Division Control No. ECD-112B

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TRACKING RIVER PLUMES WITH VOLATILE HALOCARBON CONTAMINANTS: THE ST. CLAIR RIVER - LAKE ST. CLAIR EXAMPLE

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NWRI CONTRIBUTION No. 85-83

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EXECUTIVE SUMMARY

TRACKING RIVER PLUMES WITH VOLATILE HALOCARBON CONTAMINANTS: THE ST. CLAIR RIVER - LAKE ST. CLAIR EXAMPLE.

K.L.E. Kaiser and M.E. Comba

Surface water samples from Lake St. Clair and the lower St. Clair River in June 1984 were analyzed for volatile halocarbon contaminants. The results indicate major sources of carbon tetrachloride and tetrachloroethylene along the eastern shore of the St. Clair River. Much smaller loadings of chloroform and associated haloforms and of trichloroethylene are contributed by several sources, including the St. Clair River, the Cutoff Canal and the Thames River.

RÉSUMÉ ADMINISTRATIF

DÉPISTAGE DES PANACHES DE POLLUTION D'HALOCARBURES VOLATILS : L'EXEMPLE DU LAC ST-CLAIR ET DE LA RIVIÈRE ST-CLAIR

K.L.E. Kaiser et M.E. Comba

En juin 1984, on a analysé des échantillons de l'eau du lac St-Clair et du segment inférieur de la rivière du même nom afin d'y déceler des halocarbures volatils. Les résultats ont révélé que les principales sources de tétrachloro-éthylène et de tétrachlorure de carbone se trouvaient le long de la rive est de la rivière St-Clair. On a aussi relevé de plus petites charges de chloroforme et d'haloformes associés et de trichloro-éthylène provenant de diverses sources, y compris la rivière St-Clair, le Cutoff Canal et la rivière Thames.

ABSTRACT

The concentrations of the seven widespread volatile halocarbon contaminants - tetrachlorethylene (C_2Cl_4), carbon tetrachloride (CCl_4), 1,1,1-trichloroethane ($C_2H_3Cl_3$), trichloroethylene (C_2HCl_3), chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$) and Freon 12 (CCl_2F_2) - were determined in the lower reaches of the St. Clair River and throughout Lake St. Clair. The results indicate large inputs of carbon tetrachloride and tetrachloroethylene from the St. Clair River, particularly the South Channel, Bassett Channel and Chenal Ecarte to Lake St. Clair. In addition, inputs of industrial chloroform, 1,1,1-$C_2H_3Cl_3$ and C_2HCl_3 are indicated from this source. Loadings of $CHCl_3$ and $CHBrCl_2$, reflecting chlorine treated waters are also observed to arise from the Cutoff Canal and the Thames River. Inputs of C_2HCl_3 are also noted from these two tributaries, while the concentrations of Freon 12 are almost constant throughout the lake, except for the Thames River area, where significantly higher levels of this compound are observed.

Additional Index Words: Volatile organic compounds, tracking techniques, water currents, chloroform, tetrachloroethylene, carbon tetrachloride.

Les concentrations de sept halocarbures volatils très répandus, tétrachloro-éthylène (C_2Cl_4), tétrachlorure de carbone (CCL_4), 1,1, 1-trichloro-éthane, ($C_2H_3Cl_3$), trichloro-éthylène (C_2HCl_3), chloroforme ($CHCL_3$), bromo-dichloro-méthane ($CHBrCL_2$) et fréon 12 (CCl_2F_2), ont été décelés dans le segment inférieur de la rivière St-Clair et partout dans le lac du même nom. Les résultats révèlent que la rivière St-Clair véhicule de fortes charges de tétrachlorure de carbone et de tétrachloro-éthylène, plus particulièrement en provenance du South Channel, du Bassett Channel et du Chenal Ecarte, lesquelles se déversent dans le lac St-Clair. De plus, des charges de chloroforme industriel 1,1,1-$C_2H_3Cl_3$ proviennent aussi de cette source. Le Cutoff Canal et la rivière Thames sont à l'origine de charges de $CHCl_3$ et de $CHBrCl_2$, traduisant le traitement des eaux par le chlore. Des charges de C_2HCl_3 ont aussi été imputées à ces deux affluents, alors que les concentrations de fréon 12 sont presque constantes partout dans le lac, sauf dans la région de la rivière Thames, où des niveaux nettement plus élevés de ce produit composé ont été observés.

Autres mots-clés : Composés organiques volatils, techniques de dépistage, courants, chloroforme, tétrachloro-éthylène, tétrachlorure de carbone.

INTRODUCTION

Volatile halocarbon contaminants have been shown to be widespread in surface waters of the Great Lakes (Kaiser et al. 1985, 1983; Kaiser and Comba 1983; Kaiser and Valdmanis 1979). In the U.S.A. alone, the production of eight of the most common compounds, Freon 11, Freon 12, CH_2Cl_2, CCl_4, C_2HCl_3, C_2Cl_4 and 1,1,1-$C_2H_3Cl_3$ were in the order of $1.5.10^9$ $kg.a^{-1}$ in 1982 (Kaiser et al. 1985). As their use comprises many different industrial processes, most industrial and municipal effluents contain elevated levels of the compounds.

Recently, Comba and Kaiser (1984) have shown the possibility of tracking tributary plumes in receiving lakes on the basis of volatile halocarbon concentrations. In contrast to physical measurements of currents at selected stations over long periods of time, which provide detailed but expensive information, a volatile contaminant survey can be undertaken with a minimum of resources and time and results in a nearly synoptic view of the integrated, sources/currents/sinks relationships under the conditions prevailing at the time of sampling. For a lake the size of St. Clair with a surface area of approximately 1200 km^2 (Canada Water Year Book 1977-1978), a volatile halocarbon surface water survey can be carried out by a small launch and three to four persons in approximately one week. Such a survey can cover a station grid of up to one hundred stations and includes field processing of the samples but not analysis. The latter would

take a skilled analyst approximately two week time, given the availability and functioning of a dedicated gas chromatograph. This level of resources required compares favourably with the cost of deployment of numerous current meters and the personnel and time involved to analyze the resultant data. However, the results of a volatile halocarbon survey are not necessarily representative of the plume patterns under other atmospheric (wind direction and strength) conditions. Therefore, to fully describe the natural variations of such plumes, several volatile halocarbon surveys would have to be undertaken.

The St. Clair River/Lake St. Clair/Detroit River system has become a focal point for research studies on the Great Lakes' Connecting Channels (Chau et al. 1985). Moreover, both the St. Clair and Detroit Rivers are recipients of large amounts of industrial waste products, including halocarbons and, therefore, the St. Clair River/ Lake St. Clair system should be particularly amenable to plume system analysis of volatile halocarbon concentrations in the surface water of the shallow and generally well-mixed Lake St. Clair. We report here on an application of that method to the lower reaches of the St. Clair River and to Lake St. Clair in a one-week study in June 1984.

