



Great Lakes chloride trends: Long-term mass balance and loading analysis

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ABSTRACT

Surveillance data collected over the past 150 years are compiled and analyzed to identify chloride trends in the Laurentian Great Lakes. These data indicate that chloride levels started rising in the mid-19th century and began accelerating in the early twentieth century. Lake Superior's and Lake Michigan's concentrations have continued to increase steadily and currently stand at their maximum recorded levels. In contrast, lakes Huron, Erie and Ontario reached peak levels between 1965 and 1975, but then began to decline. However, recent data indicate that the chloride concentrations in these lakes are now increasing again. Because loading data are not readily available, a mass-balance model is employed to estimate the chloride inputs required to account for the concentration trends. This inverse analysis yields computed load reductions that are consistent with reported industrial load reductions during the last three decades of the 20th century. Hence, it appears that the improvements were for the most part attributable to industrial controls. The model is also used to predict that if loads are held fixed at 2006 levels, concentrations in all lakes will continue to increase with the most dramatic rise occurring in Lake Michigan which will ultimately approach the level of Lake Erie.

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Introduction

Because it is conservative, has low natural background levels, and is readily measured with adequate precision, chloride has long served as a general indicator of anthropogenic impacts on Great Lakes water quality. Beyond its utility as an indicator, chloride can also have direct adverse impacts on lakes and other freshwater ecosystems. The Canadian federal government has recommended that road salt be added to the List of Toxic Substances under the Canadian Environmental Protection Act because of tangible threats of serious or irreversible environmental damage (Environment Canada and Health Canada, 2001). It has also been suggested that increases of chloride and other ions could be directly harmful to the Great Lakes ecosystem by fostering the spread of less-desirable halophilic organisms (Stoermer, 1978). More generally, trends of all major anions and cations are relevant as they contribute to aspects of lake chemistry such as ionic strength, pH and acid neutralizing capacity. Finally, at sufficiently high concentrations, chloride can influence density stratification as well as degrade the taste of drinking water.

Two classic papers first established that Great Lakes chloride levels had increased significantly over the past century. In a

perceptive and timely data analysis, Beeton (1965) documented substantial increases, starting as early as the late 19th century, in all the lakes with the exception of Lake Superior. In another seminal contribution, O'Connor and Mueller (1970) developed a mathematical model that successfully simulated Beeton's observations and provided insight into their underlying causes. In particular, they suggested that the increases were primarily induced by industrial discharges and runoff of road salt.

Subsequent analyses followed the trends through the 1970s and 1980s. For example, Sonzogni et al. (1983) concluded that as of the early 1980s, chloride loadings had stabilized but, if the loadings were not reduced, chloride concentrations would continue to increase significantly into the future. In particular, because of their long hydraulic residence times, they used a mass-balance model to predict that lakes Superior, Michigan and Huron would continue increasing for several centuries to levels well beyond their mid-1970 concentrations.

In fact, rather than stabilizing, chloride loadings had actually been significantly reduced beginning in the late 1960s due primarily to reductions of industrial discharges (Fig. 1). For example, the USEPA (1972) documented major reductions in chloride inputs to Lake Michigan in the early 1970s primarily from salt and chemical industries in the Manistee–Ludington area of Michigan. Dolan and Bierman (1978) indicated that the substantial chloride load to Lake Huron from Saginaw River industrial sources decreased about 600 kilotonnes per annum (kta) between 1965 and 1976. Crucil et al. (1991) reported that Lake Erie's chloride loading dropped

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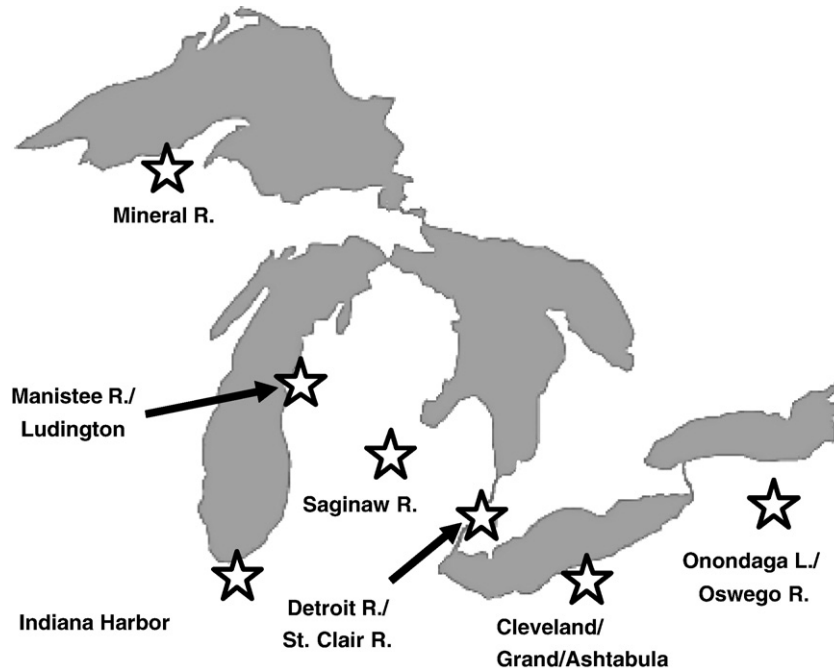


