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# SEASONAL AND TEMPORAL VARIATIONS OF CHLORIDE LEVEL IN RIVERINE ENVIRONMENT

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**Abstract:** Chloride is essential but it can also have detrimental impacts on ecosystem at elevated level. Chloride in natural water bodies often presents both intra- and inter-annual variations which could result from both natural and anthropogenic factors. Among many anthropogenic activities, the use of road salts for de-icing in winters is a common practice across Canada and has been demonstrated to elevate chloride level of rivers/streams. Besides, flow, a hydrologic factor, appears to be closely associated with chloride level regardless of the impact of anthropogenic activities. This paper aimed to further understand the variations in chloride and the possible impact of the use of road salts on chloride in rivers through statistically analyzing data collected across Canada. The results showed that the seasonal (intra-annual) variation of chloride presented in general and chloride concentration was negatively correlated with flow at most stations. The seasonal variation pattern and the dependency of chloride on flow however appeared to be different between large and small rivers. At the inter-annual scale, the increase of chloride concentration was qualitatively linked to the increase of the use of road salts in general; however the Code of Practice for the Environmental Management of Road Salts issued in 2004 appeared to help in reducing the increase trend of chloride level. Furthermore, the results from the locally weighted scatterplot smoothing (LOWESS) analysis showed that the temporal trend observed in chloride at the inter-annual scale could be primarily ascribed to the effect of either anthropogenic activities, flow, or their combination.

## 1 INTRODUCTION

Chloride is essential to all organisms including humans. Although chloride exists ubiquitously in environment, it can pose detrimental impacts on environment and human health when its level is elevated. Chloride is also inert and thus not commonly involved in geochemical reactions in aquatic environment. Therefore chloride has been often used as one of indicators of pollution and monitored in surface water bodies. Although chloride is conservative in aquatic environment owing to its inert chemical property, there are diverse factors influencing its level or concentration in surface water bodies. Among the influencing factors on chloride level in surface water bodies, the anthropogenic interference has been ascribed as the most significant cause of increasing chloride level (Eneji et al., 2012). Anthropogenic activities interfere on chloride level in many different ways, for example through releasing effluents of wastewater treatment plants and treated wastewater from industries which use large quantities of sodium chloride in many processes (point source pollution), and snowmelt contaminated by road salts (non-point source pollution) in cold regions.

A large number of studies have identified the impact of the use of road salts on various surface water bodies including rivers/streams and lakes as well as groundwater. The use of road salts has been ascribed to cause the elevation of chloride concentration, for example in the Mohawk River in New York State (Godwin et al., 2003), streams in New Hampshire (Daley et al., 2009), urban lakes in the Twin Cities Metropolitan Area of Minnesota (Novotny et al., 2007), and streams in snow-affected urban watersheds (Corsi et al., 2015) in U.S. Besides, the use of road salts has been observed to affect chloride level of groundwater, which would also affect the chloride level of surface water but at a delayed time (Kelly et al., 2012; Meriano et al., 2009; Coris et al., 2015). Therefore, the impact of the use of road salts could occur on a short time scale (during snow melting season) and a long time scale (during a year or years) as road salts can be transported into water bodies through both surface and subsurface flows.

Besides the effect of anthropogenic activities, natural factors (especially hydro-meteorological variables) could influence chloride level of surface water bodies. In riverine environment, the temporal variations/changes in hydrogeological variables such as precipitation, surface runoff, interflow, groundwater flow, and pumped in and outflows in general influence the river flow and subsequently the pollutant levels in streams (Vega et al., 1998; Rani et al., 2011). Variations in water quality and quantity have been widely investigated across the world (e.g., Gupta and Chakrapani, 2005; Rani et al., 2011; Eneji et al., 2012). It is unexceptional that flow, as one of natural factors, can largely affect chloride concentrations in rivers by either diluting chloride or transporting chloride into water bodies from both point and non-point sources. A large number of previous studies have documented that chloride level is in general negatively dependent on flow (e.g., Meriano et al.; 2009, Kelly et al., 2010; Coris et al., 2015; Boyer et al., 1999), which suggests that chloride is diluted by high flows. In such a circumstance, chloride concentration is usually higher in low flow than in high flow.

