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MONITORING SHALLOW GROUND WATER FOR INJECTED LIQUID INDUSTRIAL WASTES. SARNIA, CANADA

by

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ABSTRACT

The objective of this study was to assess the possible impact, of deep well disposal operations conducted between 1958 and 1974, on the ground-water quality in a shallow "fresh water aquifer", beneath Sarnia, Ontario, Canada. Because of the breakout of formation fluids in Sarnia and Port Huron, Michigan in the early 1970's it had been hypothesized that liquid waste from the disposal zone in bedrock had leaked through the numerous abandoned oil, gas and salt wells in the area up to the shallow aquifer and thence to the surface.

A monitoring well network of 29 2" piezometers was established in this thin (<10ft) sand and shale aquifer system, which exists between 100 and 150 ft. below ground surface. In addition, a 980 ft. deep borehole was drilled and instrumented with a Westbay multilevel casing which permitted sampling of the disposal zone.

Samples from the monitoring wells and the Westbay multilevel casing were analysed for volatiles by GC/MS. Those volatile aromatics that were conspicuously present in the deep disposal zone, e.g., ethyl toluenes and trimethyl benzene, were not detected in the shallow monitoring wells. Thus, if contaminants from the disposal zone did indeed migrate to the shallow aquifer, contamination was not widespread and probably consisted mostly of displaced chloride-rich formation waters.

RÉSUMÉ

L'objectif de cette étude était d'évaluer l'impact possible des injections de déchet pétrochimiques dans le sous-bassement rocheux effectuées entre 1958 et 1974, sur la qualité des eaux souterraines de l'aquifère d'eau douce dans la région de Sarnia en Ontario. A cause de l'éruption de liquides de formation à Sarnia et à Port Huron au Michigan au début des années 70, on avait émis l'hypothèse que les déchets liquides des zones d'injection du sous-bassement rocheux s'étaient échappés par les nombreux puits abandonnés de gaz, huile et sel de la région vers l'aquifère supérieur et de là jusqu'à la surface.

Un réseau de 29 piezomètres de 2" a été installé dans ce mince (<10 pi.) aquifère de sable et de schiste situé entre 100 et 150 pi. sous terre. En plus, un puits profond de 980 pi. a été foré et instrumenté d'un tubage Westbay à niveaux multiples qui a permis l'échantillonage de la zone d'injection des déchets.

Les échantillons du réseau piézométrique et du forage Westbay ont été analysés pour les composés organiques volatils par CG-SM. Les aromatiques volatils qui étaient les plus remarquables dans la zone d'injection e.g. les toluènes d'éthyle et le triméthylbenzène ne furent pas décelés dans les puits de l'aquifère supérieur. Alors, s'il y a eu migration des contaminants vers l'aquifère supérieur, la contamination est demeurée localisée et consistait surtout d'eau de formation riche en chlorure, déplacée par la pression des déchets injectés.

MANAGEMENT PERSPECTIVE

The objective of the study was to assess the possible impact of deep well disposal operations conducted between 1958 and 1974 on the groundwater quality of the fresh-water aquifer near Sarnia, Ontario. This study shows that there is currently no evidence of contamination of the fresh-water aquifer. One of the problems is that the petrochemical wastes contain compounds that are generally found in petroleum bearing geologic formation and this it is difficult to differentiate between the two. It is therefore recommended that monitoring be continued.

PERSPECTIVE POUR LA GESTION

Cette étude avait pour objectif d'évaluer l'impact des injections de déchet pétrochimiques dans le sous-bassement rocheux effectuées entre 1958 et 1974, sur la qualité des eaux souterraines de l'aquifère d'eau douce dans la région de Sarnia en Ontario. Cette étude a démontré qu'il n'y a dans le moment aucun impact quantifiable sur la qualité de l'eau de puits. Il est cependant difficile de différencier les contaminants provenants de déchets pétrochimiques des composés présents dans les dépots pétrolifères. Il est donc recommandé de continuer la surveillance des puits.

INTRODUCTION

From 1958 to 1974 approximately 1.7 billion US gallons (6.4 million m^3) of liquid industrial waste from oil refining and petrochemical production facilities were injected under pressure into bedrock beneath Sarnia, Ontario, Canada. The wastewaters contained high levels of phenols, sulfides and ammonia and had a pH > 9. The disposal zone was the dolomite of the Lucas formation of Devonian age, situated between 580 and 900 feet below ground surface. Wellhead injected pressures of up to 450 psi were used to obtain injection rates of 50 to 100 gpm(McLean, 1968; URM, 1984). The location of these wells is shown in Figure 1.