EXPERIMENTAL

Surface water samples (1 m) of 200 mL size were collected from a Monark launch at a total of 67 stations in the lower St. Clair River

and in Lake St. Clair between 18 June and 21 June 1984. All samples were kept in darkness and as cool as possible and were processed at a mobile processing unit at the shore within approximately four hours from collection according to the procedure reported by Comba and Kaiser (1983). The sampling locations are shown in Figure 1.

For the gas chromatographic analysis, 500 µL size injections of the headspace samples were analyzed on a Hewlett Packard 5700 gas chromatograph using an electron capture detector. Split/splitless conditions were employed with a splitless delay time of 10 seconds. Cryogenic temperatures from -20°C for 2 min to 80°C at 4°C/min were used for chromatographic separation on a 30 m OV-101 fused silica capillary column. Further details as to precision and accuracy of the method are given by Comba and Kaiser (1983).

RESULTS AND DISCUSSION

Hydrology

Lake St. Clair is a very shallow lake with mean and maximum depths of approximately 5 to 6 m. This low depth, together with frequent winds of 10 to 15 knots prevents any stratification and, therefore, surface water samples (1 m) are usually representative of the entire water column. In contrast, both the major inflow to the lake, the St. Clair River and the outflow, the Detroit River, have mean depths of 10 to 15 m. In order to facilitate navigation, several of the channels at the mouth of the St. Clair River are frequently dredged to maintain minimum depths of approximately 10 m. This is also the case for the main navigation channel through Lake St. Clair, thus connecting the Detroit and St. Clair Rivers for navigation purposes.

The mouth of the St. Clair River, downstream from Baby Point near Port Lambton spreads out into seven major channels. Of these, the Chenal Ecarte enters the lake furthest east, and the North Channel enters the lake furthest to the northwest in Anchor Bay. The mouths of these two channels are separated by a distance of over 30 km. Each of the seven major channels conduits a relatively constant proportion of the total St. Clair River flow into the lake. Table 1 gives an overview of these contributions.

In contrast to, for example, the Detroit River, where horizontal and vertical mixing of incoming tributary and effluent plumes is comparatively rapid due to many and pronounced changes in river width and depth and due to embayments, islands and many bends in the course of the river, the St. Clair River provides for little horizontal mixing. In terms of the hydraulic regime, the St. Clair River may be viewed as a large diameter trough with essentially laminar flow throughout. Evidence for this interpretation can be found in water quality surveys which indicate a continuous zone of contamination, for example with phenols, of the eastern nearshore water of the river water between Sarnia and Port Lambton (IJC 1968). Consequently, effluents discharged into the upper St. Clair River along the eastern shore will enter Lake St. Clair predominantly through the South Channel and the other channels to the east of that. Naturally then, discharges along the western river shore will enter the lake, mostly through the channels west of the South Channel. Further support for this interpretation can be seen from the distribution of benthos in St. Clair River as observed in 1968 (OME 1979).

Freon 12

Dichlorodifluoromethane (CCl_2F_2, Freon 12) is an important chlorofluorocarbon. Its major uses are in the manufacturing of foam as blowing agent, as propellant in aerosols and as working fluid in

refrigeration systems. Due to its volatility (boiling point -30°C), higher concentrations of Freon 12 dissolved in water will rapidly evaporate into the air. At lower concentrations, a more or less stable equilibrium between the levels in air and water will prevail. Freon 12 concentrations in central and eastern Lake Erie water were found to be at 76±38 ng L^{-1} in 1978 (Kaiser and Valdmanis 1979). Our present data on Freon 12 levels in Lake St. Clair (Figure 2) indicate an extremely uniform distribution of this compound. Except for the four stations (#243 to 245, 247) at the Thames River mouth and nearby, the levels at all other stations were virtually identical. For these 60 stations, a mean ± standard deviation of 32±3 ng.L^{-1} is observed. The much higher concentrations at the mouth (380 ng.L^{-1}) and immediately outside the Thames River obviously indicate a source within the river.

Chloroform and $CHBrCl_2$

Chloroform ($CHCl_3$) and bromodichloromethane ($CHBrCl_2$) are produced on the chlorination of potable and waste waters. As previously shown, the ratio of the chlorination derived concentrations of $CHCl_3/CHBrCl_2$ is primarily a function of the bromide ion concentration of the potable raw or waste water, respectively (Kaiser et al. 1983; Comba and Kaiser 1983). The distribution of both compounds in Lake St. Clair is shown in Figures 3 and 4.

Chloroform concentrations (Fig. 3) vary between approximately 10 and 300 $ng.L^{-1}$. Essentially four zones of elevated $CHCl_3$ are observed. The largest of these is emanating from the Bassett, South and Ecarte Channels and stretches in a southwesterly direction across the lake. Within this plume, a zone of higher concentrations is found in the area of the major shipping channel between stations 250 and 208. Similar levels (approximately 100 $ng.L^{-1}$) are found again in the lower reaches of the St. Clair River, stations 270 to 272 and 212 to 215. It appears that the higher levels between stations 250 and 208 and at those in the lower river represent intermittent increases on a background of elevated levels which are continuously present in the river. Possibly the zone of higher levels at stations 221 to 223, 235 and 249 also is a remnant of such spikes in the river, which may have shifted more to the south due to currents in the lake.

Two other zones of strongly increased $CHCl_3$ levels are found to the east of the Clinton River/Cutoff Canal mouths and along the southern lakeshore west of the Thames River mouth. These two areas are obviously influenced by contamination of the noted tributaries. The elevated $CHBrCl_2$ concentrations and $CHCl_3/CHBrCl_2$ ratios of approximately 4.0 confirms the contributions of mainly water chlorination derived chloroform in these areas. In contrast, however, only background levels of $CHBrCl_2$ are found in the lower reaches of the St. Clair River. For these stations (209 to 219) $CHCl_3/CHBrCl_2$ ratios of over 8.0 are observed consistently, indicating significant

contributions of industrial source chloroform to the total chloroform burdens.

1,1,1-Trichlorethane

1,1,1-Trichloroethane (TCA) levels in Lake St. Clair are shown in Fig. 5. They indicate a strong zone of contaminated water emanating from the St. Clair River through the South Channel and also from Chenal Ecarte. The area of elevated levels covers approximately one fifth of the lake and is detected in the triangle between stations 260 in the NW, 208 in the SW and 227/228 in the east.

Additional high levels of TCA are found off the mouths of the Cutoff Canal and Clinton River. As for chloroform, inputs of TCA from these tributaries is apparent.

Trichloroethylene

Trichloroethylene (TCE) concentrations are shown in Fig. 6. As for TCA, significantly higher concentrations are observed at stations within and off the mouth of the South Channel and Chenal Ecarte, the Cutoff Canal, and the Thames River. Compared to TCA, however, the TCE contamination is also observed to stretch to stations off the mouth of the North Channel. In addition, the Thames River appears to be a source of this compound as evident from the elevated levels at stations 243, 244 and 246.

