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Trace Metals in Lake Ontario, 1979

Melanie A. Neilson

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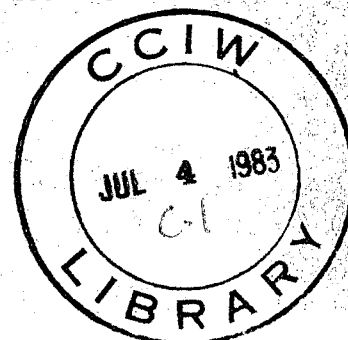
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Abstract

During 1979, three cruises were conducted on Lake Ontario to measure total trace metal concentrations on the open lake. With the exception of five isolated incidents (two for cadmium and three for iron), the lake was found to be in compliance with the objectives of the 1978 Water Quality Agreement.

Nearshore values were found to be higher than those of the open waters, with metal concentrations generally greater at the western end of the lake. To define this spatial variation, a statistical procedure was used to divide Lake Ontario into zones for each of the individual trace metals. A composite map was compiled from this information.

For each metal, on a cruise-by-cruise basis, depth-weighted concentrations were calculated for both the surface-to-20-metre stratum and the entire water column. No significant ($P \geqslant 0.05$) differences between the surface and the whole water column were found during isothermal conditions. However, during stratified conditions in late summer, significantly lower surface values were found for arsenic, chromium and iron.

Résumé

En 1979, trois levés ont été effectués dans le lac Ontario pour mesurer les concentrations totales des métaux à l'état de traces dans la zone libre. Exception faite de cinq cas isolés (deux pour le cadmium et trois pour le fer), le lac satisfaisait aux objectifs de l'Accord relatif à la qualité de l'eau de 1978.

Les concentrations à proximité des rives étaient plus élevées que dans la zone libre et étaient généralement plus élevées à l'extrémité occidentale du lac. Pour déterminer la variation spatiale, on a employé une méthode statistique divisant le lac en plusieurs zones selon la concentration de chacun des métaux, puis on a établi une carte globale pour l'ensemble de tous les métaux.

On a calculé, pour chaque métal et chaque levé, les concentrations pondérées en fonction de la profondeur pour la tranche de la surface jusqu'à 20 mètres de profondeur et pour toute la colonne d'eau. On n'a trouvé aucune différence significative ($P \geqslant 0.05$) entre la surface et toute la colonne d'eau en période d'isothermie. Toutefois, en période de stratification thermique (fin de l'été), des valeurs significativement plus faibles ont été obtenues pour l'arsenic, le chrome et le fer à la surface.

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INTRODUCTION

The Canada Centre for Inland Waters carries out annual surveys on Lake Ontario to monitor the levels of various physical, chemical and biological parameters. In 1979, three synoptic trace metal cruises were conducted, during both isothermal and stratified conditions. Samples were collected at surface (1 m) and discrete depths at 47 stations (Fig. 1). In the past, investigators studying trace metal levels in the waters of the Great Lakes (1,2,3,4) reported on filtered and/or unfiltered/nondigested samples. In compliance with the 1978 Great Lakes Water Quality Agreement (5), the current study employed revised analytical techniques to measure total metal concentrations and thus identify incidences of noncompliance with water quality objectives.

To describe the horizontal distribution of the data, the temporal and spatial variabilities were calculated so that the lake could be mapped into statistically significant trace metal zones. The vertical distribution was then studied by comparing the concentrations found in the surface-to-20-metre stratum with those of the entire water column.

MATERIALS AND METHODS

The cruises were conducted April 30 to May 4 (pre-stratified), May 28 to June 1 (early-stratified) and August 27 to August 31 (late-stratified). Analyses were done for total cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), zinc (Zn), non-filtered aluminum (Al), arsenic (As), mercury (Hg), selenium (Se) and silver (Ag) based on the methods outlined in the *Analytical Methods Manual*, 1979 (6). Samples were collected by a winch-operated Rosette Multi-Bottle Array model 1015 Mark V Sampler (General Oceanics Limited, Miami, Florida), interfaced with a Guideline EBT model 8301B probe, which gives temperature and depth readings. This submersible bottle mounting array enables the operator to remotely actuate a sequence of Niskin sampling bottles. Depths sampled were as shown in Table 1.

Total metal samples were digested with 1% HNO_3 and evaporated to 5 mL. The Cd, Cr, Cu, Fe, Pb, Ni and Zn concentrations were then determined by atomic absorption spectrophotometry after chelation with ammonium pyrrolidine dithiocarbamate (APDC) and subsequent extraction with the solvent methyl isobutyl ketone (MIBK). Oxine (8–hydroxyquinoline) was used to complex manganese before spectrophotometric determination. Arsenic and selenium were measured by flameless atomic absorption, and mercury, by cold vapour atomic absorption. The method for EDTA-chelation of silver and subsequent spectrophotometric analysis was a modified version of that method outlined in the *Analytical Methods Manual*, 1979 (7). Aluminum was not digested, as it does not exist in forms extractable by hot acid (8).

In the analysis of the data, all values reported at less than the detection limit were taken at one-half the detectable level.

A statistical procedure developed by El-Shaarawi and Shah (9) was used to zone the lake according to concentration levels of the different metals. The method involves a hierarchical classification of the surface (1 m) data, based on temporal and spatial components. The F-values are calculated to test the hypothesis of any significant differences between cruises (temporal) and/or stations (spatial). Where there was found to be no significant

Table 1. Sampling Depths for Trace Metals, Lake Ontario, 1979

Unstratified	Stratified
1 m	1 m
10 m	1 m above thermocline
25 m	1 m below thermocline
Bottom − 10 m	Bottom − 10 m
Bottom − 2 m	Bottom − 2 m

NOTE: If station depth was less than 25 m, only three depths were sampled: 1 m, 10 m, bottom − 2 m. If station depth was 50 m or less, four depths were sampled: 1 m, 10 m, 25 m, bottom − 2 m.

difference between sampling sites, the lake was regarded as a single zone. However, if the F-test showed spatial differences, the lake was divided into more than one zone. Those stations most frequently reporting the highest concentration of a particular metal fell into zone 1. Zone 2 stations reported the next highest concentrations, and so on.

A composite map was then constructed to describe various regions of the lake. In assigning stations to the zones of the composite map, an accumulated total point system was used. Any station occurring in the lowest-concentration zone for a particular metal was scored 1 point. Stations classified in the next higher concentration zone were scored 2 points, and so on in succession. A station was awarded points for each metal and the accumulated total was recorded. A score of 43 points would signify that the station was found in zone 1 (the highest-concentration zone) for all the metals. To construct the composite map, any station scoring an accumulated point total of 26 (of the 43 maximum) or more (60%) was designated to zone 1, to zone 2 if at least 22 points (50%), to zone 3 if at least 17 points (40%), and to zone 4 if less than 40%.

To examine the vertical distribution, depth-weighted concentrations for surface-to-20-metre stratum and the entire water column (surface-to-bottom) were calculated for each station for all three cruises. The Student's t test was run to determine whether there was any statistical difference in the top 20 m versus the entire water column.

RESULTS AND DISCUSSION

The metals can be classified into three categories: (1) those that were consistently reported at below the detectable level (cadmium, lead, mercury and silver); (2) those of concentrations exceeding the detection limit for all or most of the samples collected (aluminum, arsenic, copper, chromium, iron, manganese, nickel and zinc); and (3) those undetected in spring but found in late summer (selenium). Fitzsimons and Whittle (10), in analyzing the supernatant of centrifuged samples collected at the Kingston Basin in Lake Ontario, reported similar results for selenium. Samples collected before and after August had selenium concentrations at or below the detection limit; in August a mean value of 0.20 μg/L was found (11).