Many hydro-metrological variables such as flow show prominent seasonal (intra-annual) variations. Some anthropogenic activities such as the use of road salts also have seasonal nature, namely their applications occur in a specific time period in a year; whereas other anthropogenic activities such as releasing effluent from wastewater treatment plants and industries are relatively constant over a year. Therefore, the change and/or variation of chloride level can be very complicated as natural factors and anthropogenic activities drive the change/or variation of chloride level in surface water bodies differently and at different time scales. As many water quality parameters including chloride demonstrate strong seasonal variation in surface water, the characterization of their seasonal variations is very important to evaluate temporal changes of river pollution from natural or anthropogenic point and non-point sources (Ouyang et al., 2006). In addition at the inter-annual time scale, the understanding of the temporal change or trend in water quality is also required to formulate effective management strategy. Although multiple influencing factors could potentially affect the temporal variation/change of chloride at the intra- and inter-annual time scales, some influencing variables might play more important roles than other variables. Thus the role of one variable could be masked by other variables. Therefore, it might be necessary to study the potential impact of one variable after removing the impact of other variables.

In Canada, on average five million tonnes of de-icing salts have been used annually on roadways (Environment Canada, 2012). The existing body of knowledge has provided substantial evidence that the increase of chloride level results in detrimental effects on aquatic environment, terrestrial vegetation, wildlife mortality and soil chemistry. Road salts thus have been put on the second Priority Substances List of the Canadian Environmental Protection Act. The increase of chloride level in both groundwater and surface water has been linked to the increase of the use of road salts. Especially, surface runoff quality affected by the deicing operation has been blamed to cause the degradation of the water quality of freshwater bodies including streams and lakes (Environment Canada, 2012). To effectively mitigate the negative impacts posed by the application of road salts, the Code of Practice for the Environmental Management of Road Salts (referred as Code throughout this paper) was published in 2004. According to the above review, the objectives of this paper were to: (1) investigate the seasonal variation of chloride level; (2) study the temporal trend of chloride level and the effect of the Code; and (3) identify the influencing factor(s) primarily driving the temporal trend of chloride in rivers.

## 2 METHODOLOGY

### 2.1 Study Area and Data Collection

Due to the temporal and spatial nature of the road salt application, it leads to the change/variation of chloride level in water bodies at both temporal and spatial scales (Ramakrishna and Viraraghavan, 2005). Thus this paper did not target specific locations and collected data from rivers/streams across Canada. A total of 4288 datasets of chloride were collected from six provinces including Alberta, Saskatchewan, Manitoba, Ontario, Prince Edward Island (PEI), and Nova Scotia. Due to requirement for statistical analysis described below, datasets collected from 103 water quality stations in four provinces including Alberta, Manitoba, Ontario, and PEI were used.

From statistical analysis perspective, longer datasets are always desirable; whereas data availability is always the primary limiting factor for conducting statistical analysis. Schertz et al. (1991) stated that the water quality data should be collected according to a regular and fixed agenda and that a minimum record length of 5-year monthly data or longer for less sampling frequency is needed for monotonic trend analysis. In addition, Vecchia (2000) also mentioned that the data duration should not be less than 5 years to identify a true trend (including simple monotonic trends, compound monotonic trends, and step trends) in the mean because of the effect of autocorrelation in trend analysis. In addition, data both before and after the implementation of the Code in 2004 is required to investigate the potential effect of the Code on the variation of chloride concentration. Therefore, the selection of datasets was primarily conducted based on the following criteria:

- Sampling frequency is approximately monthly or less than monthly. The datasets that cover a wide range of stream flow conditions (from base flow to high flow) are preferred. Note that the datasets, which provide chloride concentration in non-winter season (e.g., at many water quality stations in Ontario), are also selected considering that the use of road salts can have prolonged impact (not only in snow-melting season) on chloride concentration in surface water bodies.

- Datasets consist of at least 5-year data before and after 2004, respectively, as the Code has been implemented since 2004.

Flow data corresponding to water quality monitoring locations were collected from Water Survey of Canada. If the water quality monitoring locations are not coincident with flow gauge stations, flow gauge stations in the proximity of the water quality monitoring locations (either upstream or downstream) were used. Therefore, flow data are not available for all water quality stations. The use of road salts was collected from municipal, provincial, and federal governments across Canada, and a total of 304 datasets were collected. Note that the data on the use of road salts are often available after the implementation of the Code in 2004.

### 2.2 Statistical Analysis

To investigate the presence of seasonal variation in chloride concentration, the analysis of variance or its non-parametric alternative, the Kruskal-Wallis test, was adopted. The analysis was employed to investigate if the mean(s)/or median(s) of a group(s) data is statistically different from other group(s). In the analysis, the dataset was grouped by months, thus there were in general 12 groups each of which corresponds to a month. If the statistical difference in medians or means is detected in the analysis, it is concluded that seasonality presents in the dataset. To investigate the presence of temporal trend in chloride concentration and the annual use of road salts, the parametric approach (regression approach) and the non-parametric trend test, the Mann-Kendall (MK) test, were employed. As shown in the section of results, seasonality commonly show in chloride data, thus the seasonal MK was primarily used; while the MK test was applied in the absence of seasonality.