During the early 1970's there were reports of discharge at the land surface of formation fluids, such as oil and gas, from abandoned wells in Port Huron, Michigan (van Everdingen and Freeze, 1971, p.30) and of what appeared to be industrial wastewater from an abandoned water well in downtown Sarnia. Subsequently, pressured injection was abandoned in favour of gravity disposal limited to areas five miles away from the St Clair River, the international boundary. It was concluded at the time that the effluent observed in the Sarnia well, if it was indeed industrial wastewater, must have migrated to the surface via an improperly abandoned oil, gas or salt borehole to the freshwater aquifer (FWA) and thence upwards via the abandoned water well (R.J. Patterson, formerly Ontario Ministry of the Environment, personal communication). In this context it is worth noting that van Everdingen and Freeze (1971) reported that "there may be as many as 30,000 unplugged wells.... near Sarnia." Various scenarios for the

upward migration of liquid industrial wastes are shown in Figure 2.

In 1986, in order to confirm that the injected liquid wastewaters were not escaping to the St. Clair River via the freshwater aquifer, it was decided by the Ontario and Federal Environment departments to drill a deep borehole to the depth of the disposal formation. This borehole was instrumented with a Westbay multilevel casing system for monitoring fluid pressures and collecting water samples for chemical analysis (see Raven et al., 1990). In addition, fifteen new monitoring wells were added to the FWA monitoring well network for a total of 29 (see Figure 1 for their locations).

Thus it became possible to monitor hydraulic heads in the disposal formation, the freshwater aquifer and the river and therefore determine the present potential for upward leakage of the industrial wastewaters. Furthermore, chemical testing of ground waters in the disposal formation through ports in the Westbay multilevel casing permitted the identification of fingerprint chemicals in that formation.

The purpose of this paper is to describe the organic ground water quality in the deep disposal formation and compare it to that of the shallow freshwater aquifer in order to determine if the latter is contaminated by industrial wastewaters previously injected into the former disposal zone.

METHODS

Monitoring Well Design

The 29 2-inch PVC piezometers/monitoring wells of the freshwater aquifer network were installed during three intervals from 1985-1987. These wells were used for ground water sampling, head measurements and hydraulic testing (i.e. slug tests). They were installed by mud-rotary drilling methods. Each well was completed with a 10 ft. (3 m) screen in the freshwater aquifer and PVC casing to the surface. Well depths below ground surface varied from 115 ft. (35 m) to 230 ft. (70 m). Following installation of the screen and casing, a sand pack was placed around the screen with a bentonite seal above it. The remainder of the hole was cemented to the ground surface and completed with a protective steel surface casing. Three of these wells were constructed on industrial property and blowout preventers were used during drilling operations in case of natural gas escape from the borehole.

Each well was developed with a hydraulic jetting tool until the return water was clean. About 10 well volumes were air lifted from the casing prior to any hydraulic or chemical testing. Water used for drilling and well development was tagged with iodide and Lissamine and subsequently monitored to ensure that samples being obtained were not contaminated. Problems with natural gas production were encountered at several of these wells and consequently they could not be used for sampling.

Field Methods

The monitoring wells in the fresh water aquifer were all instrumented with dedicated bladder pumps [Well Wizard (QED Systems, Ann Arbor, MI) or Geoguard (American Sigma, Middleport NY)] for ground water sampling. The Well Wizard pumps were equipped with inflatable packers to seal the screened section from the rest of the casing and therefore minimize the volume of water to be purged prior to sampling. The Geoguard pumps were all two stage and standing water could be rapidly removed in the air-lift mode prior to switching over to the bladder pump for sampling. The drive gas for these pumps was either filtered compressor air or prepurified nitrogen from a cylinder.

At least two well volumes of standing water were removed at high flow rates prior to field sampling or analysis. The pumping rates were then lowered to about 300 - 500 mL/min in order for flow cell analyses of pH and E_H to be conducted at the well-head. After stable readings were obtained and recorded, samples for volatiles (BTEX), major ions, phenols and isotopes were taken. BTEX samples were acidified with HCl and phenol samples were preserved with CuSO4 and refrigerated at 4°C until analysis. Trip blanks were employed in order to detect possible contamination of samples from other sources (e.g. BTEX from gasoline).