Tetrachloroethylene

Tetrachloroethylene (TECE) levels (Fig. 7) again indicate the flow of strongly contaminated St. Clair River water into Lake St. Clair. Concentrations of several hundred $ng.L^{-1}$ TECE were observed at stations 238 and 239 off the mouth of the South Channel and at station 227 off Chenal Ecarte. The distribution of elevated TECE is generally similar to those of TCA and TCE. However, the zone of contamination is larger, extending further into the lake than those of TCA and TCE and at the affected stations the TECE concentrations are approximately ten times those of TCA or TCE.

Comparatively small inputs of TECE also originate from Cutoff Canal and the Thames River. The large zone of slightly elevated TECE levels in the southwestern portion of Lake St. Clair is thought to represent a remnant of a watermass previously located off the mouth of the St. Clair River. This interpretation is consistent with the observations for chloroform (Fig. 3) and carbon tetrachloride (Fig. 8).

Carbon Tetrachloride

Carbon tetrachloride (CTC) concentrations are shown in Fig. 8. Strongly contaminated river water is found for all stations in the South Channel. In Lake St. Clair, the area of contamination is

similar to those for TCA, TCE and TECE with the hightest CTC concentrations observed just off the mouths of the South Channel and Chenal Ecarte. Except for a plume of elevated CTC at stations near the south shore of the lake which is similar to that for TECE in that area, all other stations showed only very low levels of CTC. Notably, there is a complete absence of higher CTC levels off the mouths of any of the tributaries to Lake St. Clair other than the southern and easterly arms of the St. Clair River delta. This clearly indicates that the St. Clair River is the principal source for these strongly elevated levels of CTC in the lake.

Loadings

Given the flows through the various St. Clair River channels (Table 1), the daily contaminant loadings can be estimated. For example, Figure 7 and 8 clearly indicate the major inputs of TECE and CTC to be through the South Channel and Chenal Ecarte. The combined flow of these channels represents approximately 50 percent of the total river flow. On the basis of the 13 stations within and immediately outside these channels (stations 210 to 215, 217 to 219, 227, 228, 238 and 239), and using one half of the total river flow, the following concentration means and standard deviations and daily loadings are derived: TECE, 209±125 $ng.L^{-1}$ (48 $kg.day^{-1}$); CTC, 325±267 $ng.L^{-1}$ (75 $kg.day^{-1}$); TCA, 52±22 $ng.L^{-1}$ (12 $kg.day^{-1}$); and

TCE, 17±6 $ng.l^{-1}$ (4 $kg.day^{-1}$). These values may be considered as minimum estimates which do not account for any losses through evaporation and/or degradation of contaminants between their sources and the lake.

Using the data for all 67 stations, a lake mean concentration of 83±167 $ng.L^{-1}$ CTC is found. With an area of 1200 km^2, a mean depth of approximately 5 m, a lakewide burden of approximately 500 kg CTC is calculated. The corresponding burdens are 360 kg TECE, 120 kg TCA and 60 kg TCE, respectively. As mentioned elsewhere (Comba and Kaiser 1985), these four compounds are multipurpose industrial solvents. Their presence in Lake St. Clair water at a total burden of approximately 1000 kg indicates a significant discharge to the lake via the St. Clair River.

In summer, due to its shallow basin, the water temperature in the lake increases rapidly to reach a temperature of approximately 20°C throughout the water column (AES 1984). At this water temperature, the loss to the atmosphere must be considerable. For example, Dilling et al. (1975) have shown half lifes of 20 min for TCA, 21 min for TCE, 25 min for TECE, and 29 min for CTC, respectively, for aqueous solutions containing 1 $mg.L^{-1}$ of each of these compounds. Similarly, Mackay and Leinonen (1975) concluded that many of the investigated compounds such as benzene, Lindane and polychlorinated biphenyls evaporate rapidly from solution. Also Schwarzenbach et al. (1979) concluded from a one-year study on Lake Zurich that the mass transfer

to the atmosphere is the predominant elimination mechanism for TECE. This rapid loss is further substantiated by the strong concentration gradients observed for CTC and TECE in the lake downstream from the major source of input, the South Channel. For example, the levels of approximately 900 $ng.L^{-1}$ CTC at stations 238 and 239 drop off within a distance of 10 to 20 km to near background levels of 5 to 20 $ng.L^{-1}$. It is assumed that these background levels represent the equilibrium concentrations between the water and the overlying air under the conditions prevailing at the time of sampling.

Effects on Biota

In terms of acute toxicity to aquatic organisms, even the highest observed levels of several hundred $ng.L^{-1}$ CTC or TECE are several orders of magnitude below any effect levels. For example, acute lethal concentrations to fathead minnows in 96-hr flow-through tests were 53 $mg.L^{-1}$ for TCA, 41 $mg.L^{-1}$ for TCE, and 18 $mg.L^{-1}$ for TECE at 12°C, respectively (Alexander et al. 1978). However, both the toxicity levels and the evaporation rates of these compounds are likely to be significantly different under different temperature regimes. For example, Lake St. Clair normally freezes over in the January to March period. Therefore, evaporation of the observed halocarbons will be significantly reduced, probably by one to three orders of magnitude. At the same time, aquatic organisms, subject to

slower metabolism and less abundant food may be more sensitive to these contaminants. It would therefore be of considerable interest to repeat this study under conditions of complete ice cover of the lake.

No water quality criteria for the protection of public surface water supplies appear to exist for some of the observed volatile halocarbons. However, in the USA, certain regulations regarding the treatment, technology and potable water are in effect. Among these, removal of chloroform and other trihalomethanes to acceptable levels are stipulated. While some of the observed compounds are suspected animal or human carcinogens (RTECS 1981-82) present toxicological information on the possible effects of long-term exposure to sublethal concentrations of these compounds is inconclusive. It may be assumed, however, that the removal of haloforms would also result in the removal or at least reduction of the other volatile halocarbons.

Plume Characteristics

Plumes of contaminated river water in receiving lakes have previously been described on the basis of persistent contaminant concentrations (Fox and Carey 1986; Comba and Kaiser 1984; Kaiser et al. 1983; Fogelqvist et al. 1982; Kaiser and Valdmanis 1979). It has been recognized that, due to the simplicity, speed and low cost, such investigations are of significant value. Though neither intended nor able to replace the more elaborate and time

consuming traditional physical measurements, such contaminant surveys are useful in collaboration with other more in-depth studies over longer periods. For example, our results clearly indicate the existence of contaminant plumes along the eastern shore of the St. Clair River. These plumes enter Lake St. Clair primarily through the South Channel and Chenal Ecarte and indicate little horizontal mixing across the river. Therefore, it is to be assumed that the source(s) for such contaminants are along the eastern shore of the river.

