Science of the Total Environment 846 (2022) 157336

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Salinization as a driver of eutrophication symptoms in an urban lake (Lake Wilcox, Ontario, Canada)

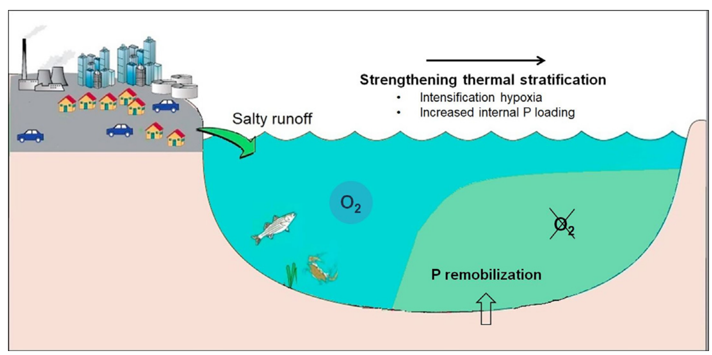
Jovana Radosavljevic [a,*], Stephanie Slowinski [a], Mahyar Shafii [a], Zahra Akbarzadeh [a], Fereidoun Rezanezhad [a,b], Chris T. Parsons [a,c], William Withers [d], Philippe Van Cappellen [a,b]

[a] Ecohydrology Research Group, Department of Earth and Environmental Sciences, University of Waterloo, Ontario, Canada
[b] Water Institute, University of Waterloo, Ontario, Canada
[c] Watershed Hydrology and Ecology Research Division, Canada Centre for Inland Waters, Environment and Climate Change Canada, Burlington, Ontario, Canada
[d] City of Richmond Hill, Ontario, Canada

HIGHLIGHTS

- Watershed urbanization causes salinization of Lake Wilcox.
- Salinization intensifies lake stratification and internal phosphorus (P) loading.
- Symptoms of eutrophication worsen even under declining external P loading.
- Reducing road salt use should be a priority to protect cold climate urban lakes.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Jay Gan

Keywords:
Lake water chemistry
Watershed urbanization
Salinization
Stratification
Nutrients
Phosphorus
Internal loading
Eutrophication

ABSTRACT

Lake Wilcox (LW), a shallow kettle lake located in southern Ontario, has experienced multiple phases of land use change associated with human settlement and residential development in its watershed since the early 1900s. Urban growth has coincided with water quality deterioration, including the occurrence of algal blooms and depletion of dissolved oxygen (DO) in the water column. We analyzed 22 years of water chemistry, land use, and climate data (1996–2018) using principal component analysis (PCA) and multiple linear regression (MLR) to identify the contributions of climate, urbanization, and nutrient loading to the changes in water chemistry. Variations in water column stratification, phosphorus (P) speciation, and *chl-a* (as a proxy for algal abundance) explain 76 % of the observed temporal trends of the four main PCA components derived from water chemistry data. MLR results further imply that the intensity of stratification, quantified by the Brunt-Väisälä frequency, is a major predictor of the changes in water quality. Other important factors explaining the variations in nitrogen (N) and P speciation, and the DO concentrations, are watershed imperviousness and lake chloride concentrations that, in turn, are closely correlated. We conclude that the observed in-lake water quality trends over the past two decades are linked to urbanization via increased salinization associated with expanding impervious land cover, rather than increasing external P loading. The rising salinity promotes water column stratification, which reduces the oxygenation of the hypolimnion and enhances internal P loading to the water column. Thus, stricter controls on the application and runoff of de-icing salt should be considered as part of managing eutrophication symptoms in lakes of cold climate regions.

* Corresponding author at: Ecohydrology Research Group, Department of Earth and Environmental Sciences, University of Waterloo, 200 University Avenue West, Waterloo, Ontario N2L 3G1, Canada.
E-mail address: jradosav@uwaterloo.ca (J. Radosavljevic).

http://dx.doi.org/10.1016/j.scitotenv.2022.157336
Received 12 May 2022; Received in revised form 4 July 2022; Accepted 9 July 2022
Available online 18 July 2022

1. Introduction

Urbanization is a global phenomenon. In 2007, half of the world's population lived in urbanized area; this fraction is expected to exceed 65 % by 2050 (McGrane, 2016). Urbanization negatively impacts biodiversity, water quality and air quality (McDonald et al., 2014; Zhao et al., 2006). Regardless of the magnitude of urban development, these impacts are generally significant (Brabec et al., 2002; Paul and Meyer, 2001; Walsh et al., 2005). Increased imperviousness in urban areas can enhance the mobility of contaminants (Garnier et al., 2013; McGrane, 2016; Pringle, 2001), causing deterioration of downstream water quality (Jacobson, 2011; Shuster et al., 2005; Walsh et al., 2005).

Examples of common urban contaminants that negatively impact water quality and aquatic life of urban lakes and streams include toxic metals, such as zinc, lead, cadmium, and nickel (Flores-Rodrîguez et al., 1994; Mansoor et al., 2018), and polycyclic aromatic hydrocarbons (PAHs) (Zehetner et al., 2009). Along with excess (Pastor and Hernández, 2012) and thermal pollution (Wang et al., 2008), nutrients represent another important class of urban contaminants (Carey et al., 2013; Garnier et al., 2013) that may result in eutrophication of the receiving water bodies (Carpenter, 2005; Carpenter et al., 1998; Filippelli, 2008; Frossard et al., 2014; Jenny et al., 2016; Tromboni and Dodds, 2017).

Phosphorus (P) is often considered to be the main limiting nutrient responsible for eutrophication in freshwater lakes (Schindler et al., 2016). Excessive nutrient P loading leads to increased primary productivity and oxygen (O_2) depletion, especially during periods of water column stratification. Jenny et al. (2016) reported that intensification of hypoxic conditions in European lakes is historically correlated with the growth of nearby urban areas and the export of P from urban point sources. In addition to the increased frequency of algal blooms and hypoxic conditions caused by algae decomposition (Carey et al., 2013; Heisler et al., 2008), eutrophication may also be accompanied by the demise of submerged vegetation and the release of toxins by harmful algal species that negatively impact fish, wildlife, pets and even humans (Carey et al., 2013; Yang et al., 2008).

In cold temperate regions, direct road salt runoff also represents a growing and multi-faceted problem for lentic ecosystems. Elevated chloride (Cl^-) and other dissolved ion concentrations from de-icing agents impact biodiversity and food webs, alter base cation balances, and modify the mixing regime of lakes that, in turn, can cause oxygen depletion of the hypolimnion (Corsi et al., 2015; Dugan et al., 2017a, 2017b; Koretsky et al., 2012; Ladwig et al., 2021).

Urban stormwater management (SWM) systems are implemented to control peak runoff and mitigate contaminant export, including that of nutrients, to surface and groundwater (Hathaway et al., 2012; Song et al., 2015, 2017). These systems include both grey infrastructure, such as stormwater ponds, and green infrastructure such as bioretention cells. These facilities help restore more natural hydrological conditions that are considered more efficient for the mitigation of urban contaminants (Goh et al., 2019; Hathaway et al., 2012; Kratky et al., 2017; Song et al., 2015, 2017; Yang and Toor, 2018). Despite their success in reducing runoff (Jefferson et al., 2017), it is still unclear whether SWM facilities permanently immobilize P (Perry et al., 2009). In fact, some stormwater ponds have been found to increase the export of the more bioavailable forms of P (Frost et al., 2019; Song et al., 2015, 2017).