Fig. 1. Areas with major industrial chloride discharges circa 1960. Significant reductions of these sources have occurred over the ensuing 35 years.

approximately 1700 kta between 1965 and 1986 due to the closing of several industrial facilities on the Detroit River. Finally, Effler and colleagues have rigorously documented that Lake Ontario's chloride load decreased approximately 700 kta following the closure of a major chlor-alkali plant in 1986 that had discharged into Onondaga Lake (Doerr et al., 1994; Effler and Driscoll, 1986; Effler et al., 1985, 1990, and Matthews and Effler, 2003).

Because of its short residence time and high industrial salt discharges, Lake Erie was the first part of the system to respond to these load reductions. Rathke and McCrae (1989) reported a downward trend in the central basin of 0.55 mg/L per year between 1966 and 1986. Whyte et al. (1990) showed similar downward trends in data collected at municipal water intakes in all three basins of Lake Erie for the same period.

The current study employs more recent observations to extend the trend analysis to the present. In addition, an inverse analysis is used to estimate time series of chloride loadings from pre-settlement (early 1800s) through 2006. An inverse analysis employs a mass-balance model to back-calculate the loadings necessary to generate the observed chloride concentration trends. Revised predictions are also made to assess the ultimate steady-state concentrations that will be approached if present loading levels are maintained.

Methods

Mass-balance framework

The Great Lakes are treated as a system of interconnected, completely-mixed systems (Fig. 2). This spatial characterization is consistent with a long-term time scale that is designed to resolve interannual variability by employing annual averages for all model variables and parameters. Hence, higher-frequency intraannual variations, such as seasonal and diel cycles, are ignored. This basic approach has been successfully applied to the Great Lakes in previous studies to simulate long-term trends of chloride (O'Connor and Mueller, 1970; Sonzogni et al., 1983), total phosphorus (Chapra, 1977; Chapra and Robertson, 1977; Lesht et al., 1991), and persistent toxicants (Thomann and Di Toro, 1983).

A direct load is delivered to each lake consisting of the total input from tributary runoff, direct point loadings, atmospheric sources and diversion inflows. Each lake has a net advective outflow that transports chloride to a downstream lake. Lake Michigan also has a diversion outflow to the Chicago River that transfers chloride directly out of the basin. In addition, diffusive mixing occurs between lakes Michigan and Huron due to two-way flows across the open boundary of the Straits of Mackinac.

A mass-balance model that accounts for these mass transfers can be written generally as

$$V \frac{dc}{dt} = W(t) + Q_{ui}(t)c_{ui} - Q_o(t)c - Q_d(t)c + E'(c_b - c) \quad (1)$$

where V = lake volume (km^3), c = chloride concentration (mg/L), W = direct loading rate (kta), Q_{ui} = inflow rate from an upstream lake (km^3/year), c_{ui} = chloride concentration of upstream lake (mg/L), Q_o = outflow rate that carries chloride to a downstream lake (km^3/year), Q_d = diversion outflow (km^3/year), E' = bulk eddy diffusion

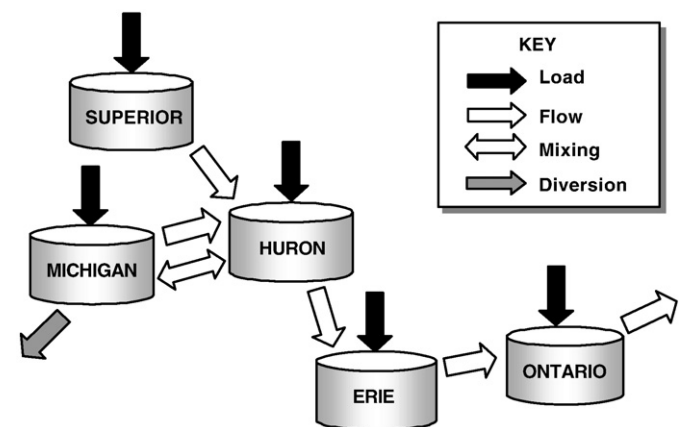


Fig. 2. Segmentation scheme for the Great Lakes showing the sources and transport of chloride.

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