To investigate if the Code issued in 2004 helps in effectively reducing the use of road salts and consequently the negative effect of the use of road salts on chloride level. Each dataset of chloride concentration was split into two sub-datasets, one for the time period before 2004 and the other for time period after 2004. The effectiveness of the Code was studied by comparing the identified temporal trends in chloride concentration and their means/medians in these two time periods (before 2004 and after 2004) using the parametric test (t-test) or the nonparametric test (Wilcoxon rank sum test).

To distinguish the role of flow from the role of the combination of all other influencing factors (except flow) in the observed temporal trends in chloride concentration, the Locally Weighted Scatterplot Smoothing (LOWESS) method was applied to remove the confounding effect of changing streamflow on chloride concentration. In the LOWESS, the smoothing factor of 0.5 was used. The smoothing factor defines the fraction the data is used to fit each local polynomial. The trend test was then applied to detect the trends in the flow-adjusted chloride concentration, which is residual of chloride concentration after the effect of flow on the variation of chloride concentration is removed, and predicted chloride concentration, which is modeled using flow as the only independent variable in the LOWESS. In addition, Pearson or Spearman's correlation coefficient between chloride concentration and flow was calculated to demonstrate their dependency.

All statistical analyses except the seasonal MK and MK tests were conducted using MATLAB. The selection of parametric and non-parametric tests in the analyses was conducted according to the normality of a dataset, which was tested using the Kolmogorov–Smirnov (K-S) test. If a dataset was normally distributed, parametric tests were applied otherwise non-parametric testes were adopted. The seasonal MK and MK tests were conducted using the computer program of USGS for trend analysis (Helsel et al., 2005). All statistical tests were conducted at a significance level of 5%.

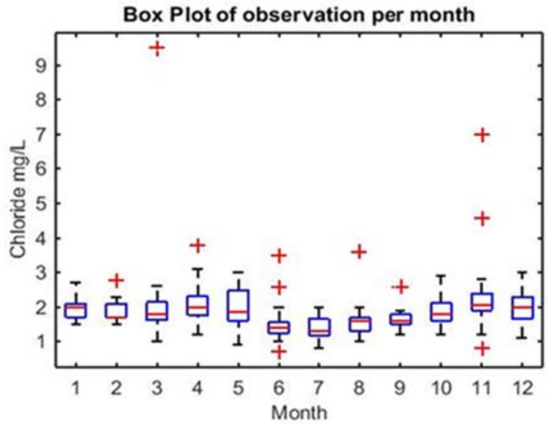
## 3 RESULTS AND DISCUSSION

### 3.1 Seasonal Variation of Chloride

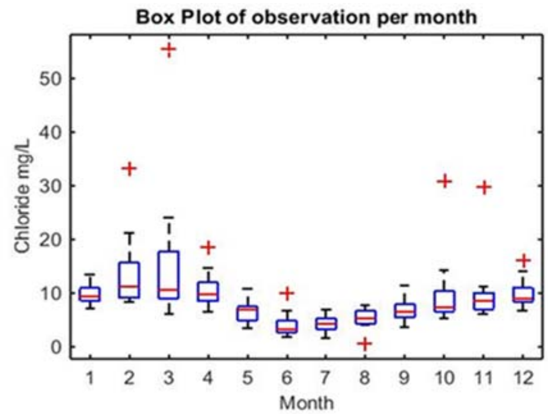
The seasonal variation of chloride concentration was statistically detected at most stations in the time periods of both before 2004 and after 2004. In the time period of before 2004, seasonal variation was detected at 82% stations (84 out of 103); while in the time period of after 2004, seasonal variation was observed at 87% stations (90 out of 103). Three different seasonal variation patterns were seen from the studied stations. Figure 1 displays the monthly boxplots of chloride concentration at three selected stations and illustrates the three different seasonal variation patterns. Chloride concentration is often lower in spring/summer months than that in other months (Figure 1(a) and (b)); while at some stations, relatively higher chloride concentration was measured in late summer and fall (Figure 1(c) and (d)). At several stations the seasonal variation was statistically absent (Figure 1(e) and (f)), although relatively high chloride concentrations were measured during summer/fall months. In general, chloride level tends to be low when flow is high. In a river, high flow is often observed in warm months while low flow is measured in winter months. The low level of chloride during high flow show that the high flow dilutes the concentration of chloride. However relatively higher chloride concentration in late summer and fall or no seasonality detected are more likely to be seen in small rivers. Small rivers are more prone to local hydrological events, such as snow melt and rain events and thus often behave differently from large rivers.