In Lake St. Clair, the plumes typically indicate an area of high contamination in the quadrangle between stations 206 and 219 in the west and 241 and 227 in the east, as shown, for example for TECE, Fig. 7. In addition, a southwesterly movement of the plume coming from the South Channel, essentially along the navigation channel through Lake St. Clair is indicated by the distribution of chloroform, Fig. 3.

Inputs from the Thames River appear to move along the south shore in a westerly direction as shown by the plumes for $CHCl_3$ (Fig. 3), $CHBrCl_2$ (Fig. 4), TCE (Fig. 6), and Freon 12 (Fig. 2). The plumes of the Clinton River and Cutoff Canal tributaries to Lake St. Clair appear to move in an easterly to northeasterly direction, as evident from the distributions of chloroform (Fig. 3), $CHBrCl_2$ (Fig. 4) and TCA (Fig. 5).

Recently, Ibrahim and McCorquodale (1985) described circulation models for Lake St. Clair. Our data on volatile halocarbon

distributions in the lake by and large appear to be in agreement with the presented submodel for vertically integrated velocities under southeasterly winds. In fact, for two out of the four days of sampling, strong southerly to southeasterly winds were prevalent. The above model is based on field observations on chloride ion distributions in the lake and it is important to note that the mean chloride ion concentrations in the various St. Clair River channels into Lake St. Clair also showed significantly higher levels for the South Channel and Chenal Ecarte. Therefore, these models should also be representative for the observed volatile halocarbon contaminants whose major sources are also indicated to be along the eastern side of the St. Clair River.

ACKNOWLEDGEMENTS

We thank Mr. John A. Kraft, Technical Operations Division, NWRI, for assistance in the sample collection and Mr. Terry Velenosi, Environmental Contaminants Division, NWRI, for assistance in the sample processing and analysis.

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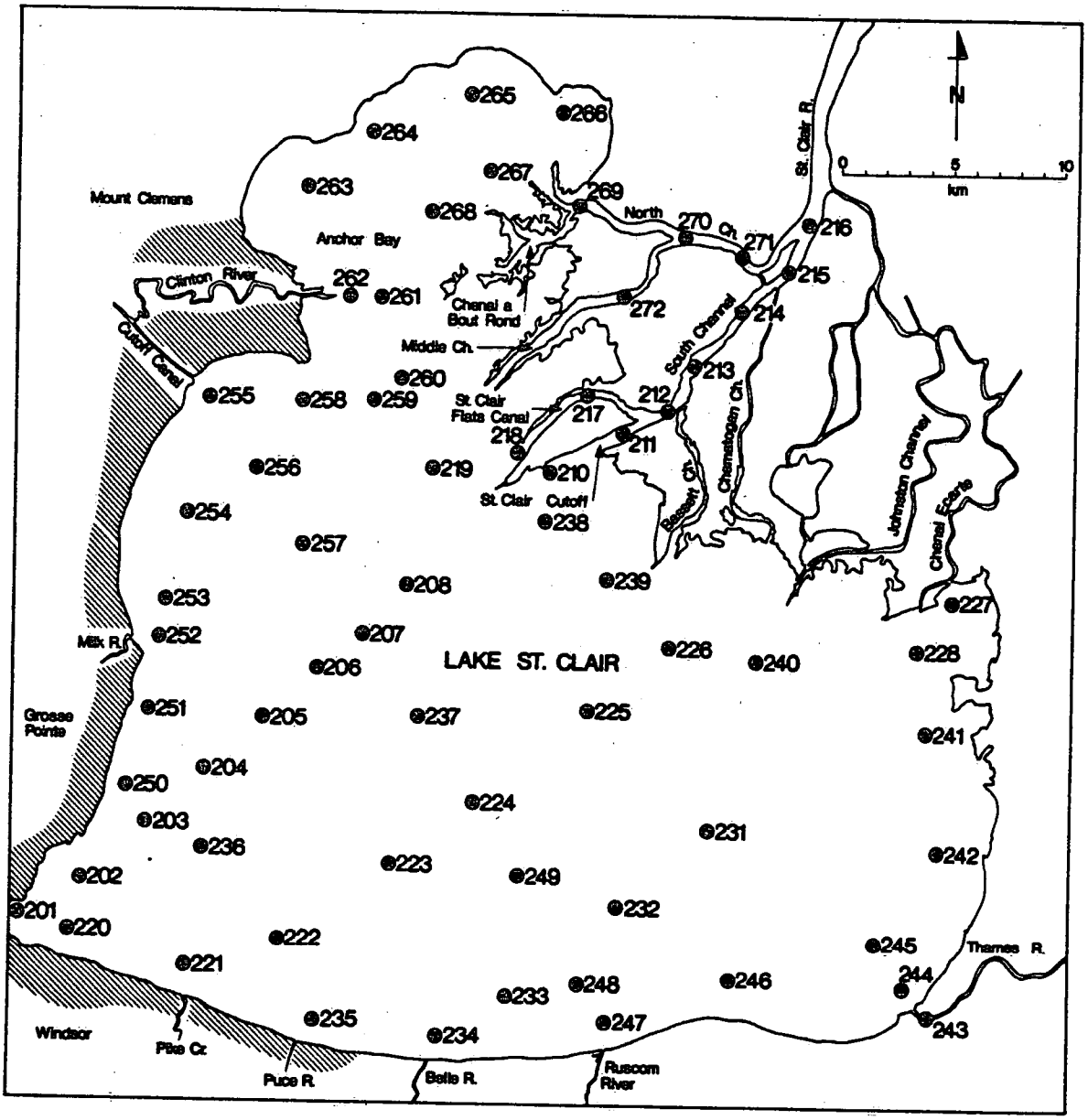
LIST OF FIGURES

Table 1. Major St. Clair River delta channels and their mean flows and proportions of total river flow. Adapted from Ibrahim and McCorquodale (1985).

Channel	Flow		
	$m^3.sec^{-1}$		% of Total
North Channel	1750		33
Northern arm	1350[a]		25
Chenal a Bout Rond	400[a]		7
Middle Channel	1060		20
South Channel	1950		36
St. Clair Flats Canal	1000[a]		19
St. Clair Cutoff	950[a]		18
Bassett Channel	280		5
Chematogan Channel	50[a]		1
Johnston Channel	120[a]	} 260	2
Chenal Ecarte	140[a]		3
Total	5350		100