Table 2 gives the mean, standard deviation, number of samples collected, and the number of values less than the detection limit for each of the three cruises. These concentrations correspond well with the data reported by Nriagu *et al.* (12). In Lake Ontario waters the latter reported average concentrations of 1.5 μg/L for Cu, 1.1 μg/L for Ni, 0.05 μg/L for Cd and 0.31 μg/L for Pb.

Waller and Lee (13) summarized trace metal data, which had been collected during 1972–73 on Lake Ontario, relative to the objectives established in the Great Lakes Water Quality Agreement. They reported multiple incidents of noncompliance with objectives. Of 207 Lake Ontario stations sampled, they found that 39% had Fe concentrations in excess of 300 μg/L; 44% exceeded the 25 μg/L

Table 2. Cruise Summaries. Mean and Standard Deviation, Number of Samples Collected and Number of Samples Less than the Detection Limit for the Metals Measured on Lake Ontario, 1979

| Metal | Detection limit (μg/L) | Maximum value (μg/L) | Cruise mean and standard deviation (μg/L)* | | | | | | | | | | | | |
| | | | Cruise 1: April 30–May 4 | | | | Cruise 2: May 28–June 1 | | | | Cruise 3: August 27–31 | | | |
			X	S.D.	N	n	X	S.D.	N	n	X	S.D.	N	n
Ag	0.1	0.8	—		0	0	0.10 ±	0.11	178	121	0.06 ±	0.02	170	159
Al	1.0	260.0	13.5 ±	15.4	180	2	13.0 ±	21.2	178	0	8.3 ±	8.5	166	12
As	0.1	1.2	0.60 ±	0.21	180	1	0.57 ±	0.28	178	0	0.72 ±	0.17	167	0
Cd	0.1	0.3	0.05 ±	0.02	180	174	0.05 ±	0.02	178	177	0.06 ±	0.03	174	137
Cr	0.2	3.2	—		0	0	0.62 ±	0.40	178	10	0.58 ±	0.35	174	6
Cu	0.1	4.5	1.21 ±	0.36	180	0	1.42 ±	0.46	178	0	1.34 ±	0.36	174	0
Fe	1.0	520.0	34.6 ±	45.3	180	0	25.9 ±	43.4	178	0	17.2 ±	20.0	174	1
Hg	0.05	0.16	0.026 ±	0.006	180	174	0.027 ±	0.012	178	173	0.025 ±	0.002	170	169
Mn	0.1	17.0	1.84 ±	1.01	180	0	2.00 ±	1.61	178	0	1.79 ±	1.52	174	0
Ni	0.1	11.0	1.16 ±	0.26	180	0	1.86 ±	0.92	178	0	1.47 ±	0.88	174	0
Pb	0.5	3.0	0.31 ±	0.26	180	161	0.40 ±	0.19	178	84	0.39 ±	0.25	174	109
Se	0.1	0.6	0.06 ±	0.06	180	162	0.07 ±	0.04	178	150	0.12 ±	0.04	167	2
Zn	1.0	13.0	1.1 ±	0.6	180	2	1.3 ±	0.8	178	14	1.2 ±	1.2	174	52

*A value of one-half the detection limit was used for values reported as "less than detection limit".
N = Total number of samples analyzed.
n = Number of samples for which "less than detection limit" was reported.

Ni objective; 79% exceeded the 30 μg/L Zn objective; and 99% exceeded the 5 μg/L Cu objective. All of the 68 stations sampled for Cd had concentrations at least five times greater than the 0.2 μg/L objective. These values are at least an order of magnitude greater than the data obtained in the present study, in which only five incidents of objective noncompliance were detected. In the Toronto-Hamilton region, 0.3 μg Cd/L was reported at station 1 (spring cruise) and at station 6 (August cruise). The heavy industry in this area may account for the high concentrations, as cadmium is an impurity associated with zinc and lead ores (14). Station 21 off the Niagara River, station 57 (Rochester) and the Black River Bay station 98 all reported total iron values near or exceeding the 300 μg/L objective.

When surface data were studied, it was evident that trace metal values were generally greater at the western end of the lake. Also, a concentration gradient existed wherein nearshore data were higher than those of the open waters. The total suspended material of nearshore waters was found by Nriagu *et al.* (12) to be higher than that of the open waters. The acid digestion method used to measure total metal concentrations leaches trace metals off the particulate matter; thus, it was expected that nearshore stations would report higher total trace metal levels. Especially high levels were recurrently found localized in the Toronto-Hamilton area and near the mouths of the Niagara, Genesee and Black rivers. It was thus decided to define this spatial variation further by utilizing the regionalization procedure developed by El-Shaarawi and Shah (9).

Table 3 presents the temporal (F_1) and spatial (F_2) values for each metal. It was generally found that the temporal variability far exceeded the spatial variability. Similar results were found by El-Shaarawi and Kwiatkowski (15) for a variety of chemical parameters in Lake Ontario. When a statistically significant F_2 value was found, the spatial distribution of that metal was described by division of the lake into statistically ($P \geqslant 0.20$) homogeneous zones.

Table 3. The Temporal (F_1) and Spatial (F_2) Variabilities for Each of the Trace Inorganics Studied on the Three Cruises on Lake Ontario, 1979

Metal	F_1	F_2
Ag	7.75*	0.99
Al	18.57*	3.28*
As	2.73	2.33*
Cd	3.99†	1.25
Cr	0.14	1.49
Cu	4.03†	1.74*
Fe	38.34*	3.47
Hg	0.51	0.90
Mn	8.01*	3.99*
Ni	21.09*	2.51*
Pb	7.83*	1.43†
Se	82.36*	1.00
Zn	2.49	1.51†

*Statistically different at 1% level.
†Statistically different at 5% level.

Figures 2 through 9 are the zone maps of Lake Ontario for the measured metals. Table 4 lists the mean and standard deviation, the number of samples collected in each zone, and the number of samples reported at less than the detection limit for each zone of the individual metal maps.

The zone map for aluminum (Fig. 2) reveals high aluminum concentrations along the northwestern and southern shores of Lake Ontario, and at Black Bay. The shoreline of the lake at the former two sites is shale, which is composed mainly of aluminosilicates; likewise, the drainage basin of the Black River (just east of the Bay) is shale (16). Hence, it appears that aluminum's zone 1 regions may be due to runoff.

