg/L). Groundwater NO₃-N concentrations ranged from 4 to 57 mg/L in 2014-
347 2015 and from 1 to 26 mg/L in 2020. Maximum NO₃-N concentrations in the nearshore
348 aquifer exceeded the Canadian Water Quality Guidelines for the Protection of Aquatic Life
349 for all sampling periods (3 mg/L; Canadian Council of Ministers of the Environment,
350 2014), and were also higher than the mean measured lake water concentrations (0.2-0.8
351 mg/L).

352 Figures 4j-1 to 4m-o presents the distributions of chloride (Cl) concentrations and electrical
353 conductivity (EC) measured along the monitoring transect in 2014, 2015 and 2020. Cl and
354 EC can be considered conservative wastewater tracers at the site(Rakhimbekova et al.,
355 2021a), and their distributions generally coincide with the locations of elevated PO₄-P and
356 NO₃-N in the nearshore aquifer Figure 4d-i. NO₃-N with EC landward of the maximum
357 shoreline position are correlated (Figure S1) indicating the suitability of using ERT for

358 characterizing the spatial extent of the high nutrient groundwater plume. Figure 5 shows a
359 2D cross-sectional ERT image that extends from $x = 0$ m (septic tile bed) to $x = 95$ m (near
360 the mean shoreline position in 2020). The location of the septic tile bed, PZ-1 and MLS are
361 indicated in the figure, along with the lake water and groundwater levels measured during
362 the survey. The highly resistive near surface layer between $x = 0$ m and $x = 78$ m (red-
363 purple; >1000 ohm-m) represents the vadose zone with a mean thickness of approximately
364 4 m. The underlying layer of low resistivity (blue-green; <150 ohm-m) represents the zone
365 of saturated groundwater, with some areas having very low resistivity (<50 ohm-m). The
366 ERT image can be validated by comparing bulk resistivity values with the porewater EC
367 measurements in Figure 4m-o. For example, the porewater EC at $x = 71$ m and $z = -3$ m
368 (PZ-1) is $1008 \mu\text{S}/\text{cm}$ which suggests a bulk EC of $209 \mu\text{S}/\text{cm}$, or bulk resistivity of 48
369 ohm-m (Archie, 1942) – this closely matches the bulk resistivity inferred by ERT at that
370 location (55 ohm-m). Figure 5 indicates that the zone of very low bulk resistivity is
371 connected to the base of the septic tile bed and extends lakeward. This zone exists through
372 the shallow nearshore aquifer and becomes shallower near the shoreline meeting the
373 sediment-water interface around $x = 92$ m. This ERT image clearly illustrates that the
374 elevated $\text{NO}_3\text{-N}$ and $\text{PO}_4\text{-P}$ concentrations observed near the shoreline originate from the
375 septic tile bed.

376 **3.1.3 Impact of shoreline recession on $\text{PO}_4\text{-P}$**

377 As seen in Figure 4, the highest $\text{PO}_4\text{-P}$ concentrations were observed 73-90 m lakeward of
378 the septic tile bed (maximum $\text{PO}_4\text{-P} = 166 \mu\text{g}/\text{L}$; $x = 73\text{-}90$ m) in 2014-2015, with
379 concentrations decreasing closer to the shoreline (particularly in 2015). However, elevated
380 concentrations of the conservative wastewater tracers, Cl and EC, near the shoreline ($x =$