a: estimate


LAKE ST. CLAIR
Mount Clemens
Anchor Bay
Clinton River
Cutoff Canal
Middle Ch.
Chenal a Bout Rond
North Ch.
St. Clair R.
South Channel
St. Clair Flats Canal
St. Clair Cutoff
Belle River Ch.
Chamatogen Ch.
Johnston Channel
Chenal Ecarte
Grosse Pointe
Milk R.
Windsor
Pike Cr.
Puce R.
Belle R.
Ruscom River
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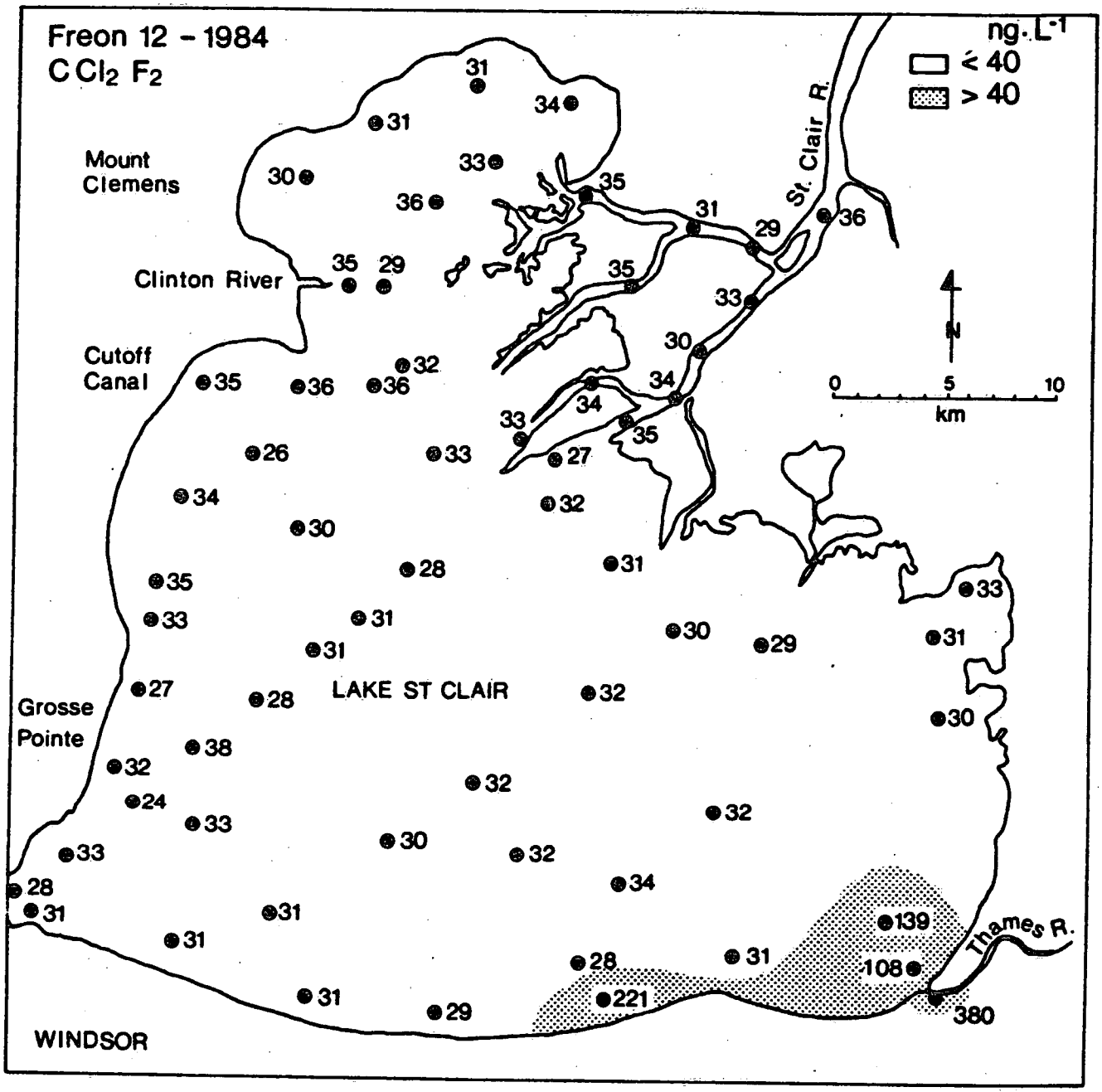


Figure 2. Freon 12 concentrations in Lake St. Clair surface water.