The remaining metals have the highest concentrations at the mouths of rivers flowing into Lake Ontario, offshore of heavily populated areas. Industrial and domestic effluents and urban stormwater runoff would appear

Table 4. The Mean and Standard Deviation, Total Number of Surface Samples Collected in Each Zone (N) and the Number of Samples Reported at Less than Detection (n) for Each Metal Zone Map

Metal	Zone 1 X	S.D.	N	n	Zone 2 X	S.D.	N	n	Zone 3 X	S.D.	N	n	Zone 4 X	S.D.	N	n	Zone 5 X	S.D.	N	n
Al	28.8 ±	26.7	23	0	11.8 ±	8.5	39	0	4.8 ±	3.0	69	2								
As	0.81 ±	0.12	15	0	0.63 ±	0.18	82	0	0.39 ±	0.19	38	0								
Cu	2.33 ±	0.58	3	0	1.71 ±	0.26	12	0	1.47 ±	0.53	59	0	1.12 ±	0.25	62	0				
Fe	107.2 ±	111.8	9	0	43.7 ±	36.1	30	0	16.5 ±	8.6	26	0	9.6 ±	6.6	70	1	0.5 ±	0.0	1	1
Mn	4.53 ±	2.01	9	0	2.54 ±	0.95	30	0	1.53 ±	0.49	40	0	1.06 ±	0.41	57	0				
Ni	5.25 ±	4.08	6	0	1.92 ±	0.52	24	0	1.37 ±	0.40	100	0	0.92 ±	0.38	6	0				
Pb	0.75 ±	0.34	12	0	0.53 ±	0.33	18	6	0.31 ±	0.12	106	80								
Zn	4.4 ±	4.4	6	0	1.4 ±	0.6	44	2	0.9 ±	0.5	86	40								

NOTE: All values are reported in micrograms per litre.

Table 5. Zone Summaries. Mean Concentration of Each Metal for the Four Zones of the Composite Map

Metal	Zone 1				Zone 2				Zone 3				Zone 4			
	X	S.D.	N	n	X	S.D.	N	n	X	S.D.	N	n	X	S.D.	N	n
Ag	0.08 ±	0.06	16	11	0.08 ±	0.09	19	14	0.08 ±	0.08	41	35	0.07 ±	0.07	14	12
Al	24.3 ±	25.9	24	0	14.6 ±	13.5	29	0	6.1 ±	5.0	58	1	4.8 ±	3.5	20	1
As	0.54 ±	0.24	24	0	0.52 ±	0.23	29	0	0.62 ±	0.22	62	0	0.52 ±	0.19	20	0
Cd	0.07 ±	0.06	24	20	0.05 ±	0.01	30	28	0.05 ±	0.01	62	58	0.05 ±	0.00	20	20
Cr	1.16 ±	0.92	16	1	0.53 ±	0.31	20	3	0.44 ±	0.23	41	4	0.34 ±	0.18	14	2
Cu	1.71 ±	0.76	24	0	1.27 ±	0.37	30	0	1.31 ±	0.32	62	0	1.10 ±	0.31	20	0
Fe	61.2 ±	77.6	24	0	32.4 ±	35.4	30	1	12.3 ±	12.2	62	0	9.3 ±	8.0	20	1
Hg	0.026 ±	0.005	24	23	0.025 ±	0.000	29	29	0.025 ±	0.003	62	61	0.025 ±	0.000	20	20
Mn	2.90 ±	1.60	24	0	2.25 ±	1.36	30	0	1.32 ±	0.50	62	0	0.97 ±	0.46	20	0
Ni	2.81 ±	2.52	24	0	1.58 ±	0.51	30	0	1.35 ±	0.39	62	0	1.23 ±	0.38	20	0
Pb	0.59 ±	0.37	24	7	0.36 ±	0.17	30	19	0.33 ±	0.14	62	44	0.34 ±	0.19	20	16
Se	0.08 ±	0.05	24	13	0.08 ±	0.05	29	14	0.07 ±	0.03	62	36	0.08 ±	0.05	20	12
Zn	2.2 ±	2.5	24	5	1.3 ±	0.4	30	2	0.9 ±	0.5	62	24	0.8 ±	0.3	20	11

N = Total number of samples analyzed.

n = Number of samples reported at "less than detection limit".

NOTE: Data are reported in micrograms per litre and include only the surface (1 m) values for the three cruises. All values reported as "less than the detection limit" were taken at half the detectable level.

to be responsible for these elevated levels. Fitchko and Hutchinson (17) studied sediment trace metal levels at the mouths of several watercourses flowing into Lake Ontario. The data were reported relative to the maxima (defined as the mean plus two standard deviations) of the tributary outlets sampled. They reported "elevated" Pb levels (greater than the maxima) in surface sediment samples collected at the mouths of the Credit and Humber rivers and the Etobicoke Creek, and "excessive" concentrations (greater than five times the maxima) at the mouth of the Don River. According to Nriagu *et al.* (12), about 60% to 90% of the runoff load of metals into Lake Ontario is imported via the Niagara River from Lake Erie; aluminum, iron and manganese concentrations were highest just at the mouth of this river (Figs. 2, 7, 9). The lead, zinc and copper concentrations off Hamilton (Figs. 4, 5, 6) most likely are a result of the steelworks foundries located there (18).

The composite trace metal map of Lake Ontario is shown in Figure 10. Table 5 lists the mean concentration and standard deviation for all the metals in each zone of the map. For the most part, concentrations decreased from the highest in zone 1 to the lowest in zone 4. However, there were exceptions; silver, cadmium, mercury, lead and selenium especially did not follow this zone-concentration trend. This is due to the high percentage of less-than-detection values reported for these metals. Greater sensitivity of detection is required for their more accurate zonation. The isolated zone 2 in the eastern region of the lake was also reported by Chau *et al.* (3) in describing the horizontal distribution of surface concentrations of iron and copper in Lake Ontario.

Because more than half the metals showed significant temporal variability (Table 3), the vertical distribution was examined on a cruise-by-cruise basis. For each station, depth-weighted concentrations were calculated for both the surface-to-20-metre stratum and the entire water column (surface-to-bottom). Student's t test values were calculated to compare the two strata; the results are shown in Table 6.

Table 6. Student's t Test Values for the 1979 Lake Ontario Trace Metal Concentrations of the Surface-to-20-Metre Stratum Versus the Entire Water Column

Metal	Cruise 1	Cruise 2	Cruise 3
Ag	—	0.00	0.00
Al	0.39	0.32	1.34
As	0.05	0.64	3.50*
Cd	0.00	0.00	0.00
Cr	—	0.74	1.79†
Cu	−0.20	−0.23	0.38
Fe	0.37	0.36	1.95†
Hg	0.23	1.01	0.00
Mn	0.47	0.44	1.46
Ni	−0.19	0.15	−0.52
Pb	−0.24	0.14	−0.18
Se	1.19	−1.32	0.84
Zn	0.29	−0.46	1.23

*Significant at 5% level.

†Significant at 10% level.

On the first two cruises, the difference between the trace metal levels reported for the surface waters and those reported throughout the water column was not found to be statistically significant for any of the metals. However,

on cruise 3, which was conducted during stratified conditions in late summer, arsenic, chromium and iron concentrations were significantly lower in the surface waters (0-20 m). Furthermore, of the metals that showed any difference in concentration between the two strata, the surface trace metal levels tended to be less than the whole water column for all the elements except nickel and lead (for these the mean concentrations were, respectively, 0.04 and 0.02 μg/L higher in the surface waters).

CONCLUSIONS

Improvement in extraction techniques and sensitivity of detection is required so that fewer values of less than detection limit are reported. This would permit more extensive statistical analysis of the data and zonation of the lake for all the metals studied. Furthermore, for more definitive zonation, more stations are required in both the central eastern and the western basins.

Having determined that higher total metal concentrations are found in areas directly influenced by runoff, major watercourses entering the lake and urban-industrial activities, it is evident that these zones are greatly affected by the concentration of suspended sediments. However, with consideration to its original intent, the composite map is still a valuable tool for defining potential problem areas and regions with a greater probability for demonstrating noncompliance with the Water Quality Agreement objectives.