### 3.2 Temporal Trend of Chloride

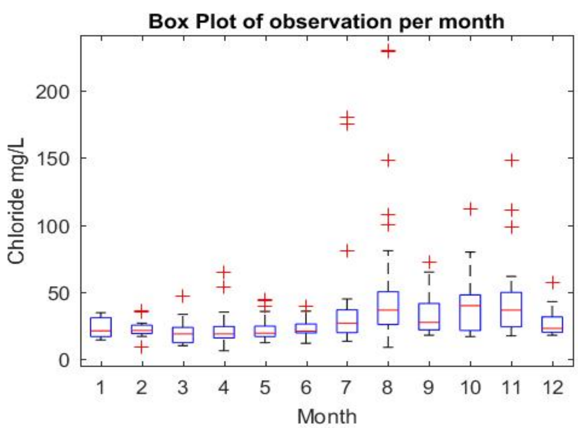
Different temporal behaviors of chloride at the inter-annual scale, namely significant and insignificant upward and downward trends, and no trend, were statistically detected in the both time periods of before 2004 and after 2004 from the studied stations. Among the 103 stations, upward trend (both significant and insignificant) of chloride level was identified at the majority of stations (about 85%) in the time period of before 2004. In the time period of after 2004, chloride level at about 43% of stations remained upward trend; whereas chloride level at 57% of stations showed downward trend (both significant and insignificant) or no trend. Figure 2 presents the trend analysis results at three sample stations. The temporal trend at a station can be resulted from various causes including anthropogenic activities, natural variations/changes (e.g., hydro-meteorological variations/changes), and the combination of the both. The anthropogenic activities such as urbanization, the use of road salts, anthropogenic point-sources (e.g., effluent from wastewater treatment plants), and anthropogenic non-point sources (e.g., snowmelt and stormwater runoff) can have immediate and/or prolonged impacts on the chloride in surface water bodies. Among the hydro-meteorological variables, river flow has often been the primary driver of seasonal variation of chloride as discussed previously. In addition, the hydrologic cycle and thus hydrologic variables are subjected to variations/changes due to the variations/changes in meteorological condition as well as anthropogenic activities. Therefore it is crucial to investigate that either anthropogenic or natural factor leads to the temporal trend of chloride in order to more effectively manage riverine water quality.