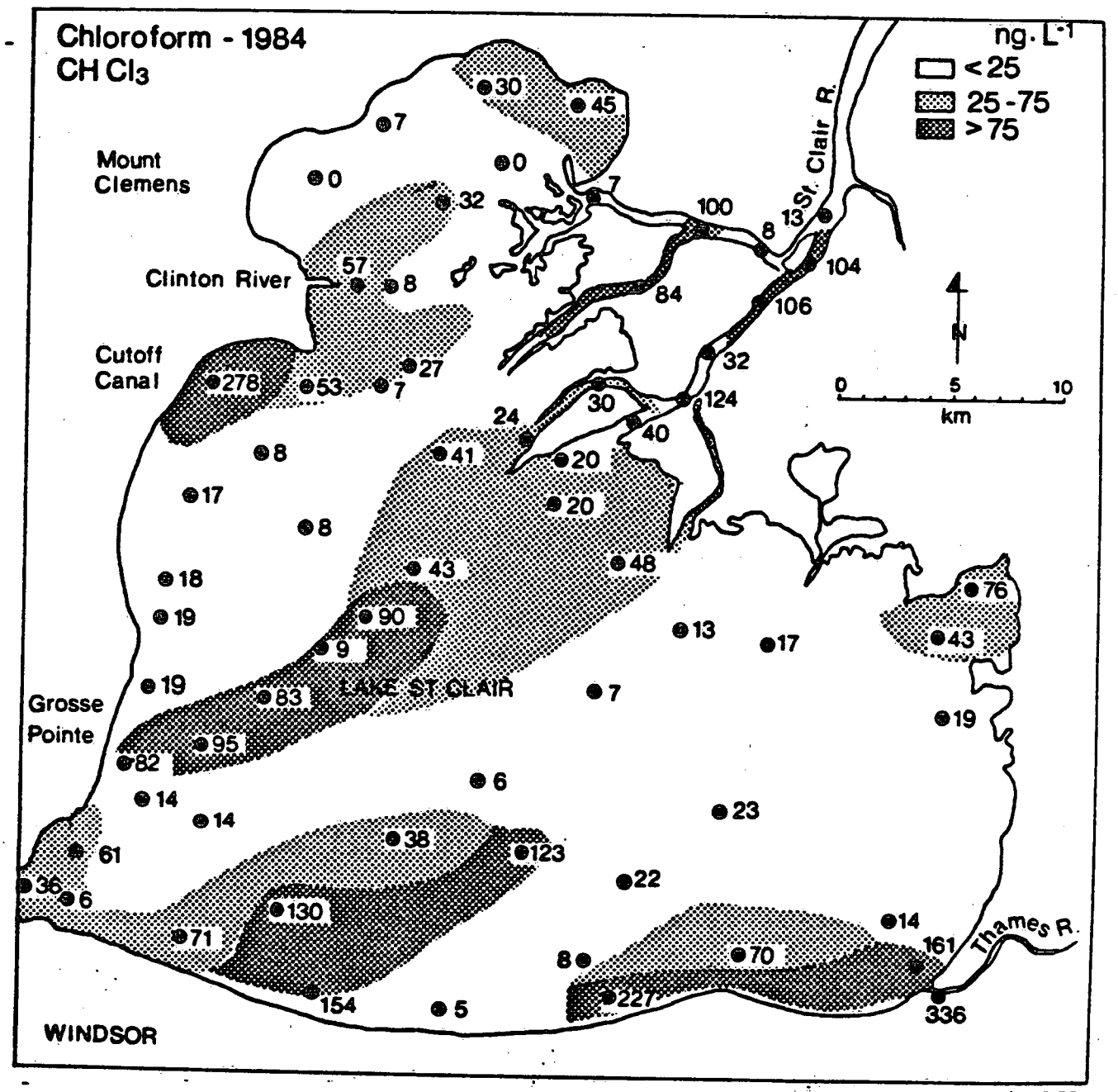
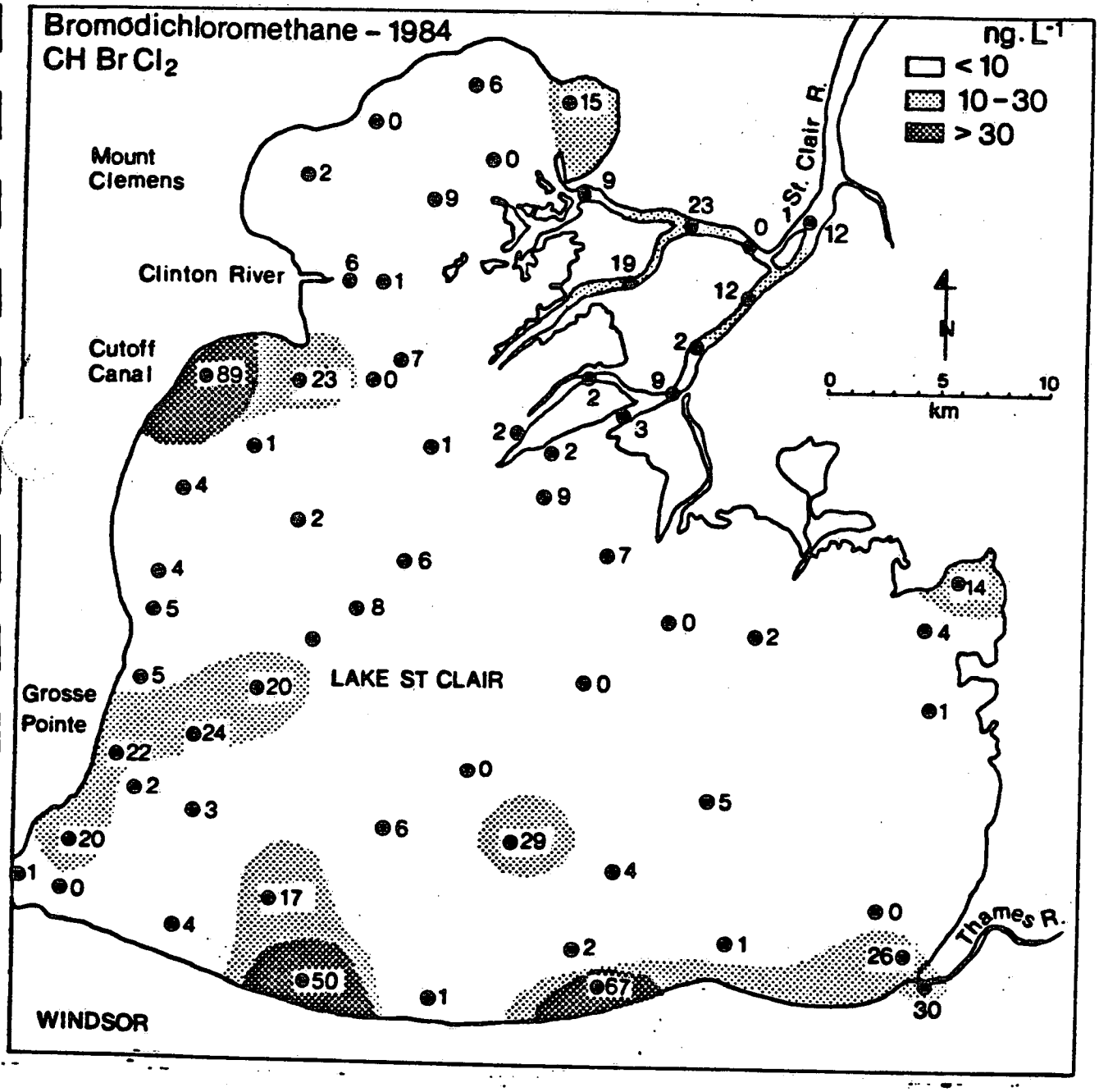


Figure 3. Chloroform concentrations in Lake St. Clair surface water.



...re 4. Bromodichloromethane concentrations in Lake St. Clair surface water.