Along with urbanization, climate change represents a potential stressor on water quality and aquatic ecosystem health. Cold and cold-temperate regions, including Canada, are warming rapidly, and this trend is expected to continue in the future, both at annual and seasonal scales (Lemieux and Scott, 2011; Vincent et al., 2018). Climate change is accompanied by hydrological changes, such as more frequent high-intensity precipitation events (Clifton et al., 2018; Stocker, 2014; Tebaldi et al., 2006; Whan et al., 2016). Extreme events, for instance heavy rainfall, windstorms, heat and cold waves, and droughts are forecasted to become the new normal in the coming decades (Emilsson and Sang, 2017; Forzieri et al., 2018; Kaushal et al., 2014; Zhang et al., 2008). In urban landscapes, heavy rainfall events create higher peak runoff flows with high pollutant loads of total suspended solids (TSS), nutrients, and toxic metals (Alamdari et al., 2020; Ekness and Randhir, 2015). Longer dry periods between precipitation events (Stocker, 2014) can favor sediment buildup in urban landscapes and cause the flushing of high TSS concentrations during subsequent runoff events (Sharma et al., 2016).

To understand the impacts of land use and climate change on water quality, previous studies have focused on individual lakes (Timothy Patterson et al., 2002) or groups of lakes (Seilheimer et al., 2007; Tomer and Schilling, 2009; Yang and Chang, 2007) using modeling (Carpenter, 2005) and data analyses (O'Reilly et al., 2015; Su et al., 2021; Taranu and Gregory-Eaves, 2008; Yang et al., 2020a). These studies typically rely on comprehensive but relatively short data time series, usually covering a year or two. Here, we investigate water quality changes in a lake using a multi-variate dataset collected over two decades. Lake Wilcox (LW) in southern Ontario, Canada is a kettle lake within an urbanizing watershed. Our goals are to delineate general trends in water quality in LW during the period 1996–2018 and identify drivers of these trends using statistical analysis methods.

2. Methods

2.1. Study site: Lake Wilcox

Lake Wilcox (LW) is a small kettle lake located in the city of Richmond Hill in Ontario, Canada, along the northern edge of the greater Toronto metropolitan area (Fig. 1). It is divided into an east and west basin that are 17 m and 14 m deep, respectively. The two basins are separated by a ridge which peaks at ~8 m water depth (Reports number 4, 5 and 6 in Table S1). The lake's surface area is 0.56 km^2 and its watershed area is 2.39 km^2. Nearby Lake St. George (watershed area: 2.5 km^2) drains into LW via a connecting channel. The high morphometric ratio (7.6 m/km) promotes stratification of LW during the summer (Report number 5 in Table S1). The lake has one outflow and seven inflow points, of which the St. George and North Shore inflows contribute the highest flows (~85 ± 5 %) to the lake (Report number 6 in Table S1). Based on discharge measurements for the period 1998–2010, the average annual surface inflow is on the order of 7×10^5 m^3 and the water retention time is approximately 2 years (Reports number 4 and 5 in Table S1).

The lake's watershed experiences a continental climate moderated by the Laurentian Great Lakes. The watershed is influenced by warm, moist air masses from the south and cold, dry air masses from the north (TRCA, 2008). The average total annual precipitation is about 853 mm (range: 700 to 1000 mm) and the average daily temperature 7.7 °C (Government of Canada, 2019). The majority of precipitation occurs during the summer months (June, July, and August). The mean annual evapotranspiration is approximately 517 mm per year (TRCA, 2018).

The originally forested watershed of LW has undergone significant development since the early 1900s when the land was converted to agriculture. This was followed by the construction in the 1940s of nearshore cottages on septic systems. The latter were decommissioned in the 1960s when monitoring revealed rising eutrophication of the lake (Reports number 1, 2 and 3 in Table S1). Sub-urban and urban development took off in the 1980s, further accelerating in the early 2000s. As a result of development, land surface imperviousness increased to over 60 % at present (Fig. 2a). The imperviousness values (in % watershed land cover) from 1996 to 2018 shown in Fig. 2a are based on estimates reported in previous studies for the years 1996, 2009, 2012, and 2018 (Reports number 1–6 in Table S1; Baker et al., 2017; Doyle and Macpherson, 2017; OMAFRA, 2019), complemented with our own estimates obtained manually from aerial images. Values for the other years were interpolated as done in previous studies (Exum et al., 2005; Li and Wu, 2016; Wu and Hung, 2016). Note that Fig. 2a shows the total imperviousness, that is, it corresponds to the sum of all impervious land parcels in Lake Wilcox's watershed. Thus, it comprises both the disconnected and directly connected impervious land cover. The directly connected imperviousness is the sum of impervious land parcels that are connected to the storm sewer network.

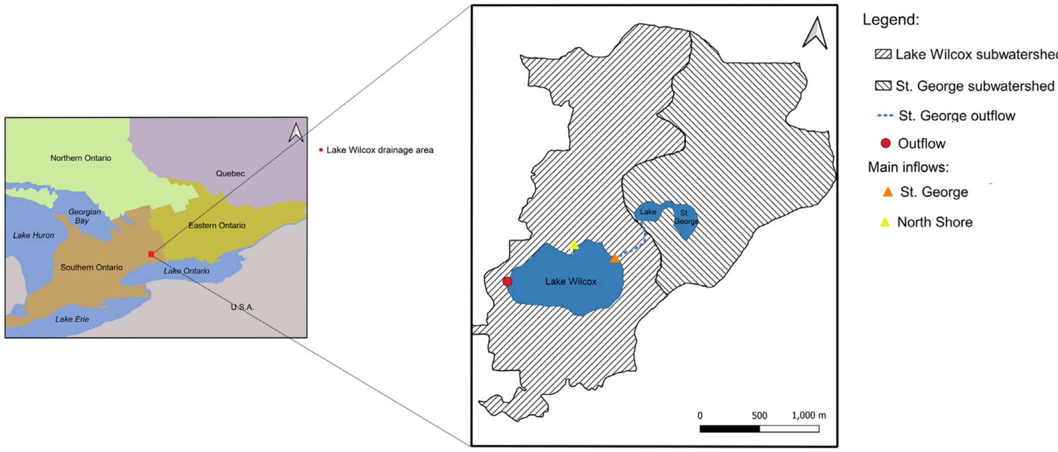


Fig. 1. Location of Lake Wilcox in Ontario, Canada (a), and sketch of its drainage area (b).

By the 1980s, worsening eutrophication, including declining dissolved oxygen (DO) levels and the appearance of cyanobacteria, had become a major issue, with the lake becoming meso-eutrophic (Reports number 4 and 5 in Table S1; Nürnberg, 1997; Nürnberg, 1996). To combat increasing algal productivity and avoid harmful algal blooms (HABs) an aeration system was installed in 1998 with the goal of slowing down internal P loading from the lake sediments to the water column – a process known to be a major driver of algal growth (Olding, 2012; Report number 5 in Table S1). The aerator, however, broke down in early 2001 and was never reactivated (Olding, 2012). Due to the lack of improvement in LW's water quality, a suite of SWM systems, notably wet ponds, were constructed in the watershed, starting in the early 2000s. Over time, more and more of the urban and suburban developments were connected to the storm sewer system and, as a result, the expansion of imperviousness from around 2010 till today saw an increase in the fraction of directly connected imperviousness (William Withers, personal communication, September 2020).