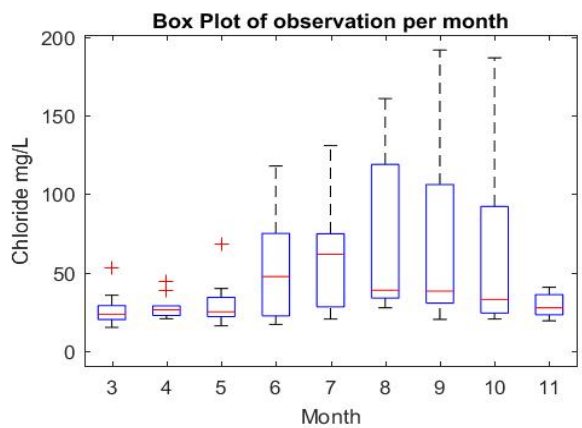

(a) Before 2004



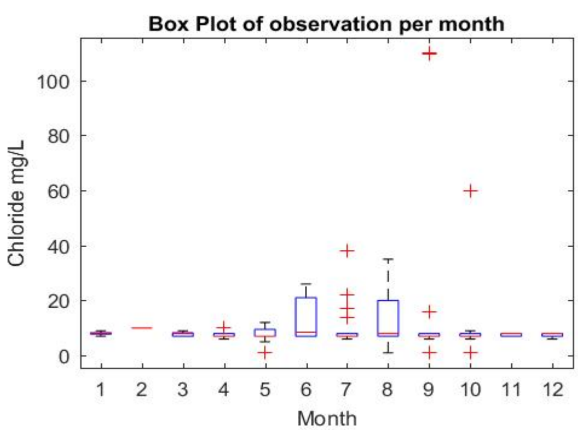

(b) After 2004



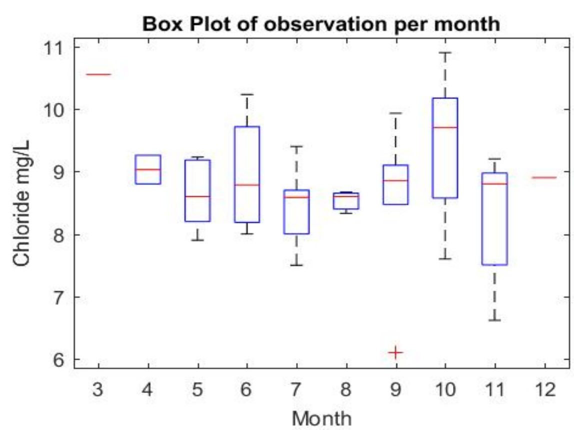

(c) Before 2004




(d) After 2004

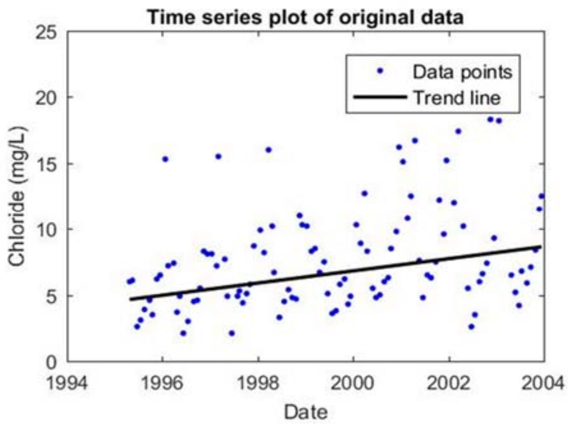



(e) Before 2004

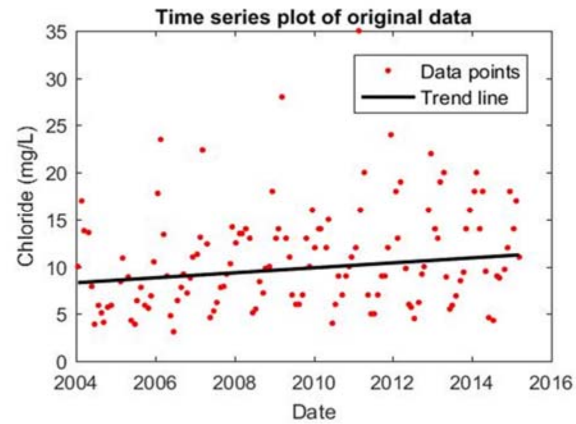



(f) After 2004

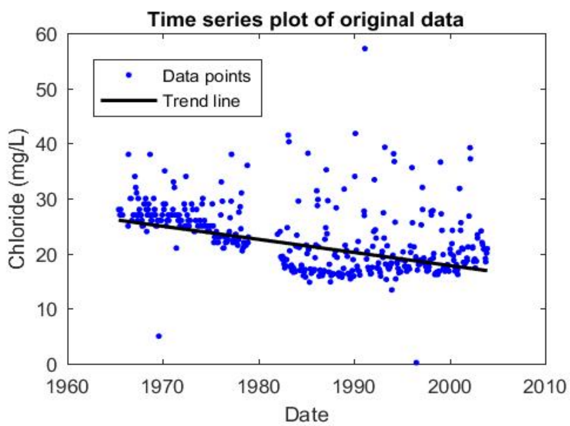
Figure 1: Monthly Boxplots of chloride concentration in the time periods of before 2004 and after 2004: (a) and (b) at the Bow River below Carseland Dam in Alberta; (c) and (d) at the Ausable River in Ontario; and (e) and (f) at the West River at Bolger Park Road in Prince Edward Island



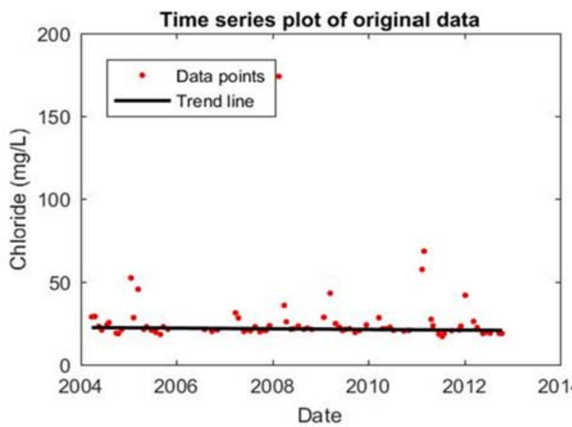

(a) Before 2004 (significant upward trend)