The temporal variability, although much greater than the spatial variability, showed the epilimnetic waters to be significantly different from the whole water column only during the late summer cruise. The vertical distribution of trace metals in the spring and early summer indicated that surface waters were representative of the entire water column.

ACKNOWLEDGMENTS

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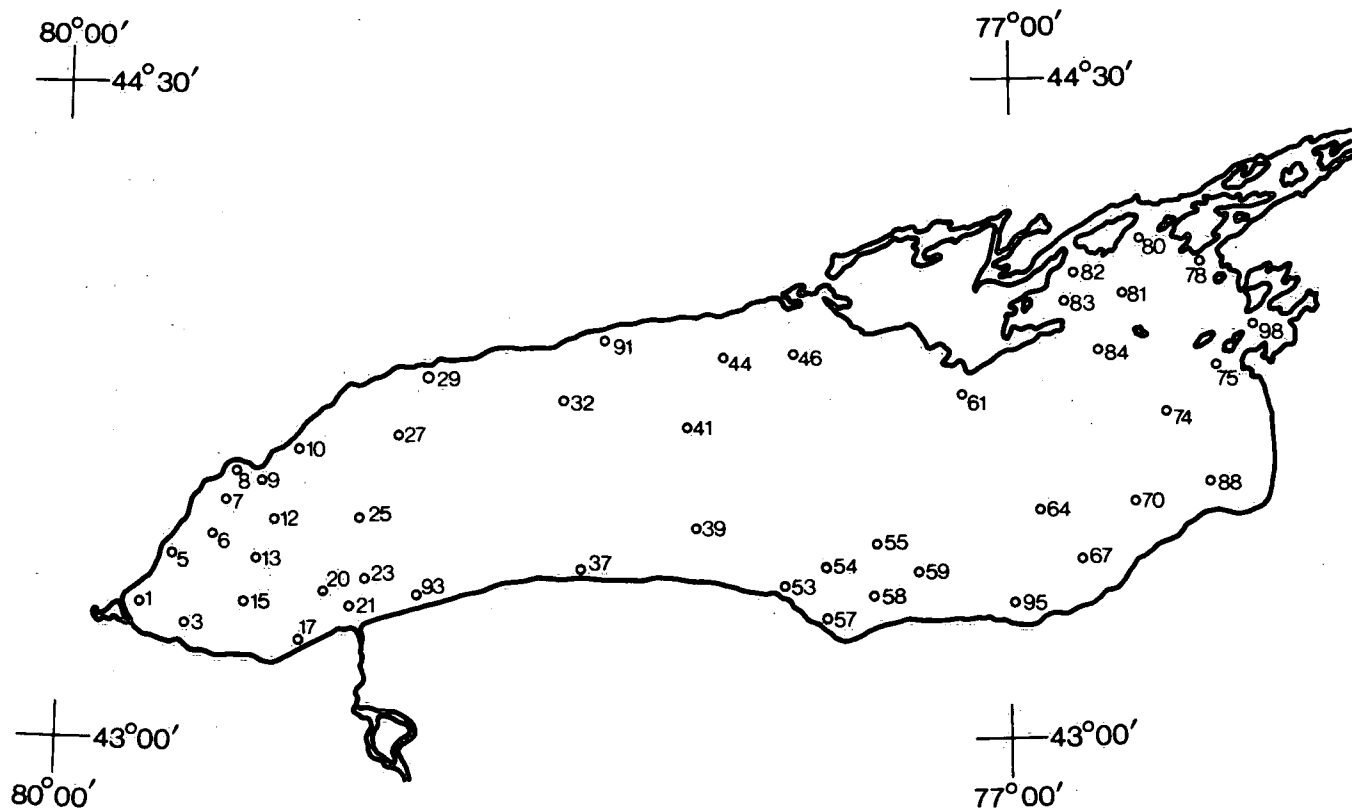
Illustrations