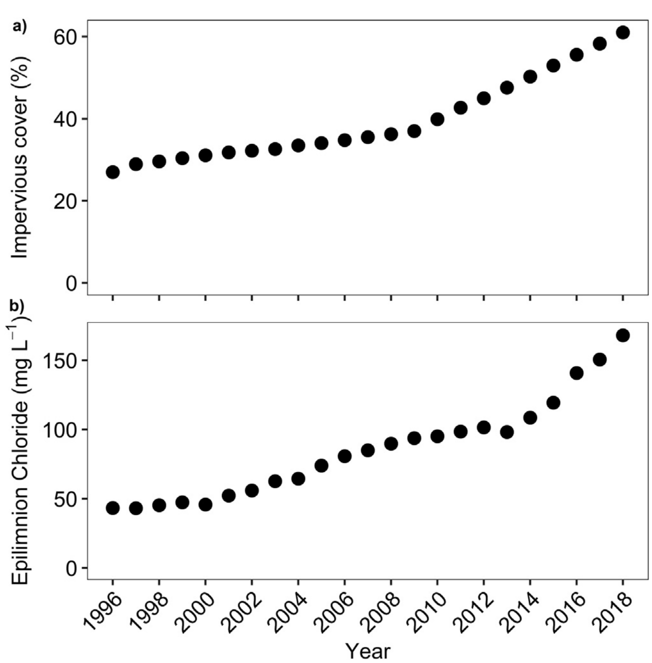


Fig. 2. Fraction of impervious land cover in Lake Wilcox's watershed (a) and in-lake surface water chloride concentrations (b) from 1996 to 2018.

2.2. Datasets

A variety of datasets were collected from different sources, including water quality monitoring data provided by the City of Richmond Hill (CRH), climate data downloaded from the Environment and Climate Change Canada (ECCC) and Toronto and Region Conservation Authority (TRCA) websites (Table S2), as well as satellite images obtained from Esri's Wayback Living Atlas Google Earth Pro and USGS Land Look. Data were synthesized for the period 1996 to 2018. From the climate records we derived values for the mean daily temperature, average yearly temperature, mean daily and yearly precipitation, and the 95th percentile of rainfall (calculated following U.S. Environmental Protection Agency guidelines (USEPA, 2009), see also Shrestha et al., 2013).

The CRH water quality dataset comprised results of bi-weekly sampling from May to November, from 1996 to 2018. Included were concentrations of dissolved oxygen (DO), total P (TP), dissolved inorganic P (DIP), nitrate (NO_3^-), ammonium (NH_4^+), dissolved inorganic nitrogen (DIN), and chloride (Cl^-) in both the epilimnion and hypolimnion, plus the concentrations of chlorophyll-a (*chl-a*) in the epilimnion. The annual TP loadings to the lake were derived from the discharge and TP concentration values in the CRH reports following the flow-weighted averaging method described in Walling and Webb (1985) and Moatar and Meybeck (2005).

Lake Wilcox was selected because of the availability of >20 years of uninterrupted data on water chemistry that overlap with the period during which the watershed experienced a significant urban development. That is, the lake provides a unique setting to relate water quality changes with urbanization of the catchment.

2.3. Anoxic factor (AF)

The DO time series data served to calculate anoxic factor values following the method proposed by Nürnberg (1996, 2004):

$$AF = \frac{\sum_{i=1}^{n} t_i \times a_i}{Ao} \tag{1}$$

where AF is the anoxic factor (in days) for a given year, t_i is the number of days of anoxia during that year or season, a_i is the hypolimnetic surface area (m^2), and Ao is lake surface area (m^2). The appearance of Ao in the denominator of Eq. (1) accounts for the variable lateral extent of anoxic bottom waters. The annual AF value thus corresponds to the number of days in a year during which a sediment area equal to the lake surface area is overlain by anoxic water. Annual AF values are a proxy for changes in the intensity of deoxygenation of the hypolimnion.

2.4. Brunt-Väisälä frequency (BVF)

The Brunt-Väisälä or buoyancy frequency (BVF) represents the amount of work required against gravity to break down thermal stratification in the water column (Mortimer, 1974a). It is calculated as follows:

$$N^2 = \frac{-g}{\bar{\rho}} \times \frac{\partial \rho}{\partial z} \quad (2)$$

where N is the BVF (s^{-1}), g is the gravitational acceleration ($m\ s^{-2}$), $\bar{\rho}$ is the average water density over the entire water depth considered ($kg\ m^{-3}$), ρ is the water density ($kg\ m^{-3}$) at a given depth z (m) (Mortimer, 1974a; Zadereev and Tolomeyev, 2007). Note that the use of Eq. (2) yields a depth profile of BVF.

The density of water was calculated from the corresponding temperature and salinity values with the Javascript calculator developed by the collaborative Computer Support Group (CSG) Network of University of Michigan and National Oceanic and Atmospheric Administration (NOAA). For those sampling times where Cl^- concentrations were reported for both the epilimnion and hypolimnion, little vertical difference in the Cl^- concentrations was observed. Because Cl^- concentrations were measured systematically in the epilimnion, but not the hypolimnion, only the epilimnion Cl^- concentrations were used and converted to salinity values (Dugan et al., 2017a, 2017b; Shambaugh, 2008). For the calculation of the BVF depth profile at a given sampling time, the same salinity value was then applied to all depths while the measured depth-dependent temperature profiles were imposed. For details on the water density calculation, see Gill (1986).

In order to assess interannual variability in the intensity of stratification, we compared BVF depth profiles calculated for the month of August for each year. In southern Ontario, July–August exhibit the highest monthly air temperatures and water column stratification is most pronounced. In addition, by August, the lake's temperature distribution retains negligible memory from the preceding spring mixing event.

2.5. Statistical analyses

We relied on multiple statistical methods to describe and interpret water quality changes. We used the Mann-Kendall (MK) test to assess the statistical significance of temporal trends in water quality variables (with $p < 0.1$ as threshold, Section 2.5.1). We further employed Principal Component Analysis (PCA) to reduce the dimensionality of the large water quality data set (Section 2.5.2). We also developed Multiple Linear Regression (MLR) models to explore the relationships between the principal components (PCs) and potential predictor variables (e.g., degree of urbanization, salinization, temperature, nutrient concentrations and speciation, etc.).

2.5.1. Mann-Kendall test (MK) and Kendall's τ

The MK tests were carried out with the XLSTAT software (Addinsoft, version 2022). Positive values of the MK statistic indicate an upward trend, negative values a downward trend, and a zero value no change over time. The larger the MK statistic deviates from zero, the greater the evidence for a significant temporal trend in the data series (Hirsch et al., 1982; Kendall, 1975; Lettenmaier, 1988; Şen, 2014). Here, we report the closely related Kendall's τ (tau) values that rescale the MK statistic values within the range -1 to $+1$, which facilitates the comparison of data series with variable numbers of observations (Helsel and Hirsch, 1992; Meals et al., 2011). More details about the MK test can be found in Hirsch et al., 1982; Kendall, 1975; and Zhang et al., 2004.

2.5.2. Principal components analysis (PCA)

To identify the drivers of water chemistry changes it is often helpful to group variables that are correlated. We selected PCA, which has been widely applied to environmental data time series (Arslan, 2013; Parinet et al., 2004; Sahoo et al., 2020; Wold et al., 1987; Yang et al., 2020b). The emerging groupings then create a new and smaller set of presumably uncorrelated variables, that is, the principal components (PCs). Details about PCA can be found in many previous works (Liu et al., 2019; Zou and Xue, 2018). The 13 water chemistry variables included in the PCA were epilimnion and hypolimnion concentrations of DIP, TP, NH_4^+, NO_3^-, DIN, and DO, plus the *chl-a* concentration in the epilimnion.

As required in PCA, we first filtered the dataset to remove missing data points. Water quality parameters that had non-existing values for >25 % of the sampling dates were removed. We further excluded data from 1997 to 2000 because during that time the DO concentrations were artificially manipulated by using an aerator (Section 2.1). The PCA was performed and visualized in R 4.1.2 (R Core Team, 2020) using the factoextra package (Kassambara and Mundt, 2020).