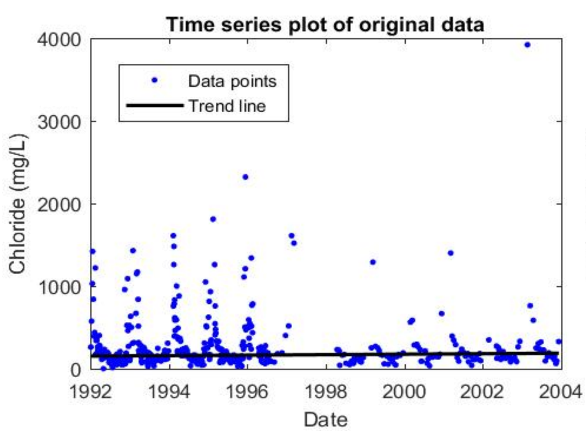

(b) After 2004 (significant upward trend)



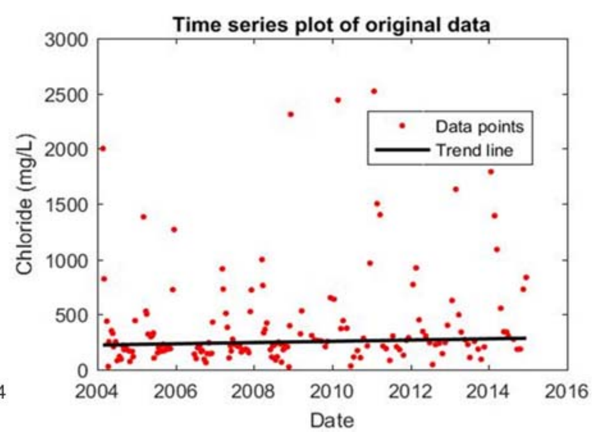

(c) Before 2004 (significant downward trend)




(d) After 2004 (significant downward trend)




(e) Before 2004 (insignificant upward trend)




(f) After 2004 (insignificant upward trend)

Figure 2: Time series plots and trend lines of chloride concentration in the time periods of before 2004 and after 2004: (a) and (b) at South Saskatchewan River above Medicine Hat in Saskatchewan; (c) and (d) at Welland Ship Canal in Ontario; and (e) and (f) at the Don River in Ontario

### 3.3 Primary Factor Driving the Temporal Trend of Chloride

Acknowledging that many potential factors including both anthropogenic and natural factors could lead to the temporal change at the inter-annual scale of chloride in surface water bodies, it is vital to differentiate their roles in order to formulate efficient and effective management strategy to reduce chloride level in surface water bodies. In the following analysis, it is assumed that flow is the primary natural factors driving the variation of chloride as the variation of other hydro-meteorological variables is either reflected into flow (such as precipitation) or negligible. The LOWESS was applied to identify the primary driving factor (either flow or combined anthropogenic factors) causing the temporal variation of chloride. In the LOWESS, flow was used as the only explanatory variable, thus the predicted chloride concentrations are those that are explained by flow. The residuals in the LOWESS represent the chloride concentrations that can be explained by all other factors (especially anthropogenic factors). The trend analysis was then applied to investigate the trends of predicted chloride concentrations and the residuals, which reflect the trend caused by flow and anthropogenic activities, respectively. The analysis was conducted for four selected stations, at which chloride concentration is associated with flow differently (significant negative correlation at two stations, no significant correlation at one station, and significant positive correlation at one station). Note that chloride concentration was found to be in general negatively correlated with flow at most stations.

The trend analysis results for original data, predicted data, residuals, and correlation coefficient (significant) are summarized into Table 2. Furthermore, the time series of predicted chloride concentration and residual from LOWESS and their trend lines are presented in Figure 3 at two stations as examples. As the results shown, the temporal trends detected in chloride concentration are primarily caused by either flow at the station on the Canagagigue Creek or by anthropogenic activities at the stations on the Elbow and Bighead Rivers. At the station on the Duffins Creek, both flow and anthropogenic activities significantly contribute to the increase of chloride concentration, whereas the role of anthropogenic activities is more prominent than that of flow. In addition, the anthropogenic activities appear to always result in the temporal upward trend in chloride; whereas flow can lead to significant increase (station on the Duffins Creek) or decrease (station on the Canagagigue Creek) of chloride concentration. Comparing the four streams/creeks, the magnitude of flow in Duffins Creek and Canadagigue Creek is much lower than that in Elbow and Bighead Rivers. Small rivers/creeks are likely more prone to local hydrological events, such as snow melt and rain events, which could largely change their flow and also transport pollutants into water bodies. This might explain the different hydrologic and water quality behavior of small and large rivers.