GC-MS Analytical Method

Because of the expected high level of contamination in the Westbay multilevel aliquots of one to five mL were used for analyses and diluted to 10 mLs with uncontaminated groundwater. The analysis was conducted on a Unacon-810 purge and trap concentrator directly interfaced to a Hewlett-Packard model 5970 GC-MSD. The analytical column was a J&W DB-624 fused silica capillary column, 30 m in length, 0.32 mm i.d., 1 um film thickness. The GC was cooled to -15°C with CO_2 and ramped to 130°c at 10°/min. For the Westbay multilevel samples, the mass spectral acquisition was done in the scan mode, from 50 to 250 a.m.u.. Chlorobenzene-d5 and difluorobenzene were used as internal standards. To obtain maximum sensitivity for the samples from the freshwater aquifer, selected ion-monitoring was used for the target analytes. Quantitation was then done by the external standard method. Standards solutions were prepared from the purest available chemicals.

HYDROGEOLOGY

Fresh Water Aquifer

The fresh water aquifer underlies much of Lambton County in Southwestern Ontario. It's extent is defined by the subcrop area of the Upper Devonian Kettle Point Formation, and consists of the upper 3-6 ft. of fractured and weathered shales and up to 6 feet of

overlying thin discontinuous layers of sand and gravel (Vandenberg et al., 1977; Intera, 1989). The fresh water aquifer is patchy and is confined directly below by unweathered Kettle Point shales and above by approximately 100 - 150 ft. of clay till, the uppermost portion of which has been reworked by the action of glacial lakes (Chapman and Putman 1984). Regionally, recharge to the aquifer occurs at the updip edge of the aquifer in eastern Lambton Country and from seepage from overlying tills. Ground water flows westward and northwestward toward the St. Clair River and Lake Huron (Vandenburg et al., 1977; Intera, 1988) as shown in Figure 3. In eastern Lambton County, the fresh water aquifer is a source of water supply for private wells, however, in the Sarnia area the aquifer is not utilized since residents obtain their drinking water from the St. Clair River.

A bedrock valley has been identified from drilling results and borehole geophysics running parallel to and 1000-3300 ft. (300-1000 m) east of the St. Clair River. It extends from Sarnia to south of Corunna where it has been identified on the Michigan side of the St. Clair River (Edsal et al., 1988). The bedrock valley has eroded 100 ft below the top of the Kettle Point Formation, which in the Sarnia area is about 50 ft. thick, and into the top of the middle Devonian Hamilton Group limestone and shales. The bedrock valley is filled with a complex sequence of clays, silts and thin sands and gravels, and forms a continuation of the fresh water aquifer. A cross section through the bedrock valley/aquifer complex is shown in Figure 4.

A variety of bail, withdrawal and recovery tests, along with a 12 hour pump test performed within the bedrock valley portion of the fresh water aquifer, have been conducted to estimate hydraulic conductivity, and are described in a number of publications. (MOE, 1986; Intera, 1987; Intera, 1989). A summary of hydraulic conductivity testing is given in Table X.

Hydraulic conductivity within the fresh water aquifer ranges from 10^{-3} - 10^{-7} m/s. Within the bedrock valley portion of the aquifer, the geometric mean of hydraulic conductivity is 1 x 10^{-4} m/s. Outside the bedrock valley, the geometric mean of hydraulic conductivity is 5 X 10^{-6} m/s. (Intera, 1989).

The potentiometric surface for the fresh water aquifer is shown in Figure 3. Horizontal groundwater flow directions in the Sarnia area are generally from the southeast, in a northwesterly and westerly direction toward the buried valley and St. Clair River. South of Sarnia, horizontal ground water flow is westward toward the buried valley. Outside of the buried valley, horizontal groundwater velocities average 2.6 ft/y or 0.8 m/a (Intera, 1989). Between the buried valley and the St. Clair River, the potentiometric surface indicates the presence of a ground water divide from which groundwater flows toward both the St. Clair River and the buried valley (see Figure 3).

Outside the bedrock valley, the potential exists for vertical ground water flow from the fresh water aquifer downward to the lower units of the Kettle Point Shale. Within the valley, groundwater movement is less clear, with some components of flow showing a downward direction to deeper formations and to the northwest and some

components of flow showing an upward direction to the St. Clair River; however, it is reasonable to conclude that all ground water flow in the bedrock valley must eventually discharge to the St. Clair River, the regional hydraulic sink.