2.5.3. Multiple linear regression (MLR)

In the MLR analyses, the water chemistry PCs were treated as the dependent (or y) variables. The goal was to identify the (independent) explanatory variables that most contribute to the temporal trends of the water chemistry PCs. To compare the regression coefficients from the MLR analyses, we first normalized the values of each explanatory variable by that variable's mean value to make them unitless. The MLR analysis was performed with the JMP statistical software (SAS Institute Inc.) based on the following formulation:

$$y = b_0 + b_1 x_1 + b_2 x_2 + \ldots + b_i x_i \quad (3)$$

where y is the dependent variable, x_i the ith explanatory (or predictor) variable, b_o the regression constant, and b_i the ith coefficient of the corresponding explanatory variable (Brix et al., 2017; Liu et al., 2015; Weisberg, 2005).

The predictors included in the MLR analyses were: watershed imperviousness (as a measure of urbanization), lake Cl^- concentration (as a measure of deicing salt usage), external P loading to the lake, water column stratification (using the Brunt-Väisälä frequency), and the following climate parameters: 14-day average temperature and 14-day total rainfall prior to sampling, average yearly temperature and rainfall, and yearly 95 % percentile of rainfall. The latter is used as a measure of a year's extreme precipitation events that can play an important role in pollutant wash-off in urban areas (Carpenter et al., 2018; Knapp et al., 2008; Rahimi et al., 2021; Ummenhofer and Meehl, 2017; Vincent et al., 2018; Whan and Zwiers, 2016).

The explanatory variables selected are known drivers of water quality changes (Carey et al., 2013; Garnier et al., 2013; McGrane, 2016; Pringle, 2001; Pickett et al., 2011; Walsh et al., 2005). Obviously, the analyses can only provide insights into the roles of the selected explanatory variables. Other variables, such as lake ice cover and duration, are likely responsible for part of the temporal variability of the PCs (Carey et al., 2012; Cavaliere and Baulch, 2018; Rodgers and Salisbury, 1981), however, they are not included here because of the lack of a corresponding data time series.

3. Results

3.1. Time series trends

3.1.1. Dissolved oxygen (DO) and anoxic factor (AF)

Hypoxic conditions were first recorded in LW in the 1980s (Reports number 4 and 5 in Table S1). For the observation period considered here (1996–2018), average DO concentrations were 10.2 and 1.1 mg L^{-1} in the epilimnion and hypolimnion, respectively. Maximum and minimum DO concentrations were 17.8 and 2.4 mg L^{-1} in the epilimnion, and 12.0 and 0 mg L^{-1} in the hypolimnion. The time series DO depth profiles shown in Fig. 3 indicate a transition around the year 2005 characterized by a lengthening of the annual periods of DO deficient conditions in the hypolimnion (i.e., the dark blue color in the figure), sometimes extending throughout the entire year.

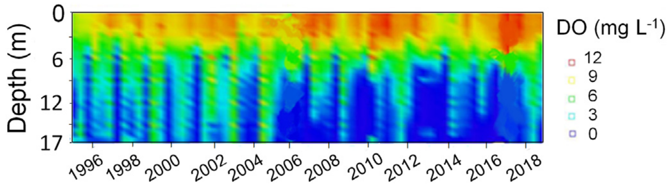


Fig. 3. Mid-lake depth distributions of dissolved oxygen concentrations (DO) between 1996 and 2018. Note the lengthening of the yearly period of anoxia (blue color) after 2003.

The depth of the redoxcline also increased, from around 4 m at the beginning of the data series to around 6 m in the most recent years. The DO trend analysis for LW revealed a statistically significant ($p < 0.1$) decrease in DO concentrations, especially in the hypolimnion (Kendall's $\tau = -0.35$) (Fig. 4). The average value of AF for the entire period (excluding years with artificial aeration) was 72 days per year. The annual AF exceeded 80 days in 2005, 2008 and 2015. As expected from the DO trend analysis, Fig. 4 shows an increasing trend of AF with time (Kendall's $\tau = +0.42$).

3.1.2. Chloride

The average Cl^- concentration in the epilimnion increased from 43 mg L^{-1} in 1996 to 175 mg L^{-1} in 2018 at which time the watershed's imperviousness exceeded 60 % (Fig. 2b, Table S3). Of all the water quality variables tested, the Cl^- time series showed the strongest positive trend with time (Kendall's $\tau = 0.94$, Fig. 4). Several distinct phases of Cl^- increase can be distinguished (Fig. 2b, Table S3). From 1996 to the turn of the century only a small increase of the Cl^- concentration was observed. This was followed by a much faster increase up to ~90 mg L^{-1} by the year 2008, while the watershed's imperviousness rose to 36 %. The next phase, from 2008 to 2012, was again characterized by a relatively slow growth of the Cl^- concentration as the imperviousness reached 45 %. The last phase saw the most rapid rise of the Cl^- concentration to about 175 mg L^{-1} in just six years. During that same period, major efforts were undertaken to expand directly connected imperviousness (Section 2.1).

3.1.3. Brunt-Väisälä frequency (BVF)

August BVF depth profiles are shown in Fig. 5 for the beginning and end of the observation period (1996 and 2018) plus intermediate year 2007.

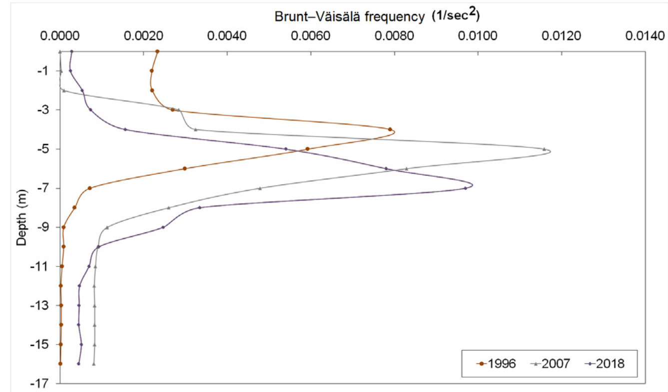


Fig. 5. Mid-lake Brunt-Väisälä frequency (BVF) depth profiles for the month of August in 1996, 2007, and 2018.

Overall, the shape of the profile became wider over the years while the maximum BVF values moved to deeper depths. The latter is consistent with the generally downward trend observed for the redoxcline (Fig. 3). As for Cl^-, maximum BVF values exhibited an increasing trend with time (Kendall's $\tau = +0.12$, Fig. 4).

3.1.4. Phosphorus: concentrations and speciation

Over the entire time period considered, the TP and DIP concentrations in the hypolimnion were about 10 and 30 times higher than those measured in the epilimnion, respectively. In the epilimnion the DIP varied from 0.001 to 0.059 mg L^{-1} with an average value of 0.005 mg L^{-1}, while TP concentrations ranged from 0.003 to 0.116 mg L^{-1} with an average value of 0.032 mg L^{-1}. The hypolimnion concentrations of DIP ranged from 0.001 to 0.513 mg L^{-1} with an average value of 0.175 mg L^{-1}, while TP varied from 0.017 to 1.33 mg L^{-1} and averaging at 0.249 mg L^{-1}. The epilimnion TP concentration showed a decreasing trend over time (Kendall's $\tau = -0.27$, Figs. 4 and 6, Table S3). The hypolimnion TP concentration similarly exhibited a decreasing trend, although less pronounced than that in the epilimnion (Kendall's $\tau = -0.24$, Figs. 4 and 6, Table S3). Since 2000, the DIP concentrations in the epilimnion and hypolimnion have experienced slight declines (Table S3).