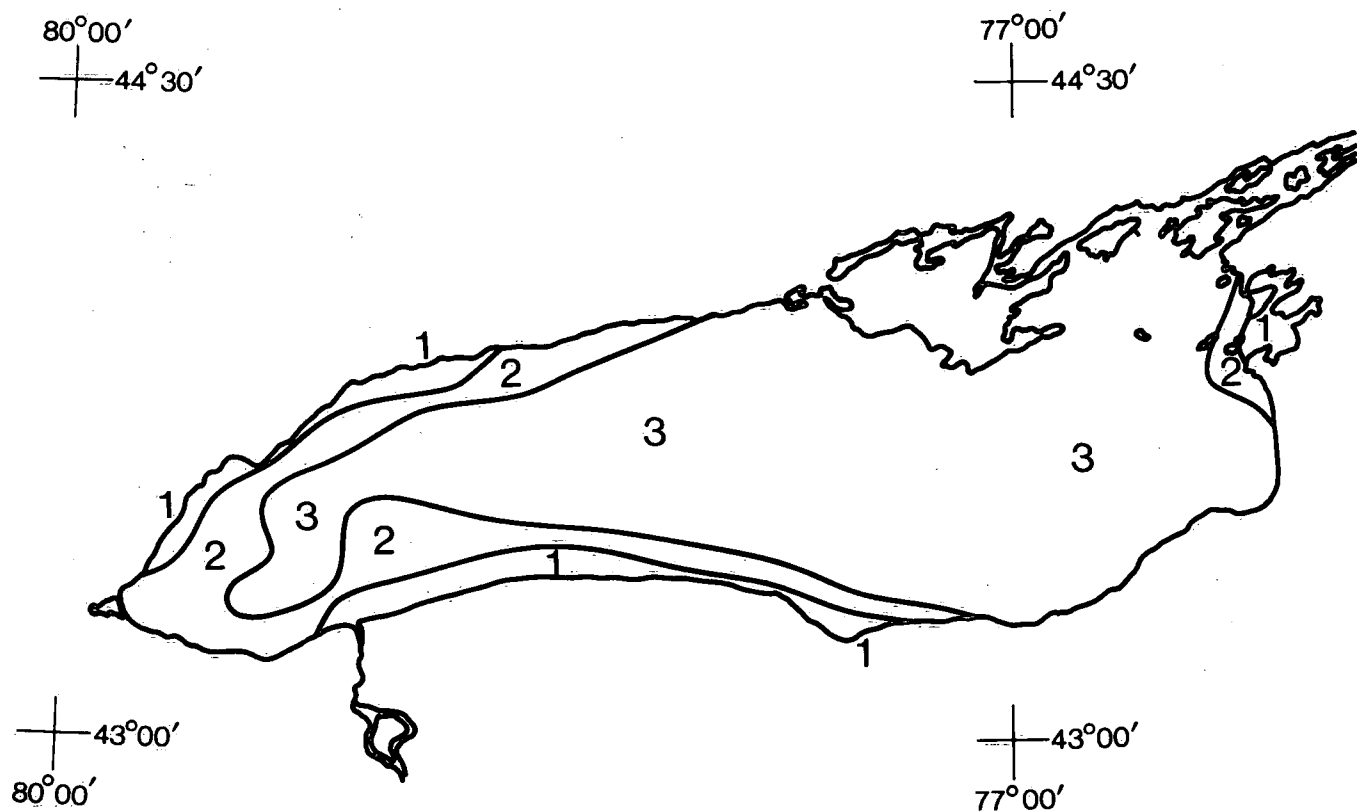Figure 1. Location of 1979 trace metal sampling stations in Lake Ontario.
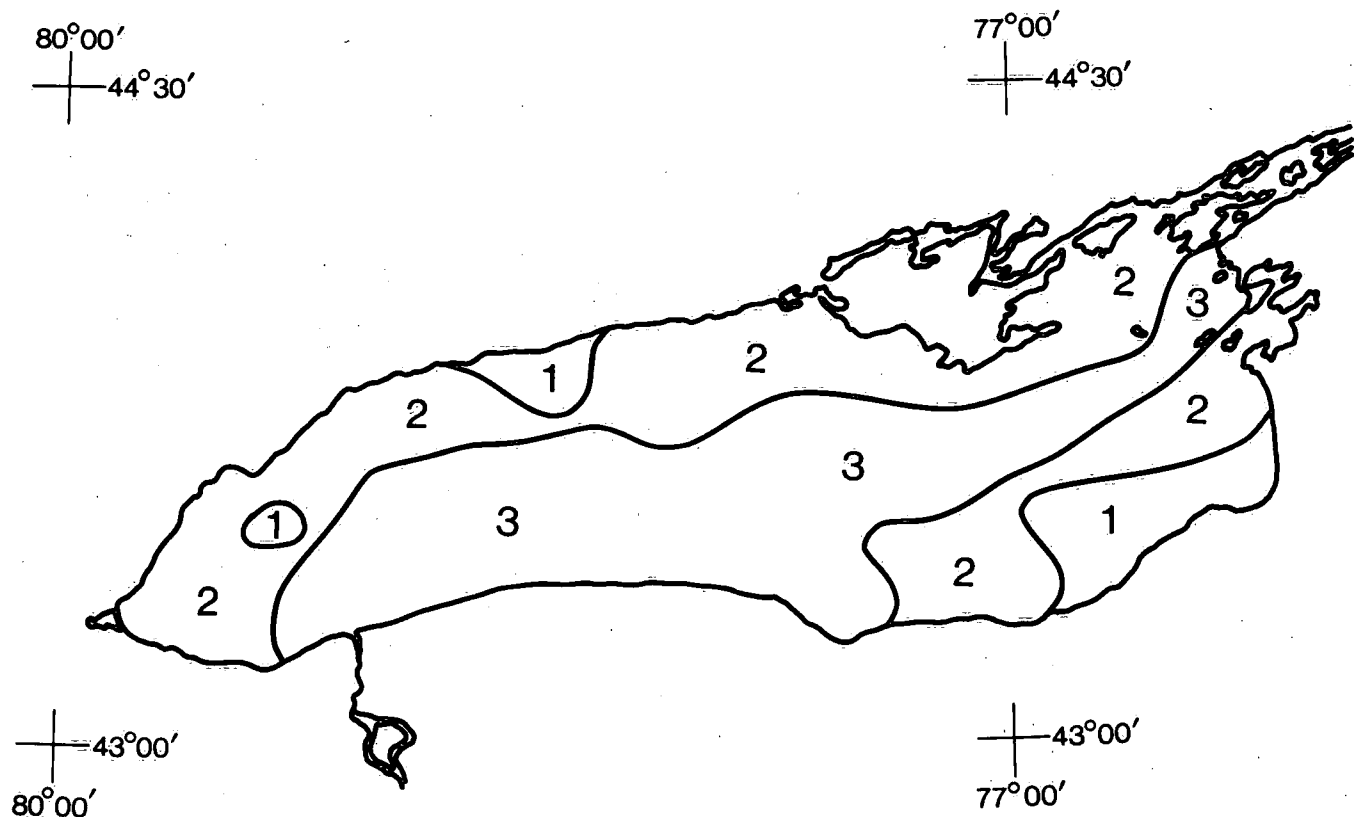

Figure 2. Zone map of aluminum, 1979.

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Figure 3. Zone map of arsenic, 1979.