381 115-125 m) for these sampling periods suggest that the septic-derived wastewater plume
382 has reached the shoreline. Taking into account septic system operation time, groundwater
383 velocity, and length of the PO₄-P plume in 2015, Rakhimbekova et al. (2021) concluded
384 that PO₄-P transport in the nearshore aquifer was mainly governed by retardation. Based
385 on the location of the PO₄-P plume in 2015, it was predicted that PO₄-P may possibly reach
386 the shoreline in approximately 10 years. However, this prediction is incorrect due to
387 shoreline recession at the site. Figure 4f shows that in 2020 (within <5 years), the PO₄-P
388 plume has reached the new shoreline location ($x = 90$ m). To provide a quantitative estimate
389 on the extent to which PO₄-P fluxes to the lake may have changed, the measured PO₄-P
390 concentrations were combined with the simulated groundwater flows for each sampling
391 period. PO₄-P fluxes were calculated by multiplying PO₄-P concentrations (interpolated to
392 a depth of 3 m below ground surface) along vertical line near the shoreline ($x = 120.3$ m in
393 2014 and 2015 and $x = 88.7$ m in 2020; Figure 4 a-c) by simulated groundwater fluxes
394 directed towards the lake for each sampling year (Additional details provided in SM S3).
395 From these calculations, it is estimated that the PO₄-P flux to the lake may have increased
396 nearly tenfold from 53 mg/d/m in 2015 to 482 mg/d/m in 2020 (Figure 6a) due higher PO₄-
397 P concentrations near the shoreline in 2020 compared to near the shoreline in 2015 as well
398 as slightly higher lakeward-directed groundwater velocity (Figure 4e, f). This result
399 illustrates that the shoreline recession can accelerate the discharge of groundwater-derived
400 pollutants that are slowly advancing towards the shoreline by decreasing the subsurface
401 travel distance and time.

402 **3.1.4 Impact of shoreline recession on NO₃-N**

403 The NO₃-N distribution in the nearshore aquifer was influenced by not only its shorter
404 discharge pathway through the nearshore aquifer due to shoreline recession but also by the
405 specific geochemical conditions near the sediment-water interface that impact reactive
406 transformations that NO₃-N may undergo prior to its discharge. Rakhimbekova et al.
407 (2021a) previously showed that the extent of NO₃-N removal that occurs in the nearshore
408 aquifer prior to discharge may vary over time as the extent of denitrification changes in
409 response to changing geochemical conditions near the sediment-water interface (e.g., due
410 to changes in organic matter availability). Rakhimbekova et al. (2021a) presented detailed
411 data analysis showing changing organic matter availability was associated with the
412 observed decrease in NO₃-N concentrations near the shoreline in 2015 (0-2 mg/L)
413 compared to 2014 (4-67 mg/L), and thus lower estimated NO₃-N flux towards the lake in
414 2015 (4 mg/d/m) compared to 2014 (124 mg/d/m) (Figure 6b).

415 NO₃-N concentrations observed near the mean shoreline position in 2020 ($x = 85-90$ m)
416 ranged from 4 to 26 mg/L in 2020 (Figure 4i). The estimated NO₃-N flux directed towards
417 the lake near the shoreline in 2020 (79 mg/d/m) was larger than in 2015 (4 mg/d/m) but
418 smaller than in 2014 (124 mg/d/m). NO₃-N flux is affected by both groundwater velocity
419 and NO₃-N concentrations. Since groundwater velocity near the shoreline was slightly
420 higher in 2020 compared to 2014 (difference in flow arrow sizes in Figure 4 a-c), the higher
421 estimated NO₃-N flux in 2014 is due to higher NO₃-N concentrations near the shoreline in
422 2014 compared to 2020. Therefore, even though shoreline recession reduced the travel
423 distance and travel time for the septic-derived NO₃-N to reach the lake, the specific
424 geochemical conditions in the nearshore aquifer near the shoreline ultimately govern the

425 flux of $\text{NO}_3\text{-N}$ to the lake rather than the overall travel distance and time. This finding is
426 in contrast to the highly retarded $\text{PO}_4\text{-P}$ plume for which shoreline recession was found to
427 directly accelerate $\text{PO}_4\text{-P}$ discharge to the lake due to the overall reduced travel distance
428 between the septic tile bed and shoreline.

429 It is important to note that while the geochemical conditions near the shoreline have a
430 controlling influence on the fate of $\text{NO}_3\text{-N}$ in the nearshore aquifer and its flux to the lake
431 at our study site, this may not be the case at other sites. The nearshore aquifer at our study
432 site (from the septic tile bed to shoreline) is predominantly oxic and therefore limited
433 denitrification occurs as $\text{NO}_3\text{-N}$ is transported towards the shoreline. At other sites where
434 the nearshore aquifer is more reducing, it is possible that the decrease in overall travel
435 distance and times could limit the extent of denitrification that occurs as $\text{NO}_3\text{-N}$ is
436 transported from the source to the coastal water (Meile et al., 2010). Further, it is possible
437 that shoreline recession could have a greater impact on the discharge of reactive pollutants
438 in nearshore aquifers with sharper geochemical (redox and pH) gradients than observed at
439 Ipperwash Beach (Rakhimbekova et al., 2021a, 2018).