The DIP:TP ratios in the epilimnion and hypolimnion exhibited statistically significant increasing trends over time (Kendall's $\tau = +0.13$ and $+0.23$, respectively in the epilimnion and hypolimnion, Fig. 4).

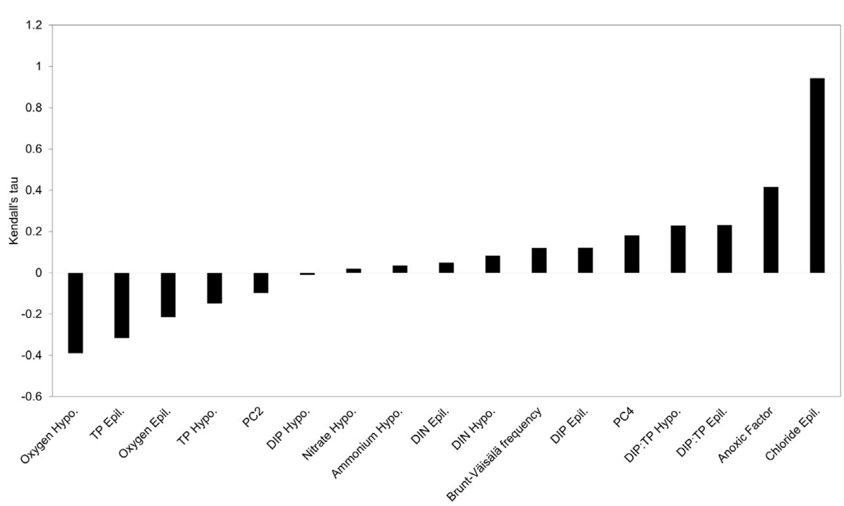


Fig. 4. Kendall's tau values for Mann-Kendall trend analyses. The figure includes the results for the water quality parameters, MLR explanatory variables, and principal components (PCs) that show a statistically significant ($p \leq 0.1$) trend with time over the period 1996 to 2018. Hypo = hypolimnion, Epi = epilimnion.

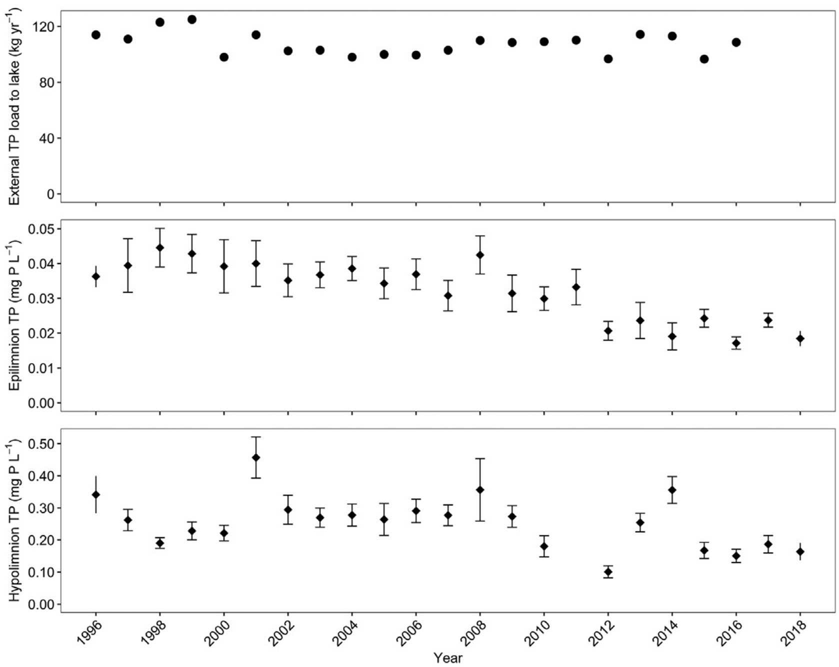


Fig. 6. Time series of total phosphorus (TP) external loads to Lake Wilcox, and in-lake TP concentrations in epilimnion and hypolimnion. The error bars represent standard deviation.

As can be seen in Fig. S1, a relatively large increase in DIP:TP occurred after 2010, particularly in the epilimnion. Further note that the minimum hypolimnion DIP:TP value of 1998 coincided with the time the aerator was in operation.

3.1.5. Nitrogen: concentrations and speciation

As for TP and DIP, the DIN (= sum of ammonium, nitrite and nitrate) concentrations in the hypolimnion were much higher than for the epilimnion (Table S3). The maximum DIN concentration in the epilimnion was 1.64 mg L^{-1} and the average value was 0.15 mg L^{-1}. The DIN concentrations in the hypolimnion reached a maximum value of 6.71 mg L^{-1}, with an average concentration of 1.54 mg L^{-1}. In both epilimnion and hypolimnion, the DIN concentrations showed increasing trends (Kendall's $\tau = +0.12$ and $+0.27$ for epilimnion and hypolimnion, respectively, see also Fig. 4).

The speciation of DIN also differed markedly between the epilimnion and hypolimnion (Table S3). In the hypolimnion, nearly all DIN was under the form of NH_4^+. The average NO_3^- concentration in the hypolimnion was 0.04 mg L^{-1} while the average concentration of NH_4^+ was 1.50 mg L^{-1}. In the epilimnion, the average concentrations of NO_3^- and NH_4^+ were 0.05 and 0.10 mg L^{-1}, respectively. The epilimnion was also more depleted in DIP relative to DIN, compared to the hypolimnion (Table S3). The yearly average DIN:DIP and DIN:TP mass ratios in the epilimnion were 32.7 and 5.3, respectively, and 9.3 and 6.8 for the corresponding values in the hypolimnion (Fig. S2). (Note: to convert to molar N:P ratios, the mass N:P ratios are multiplied by 2.21.)

3.1.6. Chlorophyll-a

The *chl-a* concentration is commonly used as a measure of the abundance of algal biomass in lakes (Jones and Bachmann, 1976; Quinlan et al., 2021; Reynolds and Descy, 1996). The long-term average value of *chl-a* for LW was 9.8 μg L^{-1} indicating eutrophic conditions. The concentrations, however, varied widely between years, in the range 0.2–41 μg L^{-1}. The average *chl-a* concentration in fall (11.4 μg L^{-1}) was higher than that observed in summer (9.3 μg L^{-1}). The *chl-a* time series data did not exhibit a statistically significant temporal trend ($p > 0.1$).

3.2. Principal components analysis

The percentages of the variations across the entire 13-dimensional water chemistry dataset that are explained by the PCs are presented in Fig. S3. The results indicate that 76 % of the variation in the data was explained by the first four PCs that, in descending order, contributed 33.3 %, 20.2 %, 11.7 % and 10.8 %. In the following section, we focus on these four PCs. Given their relative contributions, we refer to PC1 and PC2 as the major PCs and PC3 and PC4 as the minor PCs. The PC1 versus PC2 biplot is shown in Fig. 7.