Bedrock

The disposal zone chosen to receive the liquid industrial wastewaters was the Lucas formation of the Detroit River group of Devonian age. In the Sarnia area this formation is 260 to 290 feet thick (McLean, 1968) and consists of microcrystalline dolomites with halite and anhydrite layers (Winder and Sanford, 1972). Most of the industrial waste is thought to have been injected into high porosity and permeability "lost circulation" zones in the upper part of the Lucas formation that were created by anhydrite dissolution (URM, 1984). The Lucas Fm. is overlain by flat lying or gently dipping Devonian limestones and shales some of which contain petroleum.

GROUNDWATER QUALITY

Inorganic Parameters

Field testing of ground waters from the fresh water aquifer indicated a temperature of approximately 10°C, a mean pH of 7.8 (standard deviation = 0.4) and a mean platinum electrode potential or E_H of 100 mV (standard deviation = 50 mV). Such low E_H values

suggest the likelihood of sulfate reduction occurring within the fresh water aquifer (Champ et al., 1979; Jackson and Patterson, 1982), a conclusion made more likely by the widespread occurrence of natural gas in the aquifer.

The major ion chemistry of the fresh water aquifer indicates that ground waters are typically of the Na/Cl type, with some being of the Na/HCO_3 type. The chloride distribution in the fresh water aquifer records the complex hydraulic history of the area. Chloride concentrations range from 200 to 1670 mg/L, while specific electrical conductance varies from 500 to 5000 uS/cm. The highest chloride concentrations are found near the St. Clair River, particularly in those wells just north of the CN tunnel, e.g., 1-85, 2-87, while concentrations decrease to the east. In the southern part of the study area, elevated chloride concentrations extend further eastward (6-85, 5-87, 8-87, 14-87). The zones of high chloride are believed to reflect chloride-rich formation waters displaced during the period of pressurized injection of liquid industrial waste or diffusion of chlorides from the underlying bedrock.

Volatile Aromatics

Benzene, toluene, ethylbenzene, and xylenes (BTEX) were found at all levels in the Westbay multilevel (Table 2), however there was a drastic change at 622 ft. (192 m) the depth at which the liquid industrial wastes were known to have been injected. The sample at that level in the borehole showed a very characteristic pattern of distribution (Figure 5) where ethyl toluenes and other aliphatic

aromatics were predominant. This pattern is used as a fingerprint of the liquid wastes. If the freshwater aquifer was contaminated by wastes leaking upwards from this zone, it would be expected that the same contaminants would be observed in them, unless biological degradation was occuring. Under the generally anoxic conditions such as those prevailing in the shallow aquifer, it has been found that certain isomers of xylene are degraded preferentially (Barker, et al., 1986).

Surveys in two consecutive years demonstrated the possible presence of very low levels of aromatic hydrocarbons (Tables 3 and 4). As is shown in Figure 6, the levels observed were not significantly higher than zero in any of the monitoring wells and the fingerprint characteristics of the liquid waste were not observed.

The basic plotting method utilized in Figures 5 and 6 is that of multivariate plots as described by Lesage and Lapcevic (in press). However, in the case where all the values are near zero, the data plotted would be concentrated near the origin and would be difficult to visualize. Therefore, in Figure 6, the value of one was added to each data point, effectively shifting the zero at the first tick mark. If all values were zero, a hexagon would be produced. The data, once corrected for instrument blank, produces figures that do not significantly deviate from zero for any of the analytes.

b) Phenols

The organic contaminant found in highest concentration is the disposal zone was phenol. It is very mobile (log K_{ow} 1.46) hence could be a very good indicator of waste migration. No phenols were detected in the freshwater aquifer network. It thus seems unlikely that upward migration of the wastes has caused widespread contamination of the freshwater aquifer.

3) Sources of BTEX Contamination in the Freshwater Aquifer:

The current data indicates that the freshwater aquifer is not presently contaminated by wastes. However, the monitoring wells will continue to be sampled periodically because a contamination threat to the aquifer remains. Unfortunately the presence of BTEX and phenol cannot be used as indisputable evidence of contamination by industrial wastes. The Sarnia area contains some petroleum bearing geological formations that can contribute to the dissolved BTEX and phenol in groundwater (Hunt, 1979) . Concentrations up to 50µg/L total BTEX have been measured in groundwater contacting petroleum bearing formations in the Niagara Falls bedrock (Lesage and Lapcevic, in press; Novakowski and Lapcevic, 1988; Zenon Environmental, 1985).