440 ***3.1.5 Impact of shoreline recession on Fe curtain processes***

441 In addition to modifying the physical transport pathways and travel times for the septic-
442 derived nutrients being transported towards to the lake, the change in shoreline location
443 may also modify the geochemical conditions near the groundwater-lake interface. In
444 particular, it may modify the distribution, abundance and reactivity of mineral phases that
445 can sequester pollutants in the nearshore aquifer. Rakhimbekova et al. (2021b) showed
446 elevated solid phase Fe and P exists in the nearshore aquifer at the study site and suggested
447 that $\text{PO}_4\text{-P}$ derived from the decomposition of lake-derived organic matter may be

448 accumulating on Fe oxide minerals that exist close to the sediment-water interface.
449 Previous studies in marine environments have similarly found that Fe oxides that form near
450 the sediment-water interface can act as a geochemical barrier (i.e. “iron curtain”) with these
451 minerals sequestering pollutants such as PO₄-P and limiting their release to coastal
452 waters(Charette and Sholkovitz, 2002; Spiteri et al., 2006). While it is thought that
453 sequestered pollutants may later be re-mobilized if geochemical or hydrological conditions
454 change (Nisbeth et al., 2019; Spiteri et al., 2006), in this section we examine how the
455 distribution and abundance of solid phase P and Fe, including Fe oxides, in the nearshore
456 aquifer may have changed in response to shoreline recession.

457 Rakhimbekova et al. (201b) previously showed that nearshore aquifer at the study site is
458 predominantly oxidizing, and there is no distinct redox gradient near the sediment-water
459 interface. Further, Fe oxides are available through the nearshore aquifer and tend to
460 accumulate PO₄-P on their mineral surfaces. Characterization of the distribution of solid
461 phase P and Fe in the nearshore aquifer at the study site in 2015 and 2020 indicate a
462 decrease in solid phase P and Fe between these times (Figure 7a-d). Due to the landward
463 movement of the shoreline, sediment samples were taken in different locations in 2015
464 compared with 2020. To enable comparison of P and Fe solid phase concentrations
465 between two times, comparison only considers samples collected over the same area,
466 between $x = 87$ and 117 m (marked with red square on Figure 7). Generally, there was
467 higher abundance of solid phase P near the shoreline in 2015 compared to 2020 (151 ± 51
468 $\mu\text{g/g}$ in 2015, $n = 17$; $82 \pm 55 \mu\text{g/g}$ in 2020, $n = 23$; Figure 7a and b). Solid phase Fe was
469 also slightly higher in 2015 compared to 2020 ($3.1 \pm 0.6 \text{ mg/g}$ in 2015, $n = 17$; 2.7 ± 0.7
470 mg/g in 2020, $n = 23$; Figure 7c and d).