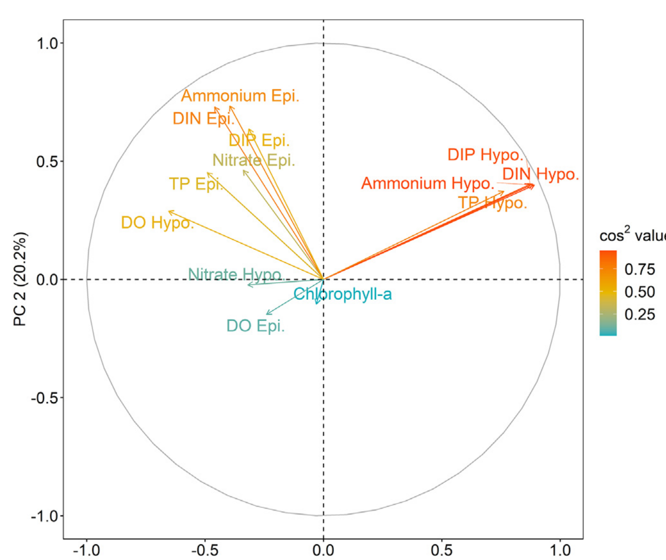


Fig. 7. PC1 versus PC2 biplot of variables included in PCA. The color of the arrows indicates how well the variable is represented by PC1 and PC2. Arrow length and direction show the extent and direction (positive or negative) of the variable's correlation with PC1 and PC2. Variables with arrows closest to the outer grey circle are best represented by PC1 and PC2.

The three variables that most strongly correlated with PC1 were the concentrations of the reactive forms of N and P in the hypolimnion: DIN, DIP, and NH_4^+ (Fig. 7, see also Fig. S4). Additionally, the correlations were all positive (Fig. 7, see also Fig. 8). For PC2 the three strongest, also positive, correlations were with the concentrations of DIN, DIP, and NH_4^+ in the epilimnion (Figs. 7 and 8, see also Fig. S4). The main three variables correlating with PC3 and PC4 were the concentrations of NO_3^-, DIP and *chl-a* in the epilimnion (for PC3), and *chl-a* and DO in the epilimnion plus NO_3^- in the hypolimnion (for PC4) (Fig. S4). These variables only weakly contributed to PC1 and PC2 (Fig. S4) and therefore plotted relatively close to the center of the PC1-PC2 biplot (Fig. 7).

The major cluster of variables that correlated positively with PC1 (hypolimnion DIN, DIP, NH_4^+, and TP) also correlated positively with PC2 (Figs. 7 and 8). In contrast, the major cluster that correlated positively with PC2 (epilimnion DIN, DIP, NH_4^+, and TP) correlated negatively with PC1. Moreover, the DO concentrations in epilimnion and hypolimnion were negatively correlated with PC1 but showed little connection to PC2.

3.3. Multiple linear regression (MLR)

We used MLR to delineate which of the 9 selected predictor variables (see Section 2.5.3) most impacted the temporal variations of the PCs. During the stepwise development and assessment of the MLR models, variables that had no impact on the PCs were discarded while the others remained in the pool of significant predictors (using a *p*-value of 0.1 as the cut-off). The remaining predictor variables are those in the grey shaded cells in Table 1. The table further identifies in bold text the predictor variable(s) with the largest regression coefficient(s) for a given PC. The value of a regression coefficient is a measure of the sensitivity of the PCs to the predictor variable. Note that because the predictor variables were normalized to their mean values, the relative magnitudes of the coefficients also provide a first-order indication of the relative importance of the predictor variables in explaining the variability of the PCs.

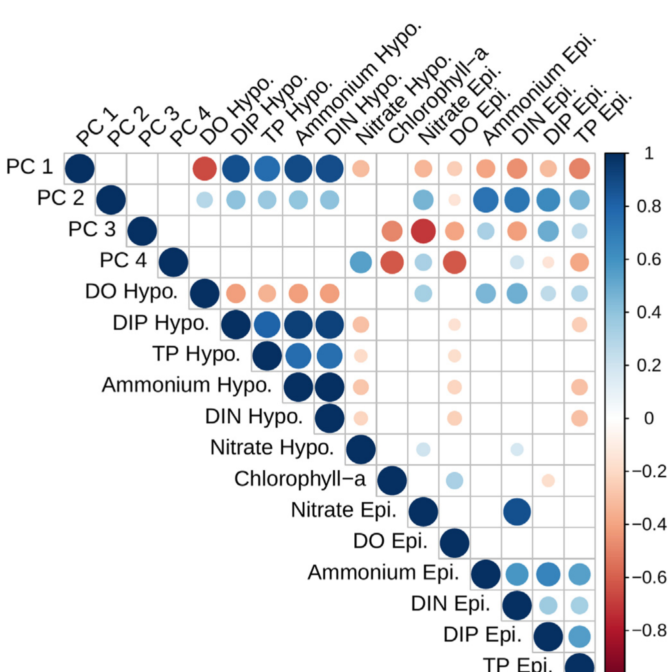


Fig. 8. Pearson's correlation between the PCs and variables included in the analysis ($p < 0.1$). Size and color of a circle indicate the strength and direction (blue is positive, red is negative) of the correlation, respectively.

4. Discussion

4.1. Water chemistry trends

The 22 years of water chemistry parameters were analyzed to help identify the drivers of symptoms of lake eutrophication. An abundant literature shows that excessive nutrient inputs, primarily phosphorus (P), are the most important driver of eutrophication of freshwater lakes (Gurkan et al., 2006; Smith, 1998; Vollenweider, 1968). Previous studies have also shown that eutrophication can be managed by reducing nutrient P inputs to lakes (Bhagowati and Ahamad, 2019; Carpenter et al., 1998; Smith et al., 1999). Nonetheless, although P inputs to LW have been reduced, the eutrophic symptoms have persisted. We therefore explored the chloride concentrations (and, hence, salinity) as potential explanatory variable because of the high increase in the lake water Cl^- concentrations (Fig. 2b) during the period that hypoxia in the hypolimnion intensified (Fig. 3). From the variables for which data series could be obtained, the two major macronutrients (P and nitrogen, N), dissolved oxygen (DO) and chlorophyll-a have all been linked to lake eutrophication and were therefore considered in the statistical analyses (see Section 4.2).

During the period 1996–2018, the water chemistry of LW underwent significant changes, especially in terms of the concentrations of Cl^-, DO, TP, DIP, plus the DIP:TP and DIN:DIP ratios (Fig. 4). The chloride concentration increased by a factor of 4, likely due to the increasing application of deicing agents as the watershed became increasingly urbanized (Fig. 2a). At the same time, however, the % imperviousness grew by only a factor of 2.3. Thus, the relative increase in LW's Cl^- concentration outpaced that of urbanization, as also noted by Corsi et al. (2010) for rivers in the northern United States. In addition, the changes in slope on Fig. 2b indicate that the expansion of urban cover alone does not explain the pace at which the Cl^- concentration has been building up in the lake. Additional factors include the type of deicing agent used (e.g., road salt versus brine) and the development of, and connectivity to, the stormwater management (SWM) infrastructure (Sorichetti et al., 2022).

In particular, the rapid increase in Cl^- between 1996 and 2010 possibly reflects the expansion of the total imperviousness in LW's catchment, while the development of directly connected imperviousness may explain the switch to a faster rise of the Cl^- concentration after 2010 (see Section 2.1). Directly connected imperviousness tends to accelerate the transfer of contaminants, including Cl^-, to streams and lakes (Brabec et al., 2002; Duan and Kaushal, 2015; Walsh et al., 2005). The spatial distribution of impervious surfaces in the watershed is an important factor controlling contaminant loads (Brabec et al., 2002; McGrane, 2016), including that of chloride (Richardson and Tripp, 2006), to the lake. More research will be needed to reconstruct the temporal trajectories of the spatial distribution of impervious land cover (Luo et al., 2018) and the connectivity to the stormwater network, as well as the types and application rates of de-icing agents, in order to fully assess their relative roles in the salinization of LW (Ramakrishna and Viraraghavan, 2005).