Only when a full suite of volatiles are analysed is it possible to compare the composition of the petroleum product with any degree of certainty. Unfortunately historical data are very scarce where all substituted benzenes are measured and individual data points reported.

In the upper part of the bedrock some petroleum bearing formations were encountered, which could easily be a source of BTEX for the freshwater aquifer. In those samples, benzene, toluene and the xylenes predominate, similarly to what was observed in the Niagara samples. This contrasts with the disposal area where ethyl toluenes are the most abundant aromatic hydrocarbons. This distribution may be altered as the contaminants migrate away from the source or if they underdo biodegradation.

CONCLUSIONS

A monitoring well network was installed in a shallow aquifer system to detect upward leakage of liquid industrial wastes injected into a deep bedrock formation. The disposal formation was characterized as containing significant quantities of fingerprint chemicals such as phenols, ethyl toluenes and trimethyl benzene. These same compounds were not present in the shallow aquifer system, indicating that widespread contamination did not occur or that biodegradation has lowered concentrations of the fingerprint chemicals to below detection limits. However, the injection operations did seem to displace chloride-rich formation waters into the shallow aquifer system.

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BIOGRAPHICAL SKETCHES

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LIST OF FIGURES

Table 1. Summary of Hydraulic Conductivity Testing of the Fresh Water Aquifer at Sarnia

Number of Wells	Method	Number of Tests	Range of Hydraulic Conductivity (m/s)	Geometric Mean Hydraulic Conductivity (m/s)	Geometric Mean Storativity	Geometric Mean Tranmissivity (m^2/s)
23	bail	80	$2 \times 10^{-4} - 4 \times 10^{-8}$	1×10^{-5}	-	2×10^{-5}
6	withdrawal	6	$2 \times 10^{-4} - 2 \times 10^{-5}$	4×10^{-5}	-	8×10^{-5}
1	recovery	1	1×10^{-9}	1×10^{-9}	-	2×10^{-9}
11	pump	2	$8 \times 10^{-3} - 1 \times 10^{-3}$	3×10^{-4}	2×10^{-4}	6×10^{-3}

Note: - assume aquifer thickness of 2m.

- sources: MOE, 1986; Intera Technologies, 1987, 1989

Table 2. Organic Chemicals Measured in µg/L in samples from the Westbay Multilevel collected on June 2, 1988

Sample Depth(a) ft (m)		Benzene	Toluene	Ethyl-benzene	Styrene	Xylenes o-	Xylenes m+p	Ethyl Toluenes (sum)	Trimethyl Benzene	Phenol[a]
200	(61)	231	214	51	7	124	230	39	73	2290
250	(75)	237	221	53	7	127	233	48	71	1240
405	(123)	269	227	47	6	112	213	38	59	2570
440	(133)	173	157	45	6	94	162	37	50	3080
635	(192)	133	168	133	33	169	205	573	151	30900
685	(207)	28	37	28	9	44	70	61	18	n.d.
752	(228)	48	19	5	1	20	27	7	11	
792	(240)	9	43	25	3	31	41	11	14	

[a] Data from Intera Technologies (1989)

Table 3. Organic chemicals measured in µg/L in samples from the freshwater aquifer monitoring wells collected on September 27, 1988