471 Selective solid phase extractions were conducted to characterize Fe-bearing solid phases
472 (amorphous, Fe_o and more crystalline, Fe_d) at the study site in 2015 and 2020. Again, only
473 sediment samples collected between $x = 87$ and 117 m were used to compare the
474 distribution of amorphous and crystalline Fe phases in 2015 and 2020. The abundance of
475 Fe_o decreased between 2015 and 2020 (0.5 ± 0.2 mg/g in 2015, $n = 10$; 0.2 ± 0.09 mg/g in
476 2020, $n = 13$; Figure 7e and f). In contrast, Fe_d was slightly higher in 2020 compared to
477 2015 (0.5 ± 0.07 mg/g in 2015; 0.7 mg/g ± 0.3 in 2020; Figure 7g vs 7h). Lower Fe_d in
478 2015 compared to 2020 may possibly be due to the formation of more crystalline Fe phases
479 over time. Larger difference in the abundance of Fe_o compared to Fe_d between 2015 and
480 2020 suggest that shoreline recession may have larger impact on amorphous Fe oxides
481 compared to crystalline Fe oxides. This is not unexpected given that amorphous Fe oxides
482 are less stable and shoreline recession may have altered factors that affect Fe oxide stability
483 (e.g. oxygen availability, presence of bacteria) (Charette and Sholkovitz, 2002; Dean et al.,
484 2003). Following shoreline recession, particularly a large erosive event, it might take time
485 for amorphous Fe oxides to reform around the new shoreline location. This may explain
486 the lower Fe_o near the new shoreline location in 2020 compared to higher Fe_o near the
487 location of shoreline in 2015, which was a more stable shoreline position over the
488 preceding years.

489 Amorphous Fe (e.g. ferrihydrite) tend to have a higher surface area that is more reactive
490 and therefore has a higher capacity to sequester pollutants, including PO₄-P, compared to
491 more crystalline Fe oxides fractions such as goethite and hematite (Fuller et al., 1993).
492 Therefore, lower abundance of Fe_o in the nearshore aquifer in 2020 (following the shoreline
493 recession) suggests that mineral surfaces along the groundwater discharge pathway may

494 have less ability to sequester PO₄-P, and this may possibly lead to higher PO₄-P fluxes to
495 lake. The impact of shoreline recession on the distribution and abundance of Fe oxide
496 minerals, particularly amorphous Fe oxides, is important as it may affect the function of
497 these minerals in sequestering and releasing pollutants thereby modifying pollutant fluxes
498 to coastal waters.

499 **3.2 Broader implications of the impact of shoreline recession**

500 The study findings demonstrate the potential impacts of shoreline recession driven by
501 coastal water level changes and/or erosion on the transport and fate of groundwater
502 pollutants and their discharge to adjacent coastal waters. The findings show shoreline
503 recession may impact pollutants moving slowly along groundwater pathways towards the
504 coastal water, as well as pollutants for which their fate and ultimate delivery to coastal
505 waters is governed strongly by the geochemical conditions in the reaction zone that exists
506 near the groundwater-coastal water interface. This section explores the broader
507 implications of the study findings on the discharge of groundwater pollutants to the
508 Laurentian Great Lakes, as well as other inland and marine coastal waters.

509 While it is known that water levels in the Great Lakes vary over 20 – 30 year cycles
510 (Gronewold and Rood, 2019), recent future projections estimate that by 2040-2049 average
511 annual water levels of Lake Superior, Lake Michigan-Huron and Lake Erie are projected
512 to increase by +0.19, +0.44, +0.28 m, respectively due to an increase in over-lake
513 precipitation and basin runoff compared to relatively smaller increase in lake evaporation
514 (Kayastha et al., 2022). While there are no studies predicting future erosion rates and
515 shoreline recession in response to the projected water level changes for the Great Lakes,
516 several studies show that high water levels correspond with accelerated shoreline erosion,

517 including bluff erosion, on short time scales (e.g. Kaczmarek et al., 2016; Krueger et al.,
518 2020). For instance, Castedo et al. (2013) simulated erosion rates to range between 0.5-1.5
519 m/yr in response to lake water level variations of 0.5-1 m. A more recent study by Volpano
520 et al. (2020) on coastal bluff erosion along Western Lake Michigan's shoreline found that
521 erosion rates increased from 0.18-0.43 m/yr between 2009 and 2014 (period with below
522 long term average water levels) to 0.49-1.19 m/yr between 2014 and 2018 (period with
523 above long-term average water levels).