Rising Cl^- concentrations in lakes negatively impact water quality and drinking water supply (Kaushal et al., 2005, 2018), as well as aquatic ecosystem health (MacLeod et al., 2011; Tiquia et al., 2007). They can further cause changes in a lake's mixing regime (Bubeck et al., 1971; Novotny et al., 2008). With increasing Cl^- concentrations and, hence, increasing density of the water, more energy is required to overcome the thermal stratification (Mortimer, 1974b). In turn, reduced vertical mixing causes the expansion of DO depleted conditions in the hypolimnion (Fig. 3) and is consistent with the broadening of the depth interval of positive BVF values (Fig. 5) plus the increasing trend of the maximum BVF value over the time period considered (Fig. 4).

The external TP loads to LW, as well as the TP concentrations of both epilimnion and hypolimnion, have been generally declining (Figs. 4 and 6). A possible reason is increasing TP retention by SWM infrastructure, especially stormwater ponds, in the urbanizing watershed of LW (Goh et al., 2019; Kratky et al., 2017, 2021). In contrast to TP, however, DIP

Table 1
Results of the MLR analysis. Significant variables ($p \leq 0.1$) are identified by the shaded boxes with **bold** marking the most important explanatory variables.

	Urbanization		Climate					Loadings	Stratification
	Urbanization	Chloride	Mean temperature 14 days prior sampling	Average yearly temperature	Total rainfall 14 days prior sampling	Total yearly rainfall	>95% of the events	P loadings	BVF
PC1 (mixing/stratification)	0.87	3.05	3.30	0.99	-1.36	0.81	-0.66	0.56	**4.08**
PC2 (nutrient state and trophic conditions)	0.44	0.08	**2.97**	1.36	0.68	-1.21	-0.43	0.42	-0.78
PC3 (SWM infrastructure)	**2.52**	-1.53	-0.62	0.61	0.38	**-3.37**	1.22	0.75	-1.57
PC4 (extreme events)	1.18	-0.60	-0.68	0.63	-0.37	0.84	**1.70**	0.89	-1.38

concentrations exhibit negligible, or only very slightly decreasing, trends during the 1996–2018 period (Fig. 4). As a result, the DIP:TP ratios in the lake have been on the rise (Figs. 4 and S1). Also worth noting are the minimum DIP concentrations measured in 1998 and 1999 in the hypolimnion, which occurred during the short-lived operation of the aerator in the lake (see Section 2.1). Oxygenation of the bottom waters by the aerator probably inhibited the internal DIP loading from sediments to the water column.

Although the ultimate retention of P in bottom sediments is dependent on a variety of factors, including the flux of organic matter to the sediment and the relative concentrations of iron and sulfur (Orihel et al., 2017), the influence of dissolved oxygen at the sediment-water interface on internal DIP loading is well established (Markelov et al., 2019; O'Connell et al., 2020; Orihel et al., 2017; Parsons et al., 2017). When the hypolimnion remains fully oxygenated, P tends to be retained in the sediments, while the sediments tend to release DIP when the bottom waters become anoxic. Thus, the general trend toward longer annual periods of DO depletion of the hypolimnion (Fig. 3) and the corresponding increase of the AF has likely promoted internal P loading in LW. This is consistent with generally increasing trends of the DIP:TP ratios (Fig. S1). In the epilimnion, the most pronounced increase in the DIP:TP ratio occurred after 2012, that is, when LW's Cl^- concentration rose above 100 mg L^{-1}.

The DIP:TP ratio provides a rough indicator of the relative bioavailability of P, given that DIP represents the fraction of TP that is most easily assimilated by biota (Kao et al., 2021). Thus, the decreasing trends of the average TP concentrations in both the epilimnion and hypolimnion are in part being offset by the increase of the DIP:TP ratios. In addition to in-lake internal loading, SWM infrastructure in the watershed may also cause changes in the lake's DIP:TP ratio. Stormwater ponds, which are the dominant SWM structures in LW's watershed, generally reduce downstream TP loadings (Goh et al., 2019; Jefferson et al., 2017; Kratky et al., 2017). However, they also modify P speciation, often preferentially accumulating particulate P or even becoming net sources of DIP (Duan et al., 2012; Frost et al., 2019; Marvin et al., 2020).

The relative roles of internal and external P loading in modulating the trends in LW's DIP:TP ratios will need to be further investigated. The available data show that despite the decreasing average TP concentrations in LW, there has been no detectable improvement in the DO concentrations in the hypolimnion. We hypothesize that the rising salinity of LW is responsible for strengthening water column stratification, hence reducing the DO supply to the hypolimnion while enhancing internal P loading.

4.2. Statistical analyses

The first principal component (PC1), which explains around 33 % of the water chemistry trends, is positively correlated with the hypolimnion DIP, DIN and NH_4^+ concentrations and inversely with the hypolimnion DO (Figs. 7, 8 and S4). Thus, we propose that PC1 encapsulates the water column stratification, the redox state in the hypolimnion and the mixing regime of LW. As PC1 increases, stratification strengthens, DO in the hypolimnion decreases and the concentrations of DIP and NH_4^+ increase. Conversely, as PC1 decreases mixing increases resulting in higher hypolimnetic DO, lower DIP and decreased NH_4^+. This is supported by the observed seasonality of PC1 (Fig. S5). In southern Ontario lakes, thermal stratification typically starts in May and then increases until late summer (Kao et al., 2021). As expected, PC1 shows an increasing trend from May to September–October. The drop in PC1 between October and November can then be attributed to the breakdown of the summer stratification during fall turnover. Moreover, the trend analysis of PC1 shows a positive, admittedly weak ($p = 0.38$), temporal trend during the period of observation potentially suggesting a strengthening of the summer thermal stratification over time (Fig. S6).

The results of the MLR analysis indicate that BVF is the top predictor of PC1 (Table 1), hence confirming that the variability in the data explained by PC1 is closely related to the lake's stratification and vertical mixing regime. PC1 also correlates positively with the Cl^- concentration (Table 1), which is attributed to the stabilization of the water column stratification by the increasing water density. The other significant predictor for PC1 is the average air temperature (positive correlation) during the two weeks preceding a given sampling date. The latter likely reflects the steeper thermocline during warmer summer months.

The depth distributions of DO indicate a deepening of the redoxcline by approximately 2 m between 1996 and 2018 (Fig. 3). A comparable observation has been made by Kraemer et al. (2015) based on data from 26 lakes around the world. These authors found that, from 1970 to 2010, stratification in most of the lakes became more stable while also experiencing a deepening of the thermocline. Jane et al. (2021) report widespread declines of DO concentrations in both the surface and deep waters of temperate lakes. These authors invoke warmer air temperatures for lowering the O_2 solubility in surface waters and a strengthening of the thermal stratification for causing DO loss in the deeper waters. We propose that, in regions where the use of deicing agents is extensive, salinization is an additional driver of the intensification of lake stratification and, hence, the lowering of hypolimnetic DO concentrations.

While PC1 correlates negatively with the DIP, DIN and NH_4^+ concentrations in the epilimnion, PC2 is positively correlated with the DIP, DIN and NH_4^+ concentrations in both the epilimnion and hypolimnion (Figs. 7 and 8). The decreasing trend of PC2 (Figs. 4, S6) over the period 1996–2018, further parallels that of the TP concentration in the epilimnion (Fig. 6). Also, in contrast to PC1, the monthly average PC2 values do not experience a distinct drop in November (Fig. S5). This points to a link between the long-term changes of PC2 and the overall trophic state and nutrient enrichment of the lake, rather than to internal loading processes. The dependence of PC2 on the air temperature, both during the two-week period preceding sampling, as well as averaged over the year (Table 1), could reflect the enhanced transfer of biomass-associated N and P produced in the watershed during warmer years (Butturini and Sabater, 1998; McDowell et al., 2017; Wood et al., 2017). We therefore tentatively relate PC2 to the overall trophic and nutrient conditions of LW.