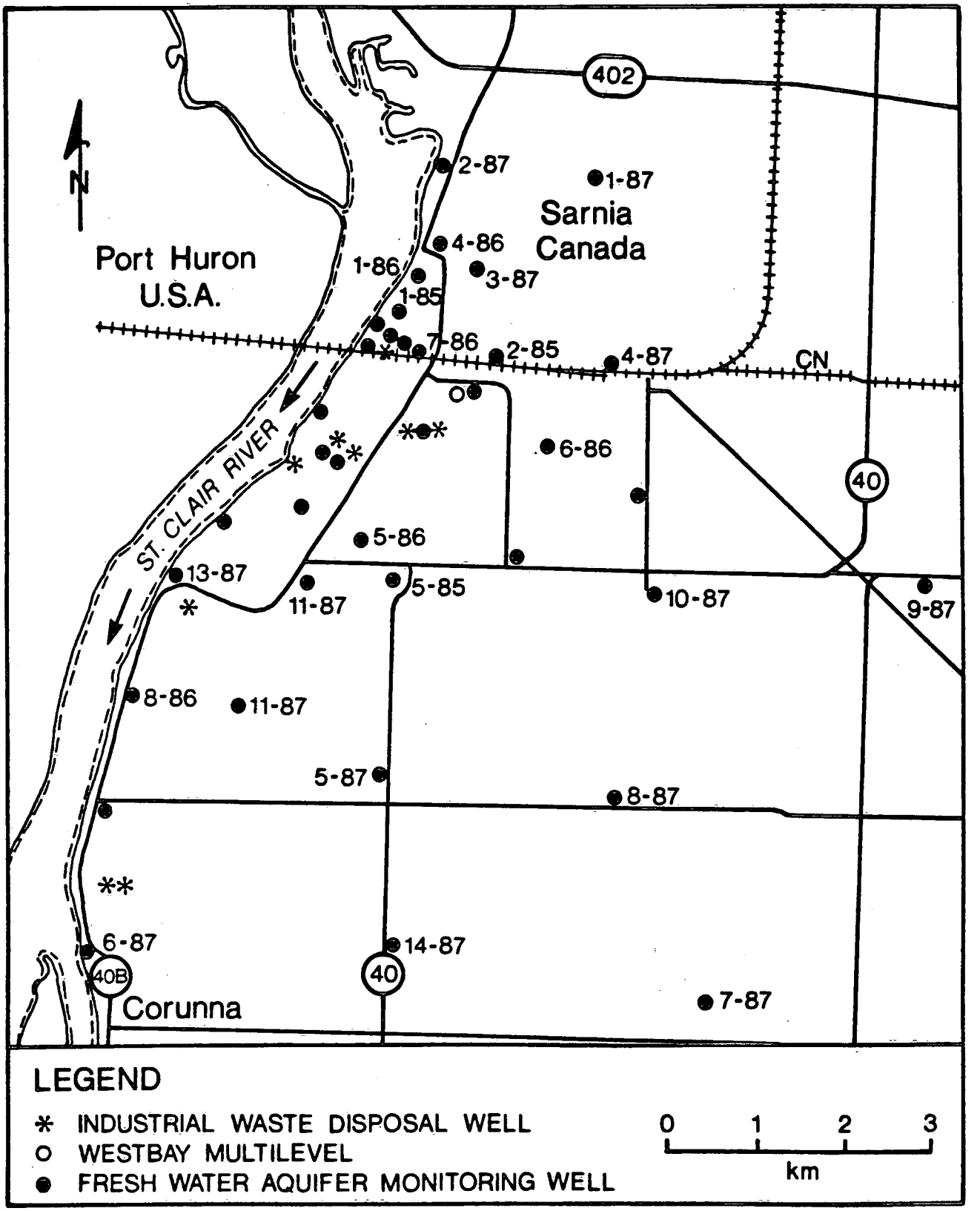
Well #	Benzene	Toluene	Ethyl-Benzene	Styrene	Xylenes (sum)	2-Ethyl Toluene	Phenols [a]
1-87	0.00	0.00	0.00	0.00	0.00	0.00	<0.1
4-87	0.00	0.00	0.08	0.10	0.00	0.10	<0.1
5-87	0.00	0.00	0.00	0.00	0.00	0.50	<0.1
6-87	0.40	0.10	0.00	0.00	0.10	0.05	<0.1
8-87	0.00	0.07	0.17	0.00	0.25	0.18	<0.1
10-87	0.00	0.27	0.25	0.00	0.34	0.21	<0.1
11-87	0.00	0.15	0.10	0.00	0.28	0.07	<0.1
12-87	0.58	0.32	0.16	0.00	0.42	0.08	0.1
13-87	0.00	0.00	0.00	0.00	0.00	0.05	<0.1

[a] Data from the Ontario Ministry of the Environment, London, Ontario Total phenols by the 4-AAP method.