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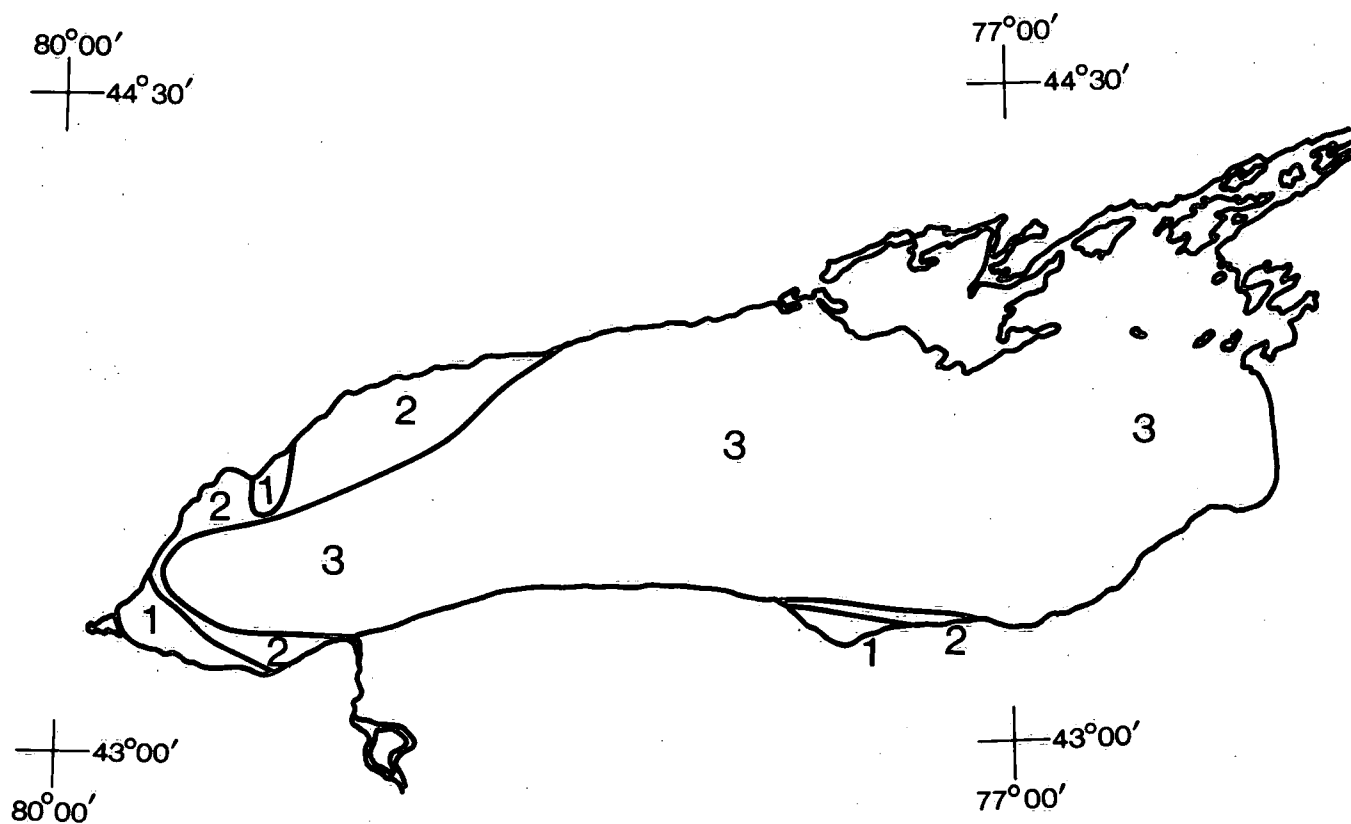
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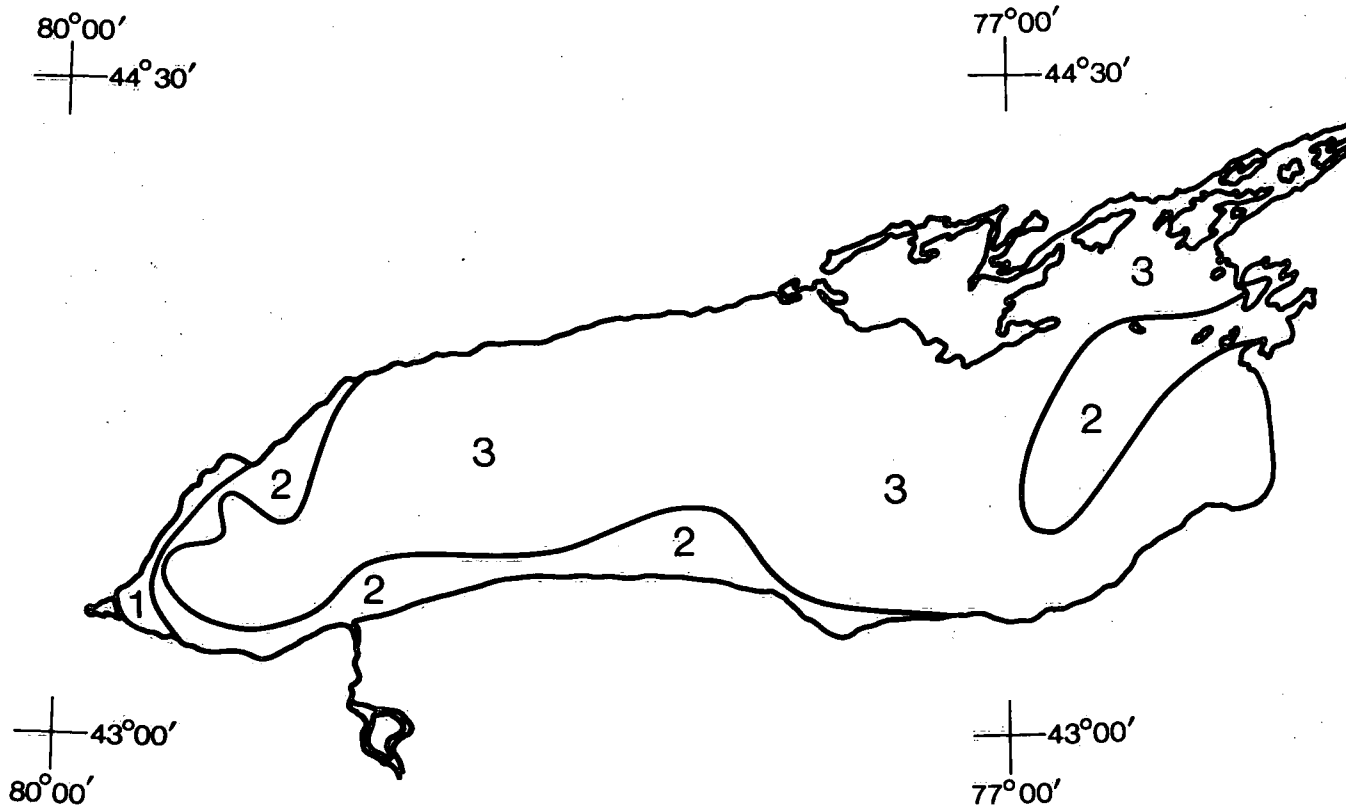
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Figure 4. Zone map of lead, 1979.
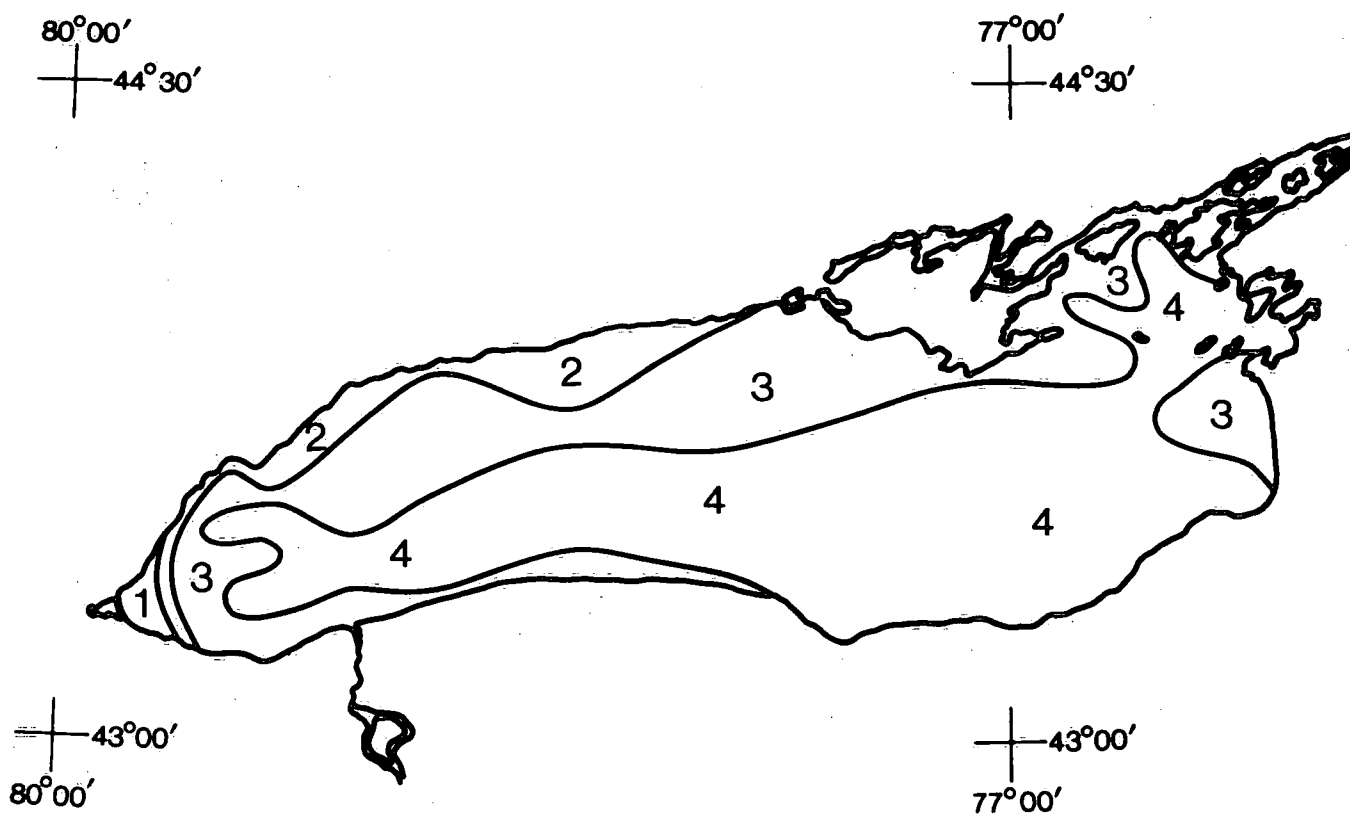
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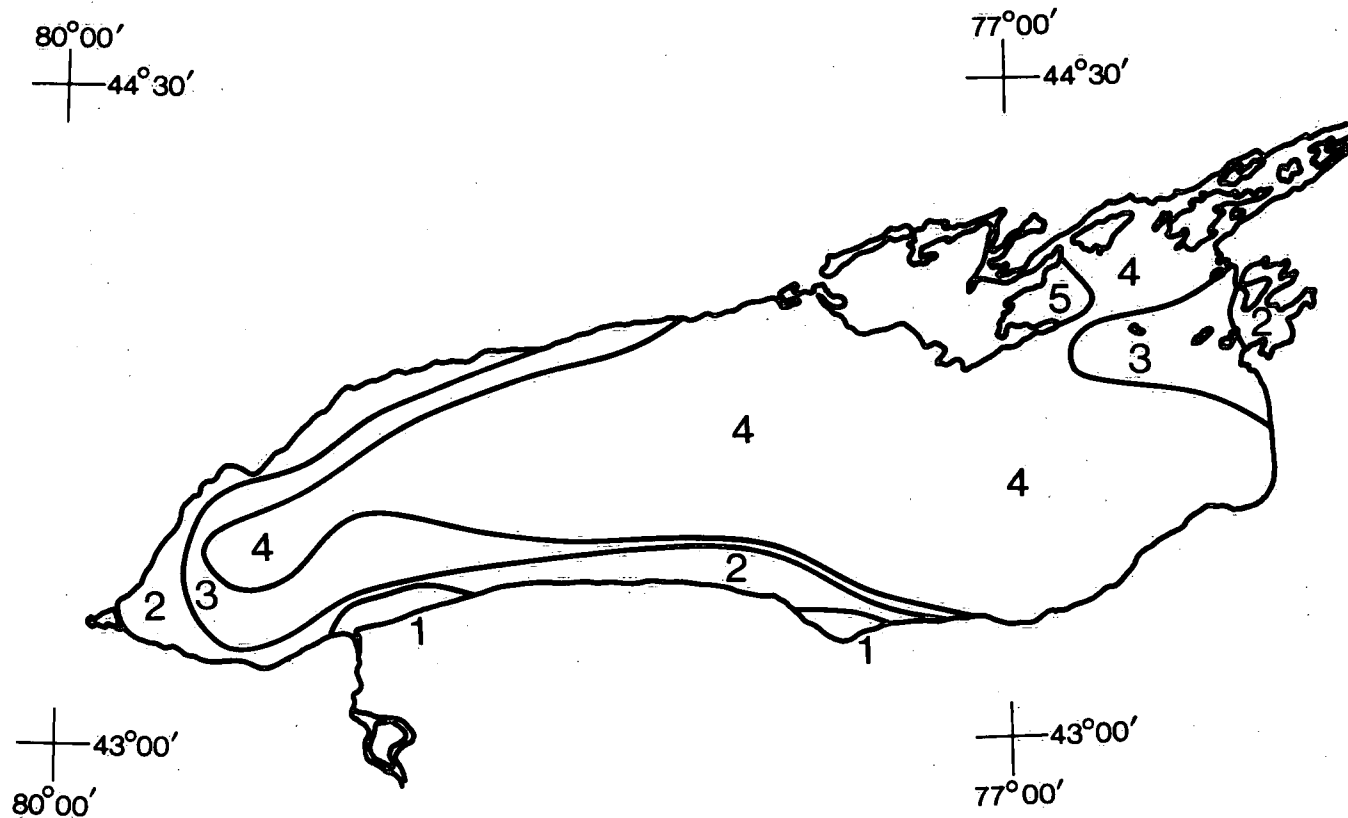