524 The outlook for marine shorelines is also concerning. On average, global sea level has risen
525 at a rate of 3-4 mm/yr in the last 20 years (Yi et al., 2015). Future projections estimate sea
526 level rise from 0.1-0.2 m by 2050 and from 0.3-2 m by 2100 due to ocean warming and
527 land-based ice melt (Kopp et al., 2014; Vitousek et al., 2017). While shoreline recession is
528 generally the combined result of various contributing factors and may be complicated by
529 local and regional differences, it is widely acknowledged that increasing sea levels will
530 promote shoreline recession (e.g. Anderson et al., 2015; Bruun, 1962; Dean and Houston,
531 2016). For instance, a recent study predicted that 15% of world's sandy beaches could face
532 severe coastal erosion by 2050 and up to 50% by the end of the century (Vousdoukas et
533 al., 2020). Taking into consideration the projected sea level changes and associated
534 shoreline recession, together with the findings of our study, there is high potential that
535 future shoreline recession will modify inputs of groundwater pollutants to coastal waters.

536 While this study focuses on the wastewater plume from one septic system, there are 1.2
537 and 1.3 million septic systems in Ontario, Canada (DeRabbie, 2020) and Michigan, United
538 States (Department of Environment, Great Lakes and Energy, 2022), respectively, with
539 many of these septic systems located in close proximity to the shorelines of the Great Lake.

540 It is also important to note that in addition to nutrients, septic-derived wastewater effluent
541 also contains other pollutants of concern including pharmaceuticals, personal care products
542 and pathogens, all of which may pose a risk to coastal water pollution with associated
543 human health and ecosystem impacts. Further, many other groundwater contaminant
544 sources are often located in coastal areas whereby shoreline recession may modify and
545 potentially accelerate pollutant inputs to coastal waters. This includes landfills (both
546 historic and currently operating) (Brand et al., 2018; Stefania et al., 2019) as well as
547 industrial sites including Superfund sites which are the most contaminated sites in the
548 United States (Carter, 2020).

549 **4 Conclusions**

550 This study showed the impact of shoreline recession in modifying the fate of groundwater
551 pollutants discharging to coastal waters at a specific study site, and discussed the broader
552 potential implications for inland and marine coastal waters. The study findings show that
553 the shoreline recession may accelerate the discharge of groundwater pollutants to coastal
554 waters by decreasing the subsurface travel distance and travel time. In addition, shoreline
555 recession may modify reactive processes close to the sediment-water interface, which may
556 in turn affect the fate of reactive pollutants and their discharge to coastal waters.
557 Importantly, shoreline recession may result in a landward shift of the recirculation zone
558 that commonly exists close to the shoreline, and in doing so, alter the stability and
559 abundance of sediment phases (e.g., Fe oxides) that often act to sequester pollutants and
560 potentially limit their release to coastal waters. In particular, the study findings show that
561 amorphous Fe oxides may be particularly impacted by the shoreline recession and may

562 take time to reform and function to sequester pollutants along the modified groundwater
563 discharge pathway.

564 As it is expected that climate change will accelerate shoreline recession on coastlines
565 worldwide due to increase in the magnitude of lake water level fluctuations, sea level rise,
566 as well as increased frequency and intensity of storm events, it is recommended that future
567 work evaluate the impact of shoreline changes of different time scales (e.g., event based,
568 yearly, decadal) on pollutant fate in nearshore aquifers. The nearshore aquifer at the study
569 site was relatively homogeneous and predominantly oxidizing and it is recommended that
570 future studies evaluate the impact of shoreline recession in different nearshore aquifer types
571 including more heterogeneous aquifers and those with more reducing conditions. Further,
572 while this study focused on a freshwater coastal aquifer, field investigations are required
573 to determine the impact of shoreline recession on the functioning of the reaction zone in
574 nearshore aquifers along marine shorelines (i.e., subterranean estuary) and the
575 corresponding impact on pollutant fluxes to marine waters. As the study data also
576 demonstrate that the abundance and form of solid phase Fe near the shoreline may vary in
577 response to shoreline change, there is a need to better understand the mineralization process
578 for Fe oxides to better predict the impact of future changes on their functioning especially
579 with respect to sequestering and releasing pollutants of concern. While this study focused
580 on the impact of shoreline recession on nutrients derived from a septic system, it is
581 recommended that future studies explore the potential impacts of shoreline recession on
582 the discharge of other anthropogenic pollutants of concern that are commonly elevated in
583 coastal groundwater systems.

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