Table 4. Organic chemicals measured in µg/L in samples from the freshwater aquifer monitoring wells collected on June 5-6, 1989

Well #	Benzene	Toluene	Ethyl-Benzene	Xylenes m + p[a]	Cumene	2-Ethyl toluene
4-87	0.00	0.00	0.00	0.05	0.00	0.00
5-87	0.01	0.07	0.00	0.03	0.00	0.02
6-87	0.14	0.18	0.00	0.06	0.00	0.02
11-87	0.00	0.00	0.00	0.03	0.00	0.02
13-87	0.03	0.10	0.00	0.05	0.00	0.03
15-87	0.16	0.03	0.00	0.00	0.01	0.00
PW Deep	0.04	0.04	0.00	0.05	0.00	0.02
CN-7	0.03	0.10	0.33	0.02	0.00	0.02

[a] O- xylene → all none detected.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U.S.A.
Sarnia
Canada
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1-85
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9-87
ST. CLAIR RIVER
8-86
11-87
5-87
8-87
6-87
14-87
40B
40
Corunna
7-87
LEGEND
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WESTBAY MULTILEVEL
FRESH WATER AQUIFER MONITORING WELL
0 1 2 3
km

Figure 1

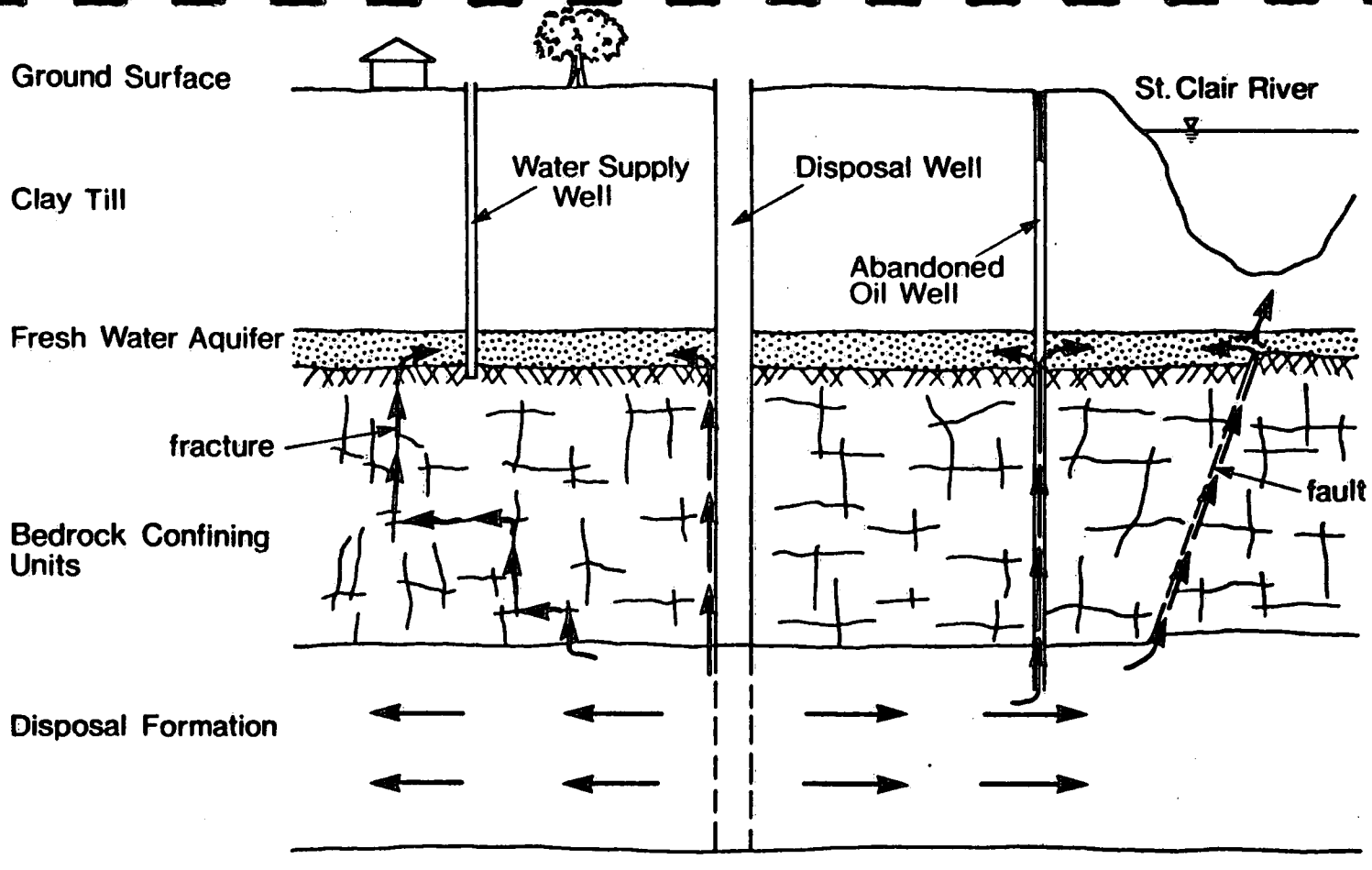
Ground Surface
Clay Till
Fresh Water Aquifer
fracture
Bedrock Confining Units
Disposal Formation
Water Supply Well
Disposal Well
Abandoned Oil Well
St. Clair River
fault