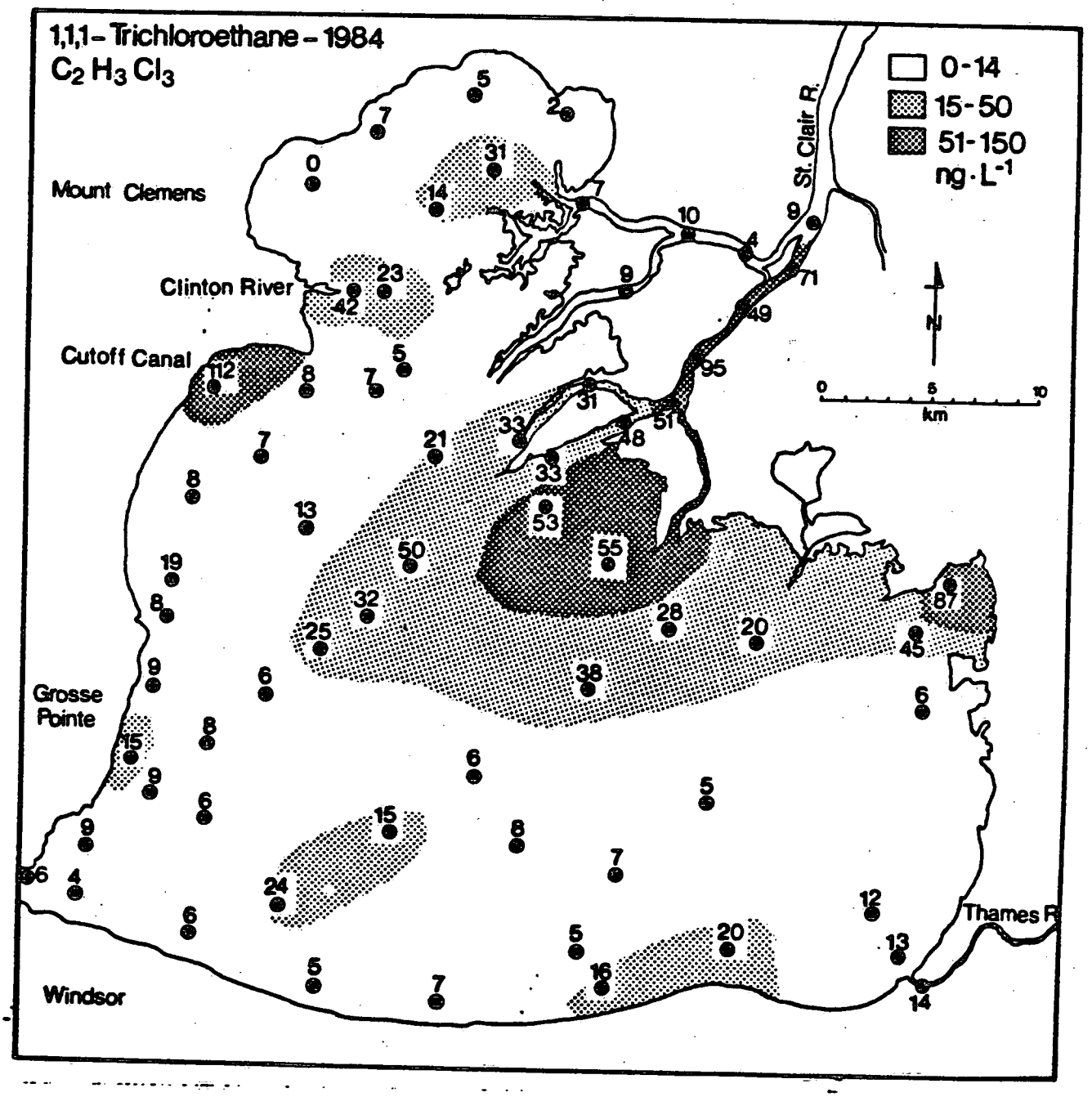


Figure 5. 1,1,1-Trichloroethane concentrations in Lake St. Clair surface water.

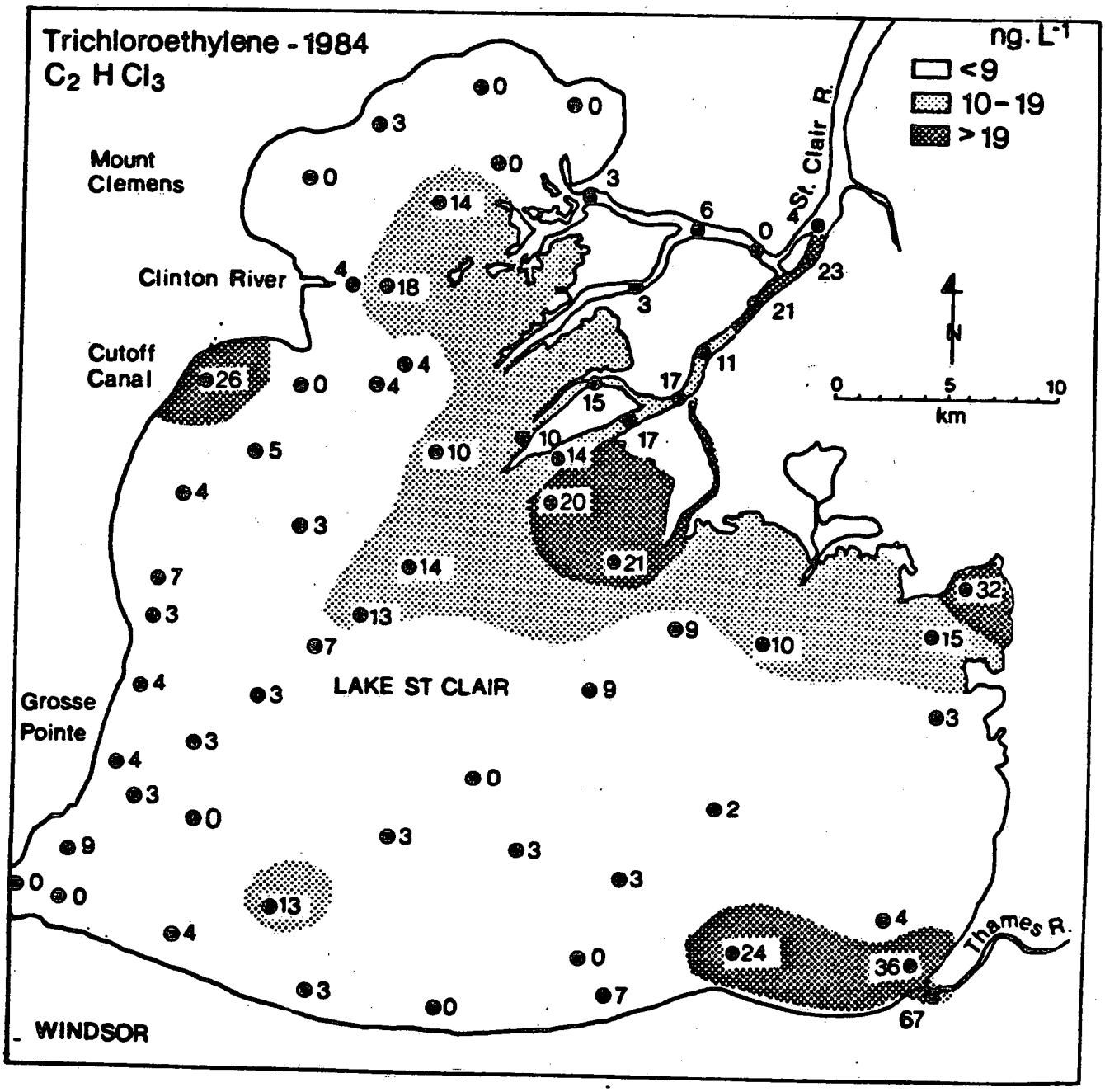


Figure 6. Trichloroethylene concentrations in Lake St. Clair surface water.