Figure 6. Zone map of copper, 1979.
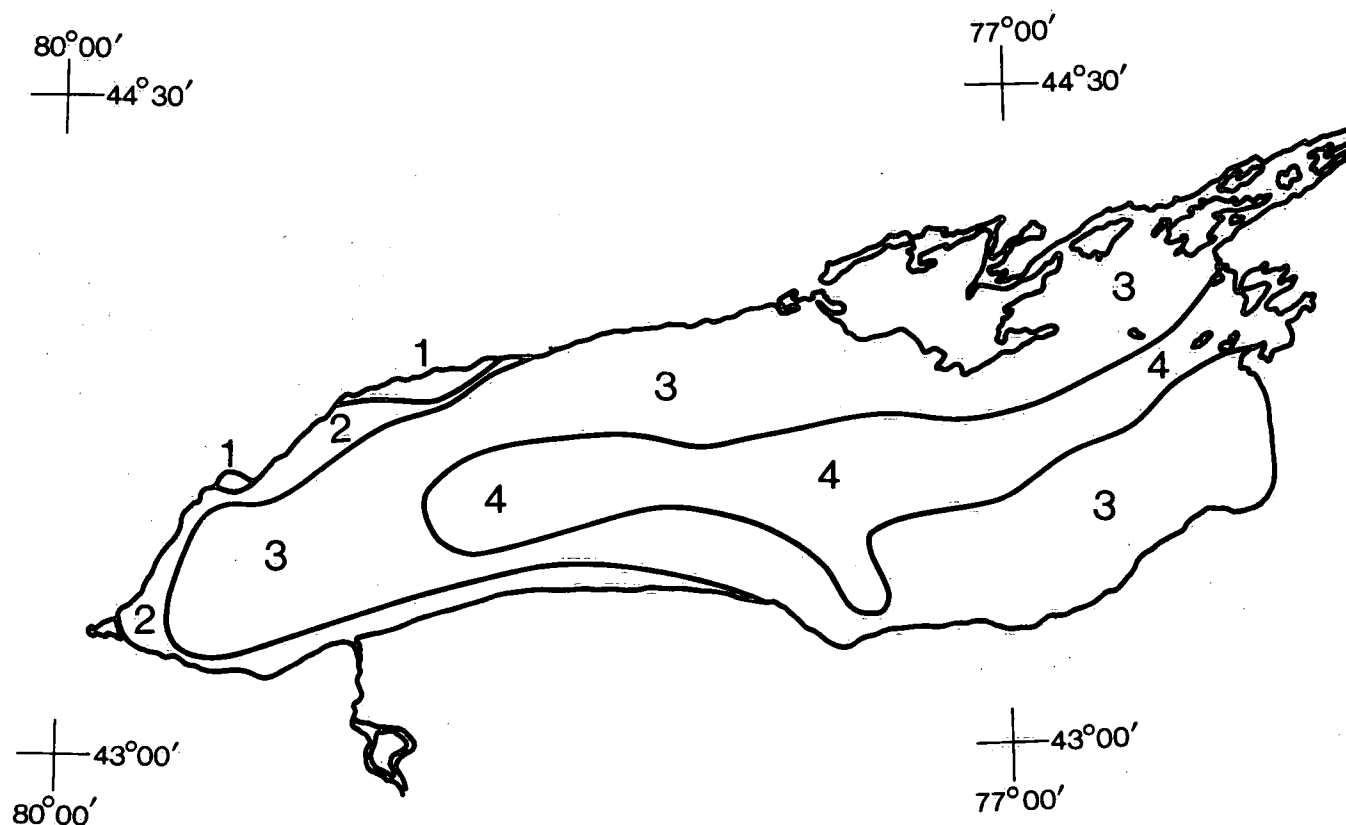
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Figure 7. Zone map of iron, 1979.
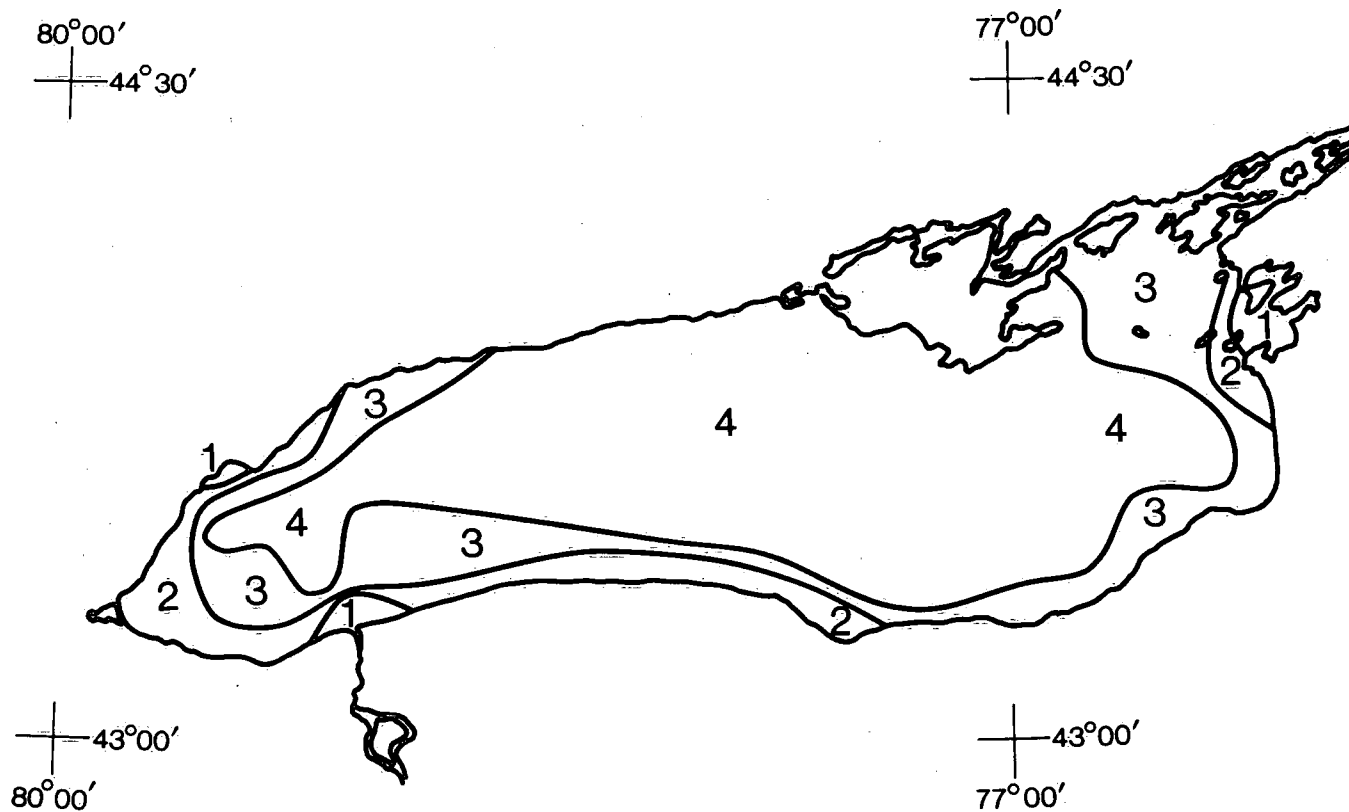