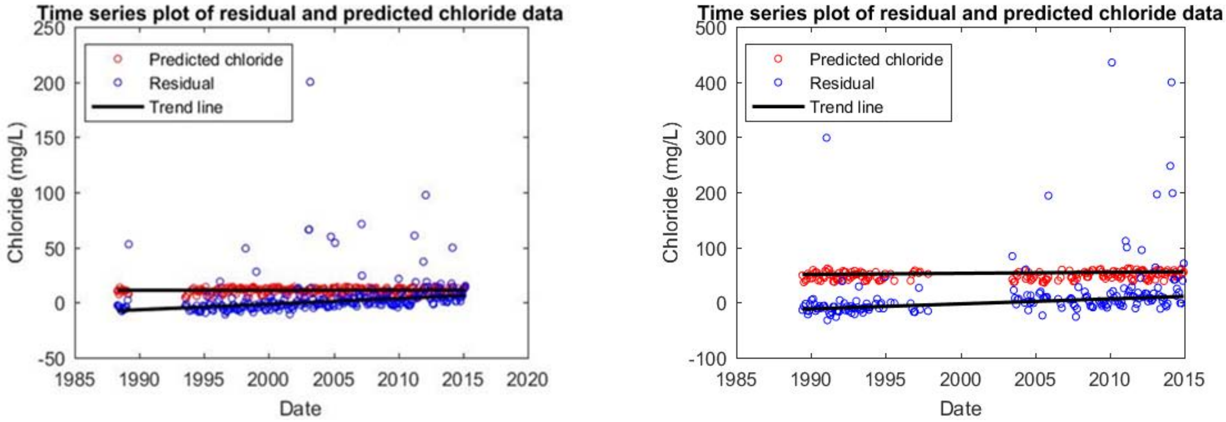
### 3.4 The Effect of the Code

From six datasets of the annual use of road salts for the time period of before 2004 collected across Canada, upward trend (both significant and insignificant) was detected from four out of six datasets; while insignificant downward trend was found from two datasets. After 2004, upward trend was found from 70% of datasets. Thus it can be concluded that the annual use of road salts increases significantly or insignificantly in general after the implementation of the Code in 2004.

To further illustrate the change in chloride level after the implementation of the Code, the medians of chloride concentration were compared between the two time periods of before 2004 and after 2004. The results of the comparison are shown for the southern part of Ontario where all water quality stations are located in in Figure 4 (as example). The median of chloride level was elevated at the majority of stations (about 83% of the studied stations) after 2004; while at the remaining stations, the medians of chloride concentration either decreased or were remained same. Although chloride level in terms of median did not decrease statistically in general, chloride concentrations were found to decrease after 2004 at some stations where the trend of chloride concentration changed from upward trend before 2004 to downward or no trend after 2004. The results suggest that the effect of the Code at a certain degree, however the implementation of the Code has not reversed the increasing trends in chloride in general. The previous results demonstrate that both natural and anthropogenic factors could result in the temporal variation in chloride. In addition, urbanization can lead to the increase of the use of road salts, which can possibly be masked the effectiveness of the Code in reducing the use of road salts. A more elaborate research, which takes all potential influential factors on chloride into account, is recommended to assess the effect of the Code.

Table 2: Trend analysis results of the original data, predicted chloride concentration, residual, and significant correlation coefficient between chloride concentration and flow at four selected stations.

| Station number (station name) | Time | Data set | Trend | | | Correlation |
|---|---|---|---|---|---|---|
| | | | Significant or Insignificant | Upward or Downward | Slope | |
| AB05BJ0450 (Elbow River at 9th Ave Bridge) | 1988-2015 | Original data | Significant | Upward | 0.48 | -0.37 |
| | | Predicted data | Insignificant | Upward | 0.01 | |
| | | Residual | Significant | Upward | 0.51 | |
| 03003000202 (Bighead River) | 1975-2014 | Original data | Significant | Upward | 0.09 | -- |
| | | Predicted data | Insignificant | Downward | -0.00 | |
| | | Residual | Significant | Upward | 0.09 | |
| 06010400102 (Duffins Creek) | 1989-2014 | Original data | Significant | Upward | 1.11 | 0.38 |
| | | Predicted data | Significant | Upward | 0.19 | |
| | | Residual | Significant | Upward | 0.91 | |
| 16018401602 (Canagagigue Creek) | 1966-2014 | Original data | Insignificant | Downward | -0.08 | -0.64 |
| | | Predicted data | Significant | Downward | -0.32 | |
| | | Residual | Insignificant | Upward | 0.09 | |




(a) (b)

Figure 3: Time series of predicted chloride concentration and residual with their trend lines at: (a) the station on the Elbow River in Alberta; and (b) the station on the Duffins Creek in Ontario