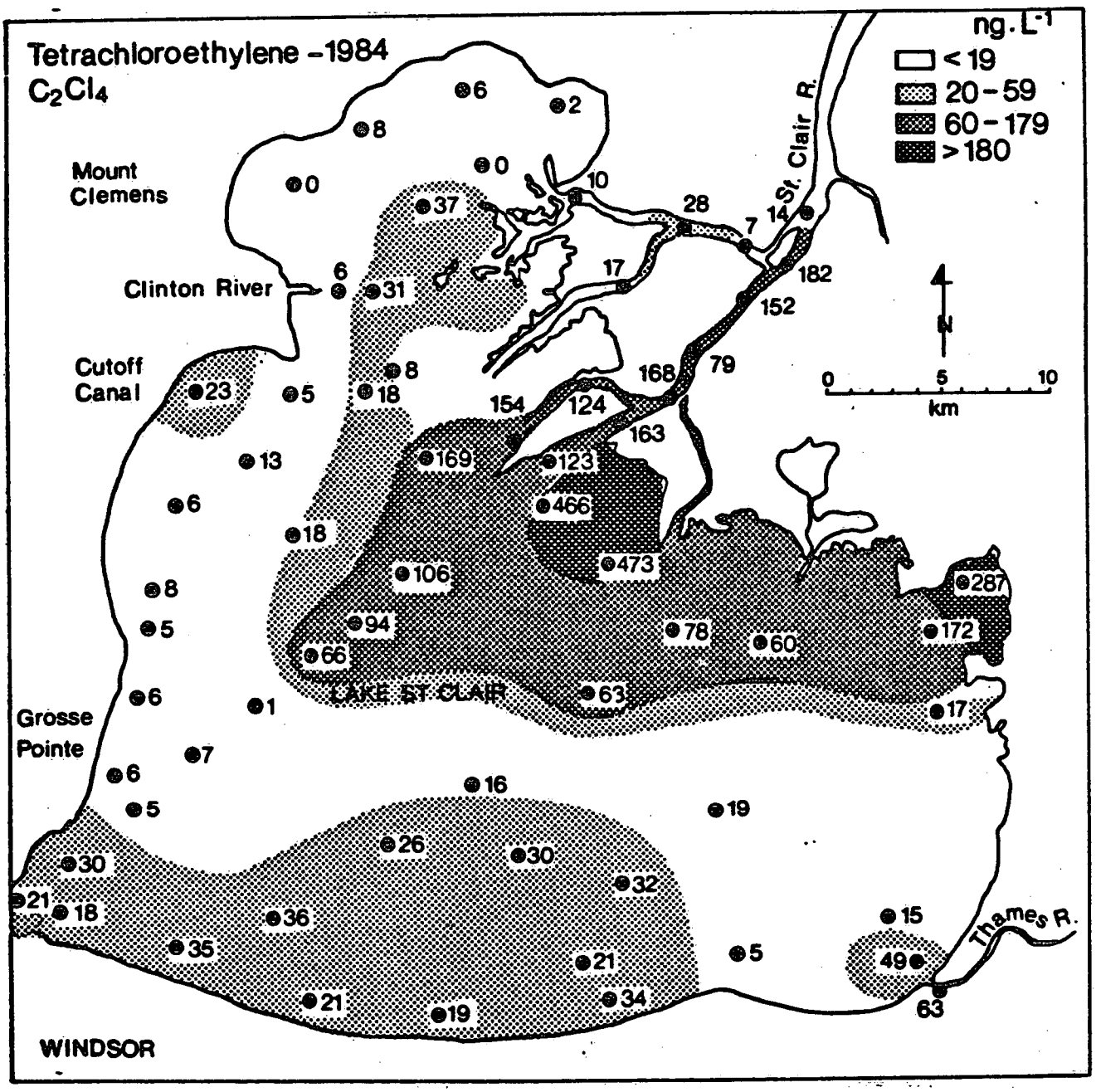


Figure 7. Tetrachloroethylene concentrations in Lake St. Clair surface water.

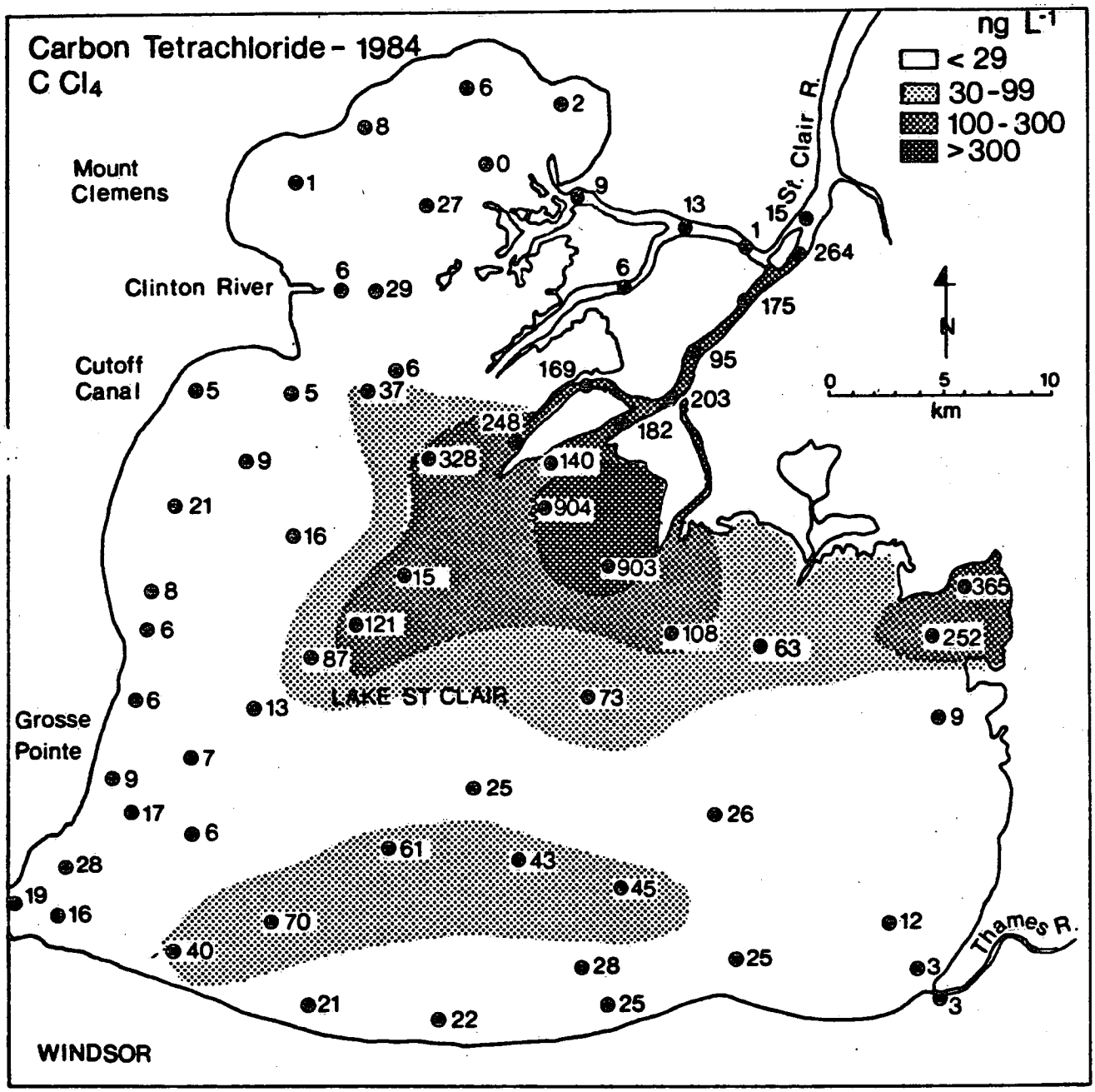


Figure 8. Carbon tetrachloride concentrations in Lake St. Clair surface water.