The interpretations of PC3 and PC4 are less straightforward. The epilimnion *chl-a*, NO_3^-, and DO concentrations contribute most to both PC3 and PC4 (Fig. S4). The correlations with *chl-a* and DO are negative, however (Fig. 8). Also, in contrast to PC1, PC3 and PC4 are inversely related to BVF (Table 1). For PC3, the strongest predictor variables are the extent of urbanization (positive) and the yearly precipitation (negative). Hence, PC3 may in part reflect nutrient processing and movement of nutrients within the SWM infrastructure in the lake's watershed. We speculate that, during drier years, longer water residence times and the accompanying more stagnant conditions in SWM ponds promote the mobilization of DIP and the elimination of NO_3^- by denitrification as seen in dammed reservoirs and during anoxic conditions in wetlands (Maavara et al., 2020; Parsons et al., 2017). The proposed impact of variable water residence time in the SWM ponds is consistent with the negative and positive correlations of PC3 with the epilimnion NO_3^- and DIP concentrations, respectively (Fig. 8). Drier years likely also result in less flushing out of deicing salt applied during preceding winters, hence, providing a possible explanation for the inverse relationship between PC3 and the Cl^- concentration in the lake (Table 1).

The rather similar correlation and regression coefficients for PC3 and PC4 seen in Fig. 8 and Table 1 imply a closeness of the two principal components. The most notable differences include the statistically significant increasing trend of PC4 with time ($\tau = 0.18, p = 1.6e-05$), compared to little change in PC3 ($\tau = -0.0063, p = 0.88$) (Figs. 4, S6), and the importance of the yearly 95 % percentile of rainfall as a predictor of PC4 rather than the yearly rainfall for PC3 (Table 1). Therefore, PC4 could record the impact of more extreme precipitation events on the transfer of N and P to the lake. Intense rainfall tends to cause more direct runoff from surrounding soils. Thus, the inverse relationships of PC4 with *chl-a* and DO could be attributed to increased turbidity that negatively impacts photosynthetic activity while enhancing heterotrophic activity in the lake's surface waters. Nutrient loads exported from soils also tend to be characterized by high N:P ratios (Downing and McCauley, 1992), consistent with the positive correlation of PC4 with the epilimnion nitrate concentration but a negative one with the epilimnion TP concentration (Fig. 8).

4.3. Implications

Seasonal stratification plays a key role in the biogeochemical functioning of lakes. With the establishment of a stable thermocline, the hypolimnion becomes increasingly isolated from the atmosphere thereby causing deoxygenation and the build-up of reduced chemical species, such as ammonium. It also activates the recycling of DIP from the sediments. That is, internal P loading gains in importance relative to the external P loading of the lake (Markelov et al., 2019).

According to our results, a key impact of urbanization on the biogeochemistry of LW is related to the growing impervious land cover and the accompanying increase in road salt application, which leads to a strengthening of the lake's water column stratification. Lake eutrophication is usually attributed to excessive P enrichment fueling enhanced algal growth (Schindler, 1971). Since the year 2000, however, the concentrations of TP and DIP in LW exhibit downward trajectories, while the *chl-a* data series imply little change in primary productivity of the lake. Thus, the symptoms of eutrophication, in particular the lengthening of the annual period of low hypolimnion DO concentrations, appear to be mostly caused by the progressive salinization of the lake.

Many temperate lakes experience declining DO concentrations and more intense stratification, changes that tend to be ascribed to climate warming (Jane et al., 2021). Our results suggest that, in cold and cold-temperate regions, salinization may be an additional driver of these changes. As shown by Dugan et al. (2017a, 2017b), in the past decade the Cl^- concentrations of lakes in urban watersheds in North America and Europe have been exhibiting generally increasing trends (Sorichetti et al., 2022), like the one observed in LW. Thus, the proposed salinization-driven water column stratification may be widespread. Efforts to control the external nutrient loading to such lakes may therefore be offset by the enhanced internal P loading caused by growing DO depletion of bottom waters.

Historical use of road salt in urban watersheds has also likely created chloride legacies in groundwater and soils (Mazumder et al., 2021). These legacies could continue to supply excess Cl^- to the lakes in these watersheds, even if winter salt applications were reduced or altogether halted. Protecting the health of urban lakes therefore calls for integrated management strategies that account for the impacts of past and ongoing road deicing practices on a lake's mixing regime. This, however, will require further work to better understand the relative contributions of climate change, urbanization, and salinization to changes in lake water quality.

5. Conclusions

The impervious urban land cover increased significantly in LW's watershed during the period for which water quality data are analyzed in this study (1996–2018). The growing impervious and directly connected impervious land covers have been accompanied by an increase in the annual duration of anoxic conditions in the hypolimnion, as well as an increase in the DIP:TP ratio. The increasing anoxia and intensification of the water column stratification have been occurring despite stable, even slightly decreasing, external TP loads to the lake, likely because of the expansion of stormwater management infrastructure in the watershed since the early 2000s. At the same time, in-lake TP concentrations have declined, while *chl-a* concentrations show no evidence of increasing algal productivity.

Statistical analyses (PCA and MLR) of the water chemistry time series data shed light on the mechanism(s) responsible for the intensification of eutrophication symptoms in LW. The progressive salinization of the lake emerges as a major driver of the strengthening of water column stratification and the accompanying changes in water quality, in particular the expansion of hypolimnion anoxia and the enhancement of internal P loading. The latter contributes to maintaining LW in its eutrophic state. To our knowledge, this is the first study that links salinization with lake eutrophication symptoms. Lake management in regions where the use of de-icing agents is widespread therefore needs to consider the multi-faceted role of salinization in changes lake water quality, including those that traditionally are associated to increasing external nutrient loading. Further research on the water quality impacts of de-icing agents will require reliable data on their types and application rates.

Data availability statement

The data that support the findings of this study are openly available from the Federated Research Data Repository (FRDR) website at DOI: 10.20383/103.0577. Additionally, the raw data provided by the City of Richmond Hill and used in this study can be obtained at DOI: 10.20383/102.0540 on the FRDR website.

CRediT authorship contribution statement

Jovana Radosavljevic: Conceptualization, Methodology, Validation, Visualisation, Formal analysis, Interpretation, Data curation, Writing - original draft.

Stephanie Slowinski: Methodology, Validation, Formal Analysis, Interpretation, Writing – Review and Editing, Visualization.

Mahyar Shafii: Methodology, Writing – Review and Editing.

Zahra Akbarzadeh: Visualization.

Fereidoun Rezanezhad: Writing - Review & Editing.

Chris T. Parsons: Interpretation, Methodology, Writing - Review & Editing.

William Withers: Resources, Data interpretation.
Philippe Van Cappellen: Interpretation, Writing - Review & Editing, Supervision, Funding acquisition.

Declaration of competing interest

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

Acknowledgements

Financial support was provided by a Natural Science and Engineering Research Council of Canada (NSERC) Strategic Partnerships Grant (STPGP 521515-18), and the Lake Futures project and Managing Urban Eutrophication Risks under Climate Change project within the Global Water Futures (GWF) program funded by the Canada First Research Excellence Fund (CFREF). We thank the City of Richmond Hill for providing the data. Nina Sattolo helped with the PCA analysis; other undergraduate student assistants in the Ecohydrology Research Group helped with assembling the data set. We thank Bhaleka Persaud for help with publishing and managing data. The constructive comments of two journal reviewers improved the clarity of our manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.157336.

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