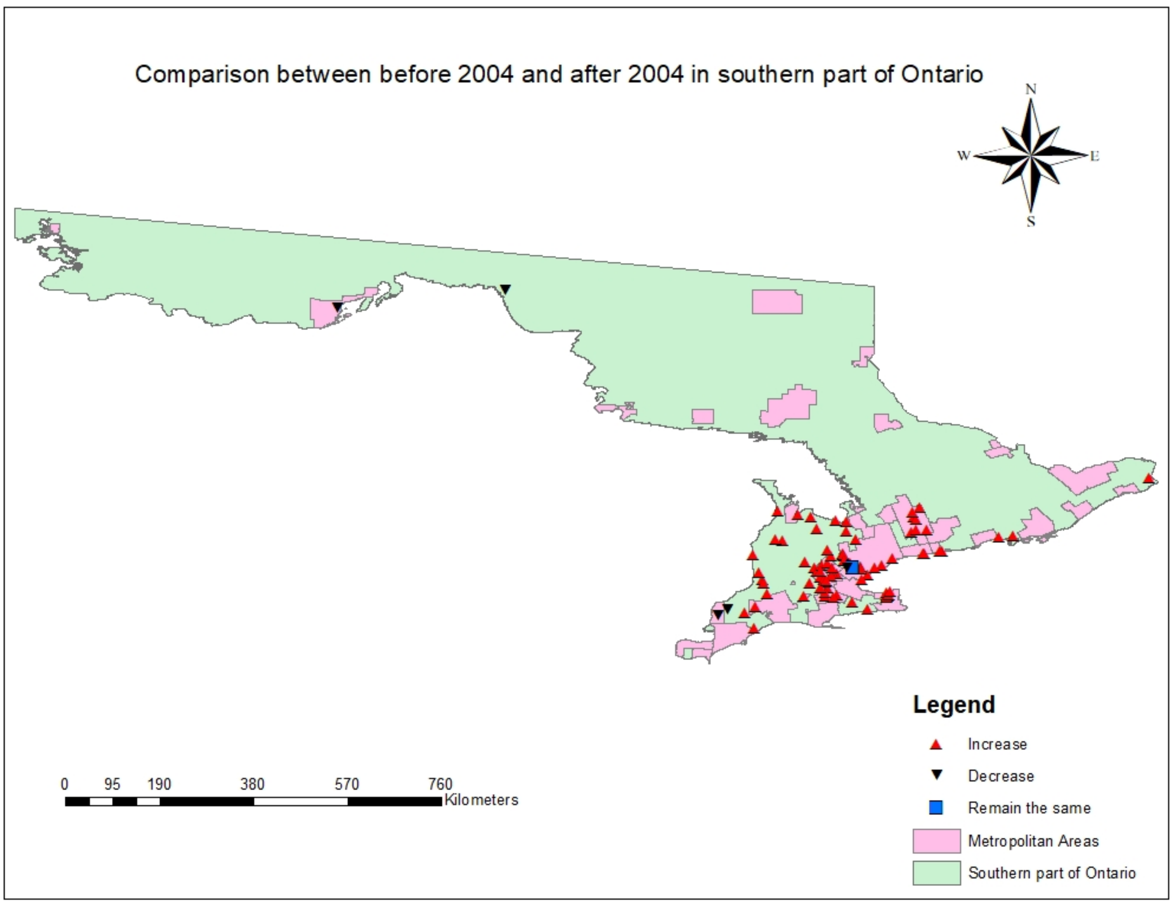



Figure 4: Comparison of the medians of chloride concentration between the two time periods of before 2004 and after 2004 for southern part of Ontario

## 4 CONCLUSION

In general, prominent seasonal variation in chloride concentration was observed at the majority of studied stations. Low concentrations were often measured in warm months, while high concentrations were measured in cold moths when the road salts are applied. Thus chloride concentration was often negatively dependent on flow. However different seasonal variation patterns and dependence of chloride on flow were also identified at some stations (especially on small rivers). In the time period of before 2004, significant upward trend of chloride was detected at many stations; while in the time period of after 2004, no trend and downward trend were detected at many stations, especially in Ontario. Although compared to the time period before the implementation of the Code, the chloride level (in terms of median of chloride concentrations) was elevated and the use of road salts also increased, the change from upward trends to downward/or no trends might imply the effectiveness of the Code. Furthermore, the analysis results confirmed that the anthropogenic activities often result in the increase of chloride in general; while flow can also be the primary or the secondary driving force of the detected temporal trend in chloride, especially in small rivers/creeks. Therefore to further investigate the role of the Code and formulate more efficient management strategy, more elaborate research in which the effects of various potential influential factors on chloride as well as the river physical characteristics (flow magnitude, river watershed, and anthropogenic activities in watershed) are taken into consideration is recommended.

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