Figure 8. Zone map of nickel, 1979.

Figure 9. Zone map of manganese, 1979.
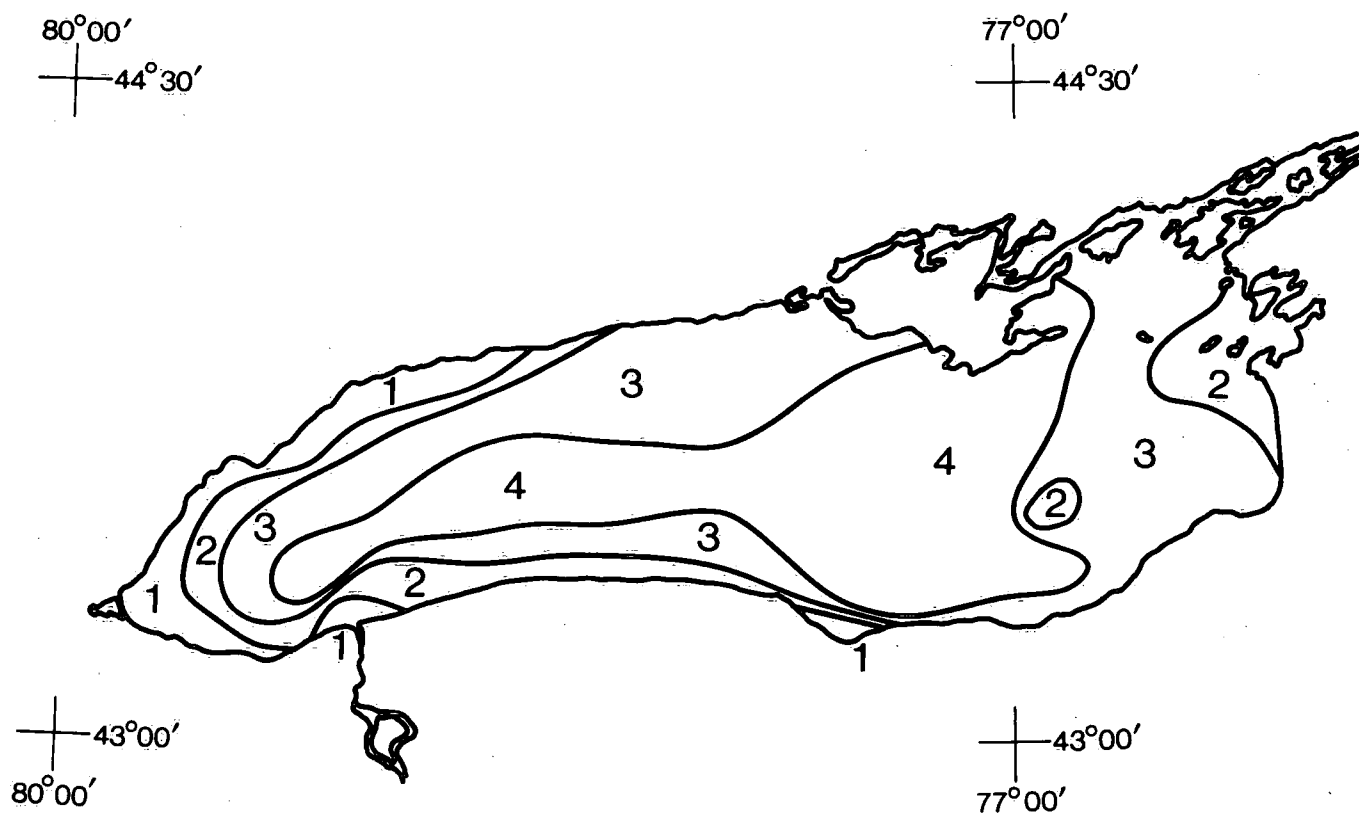
Figure 10. Composite map of trace metals in Lake Ontario, 1979.

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