Figure 2

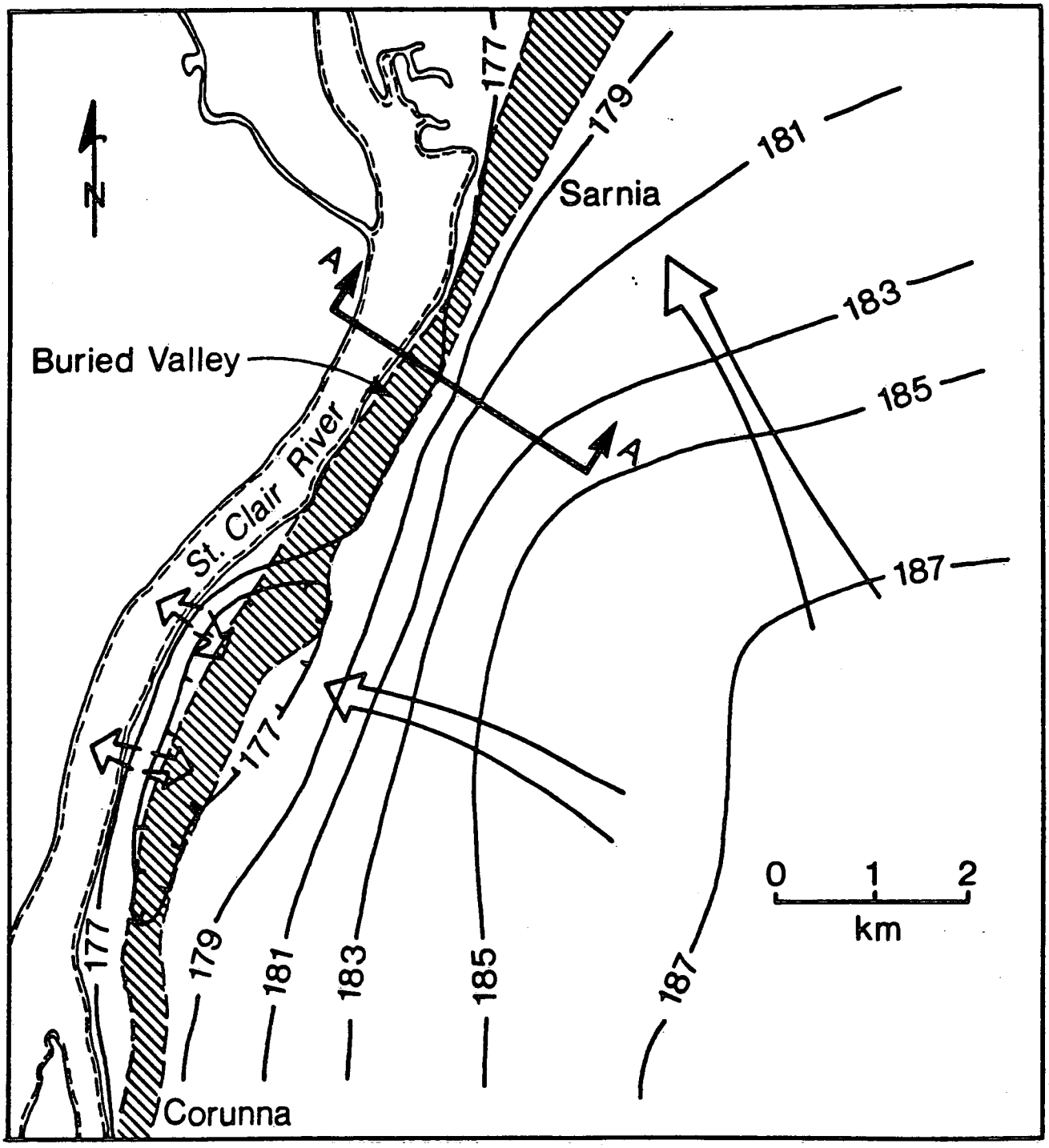
N
Sarnia
Buried Valley
St. Clair River
Corunna
A
A
177
179
181
183
185
187
0
1
2
km

Figure 3

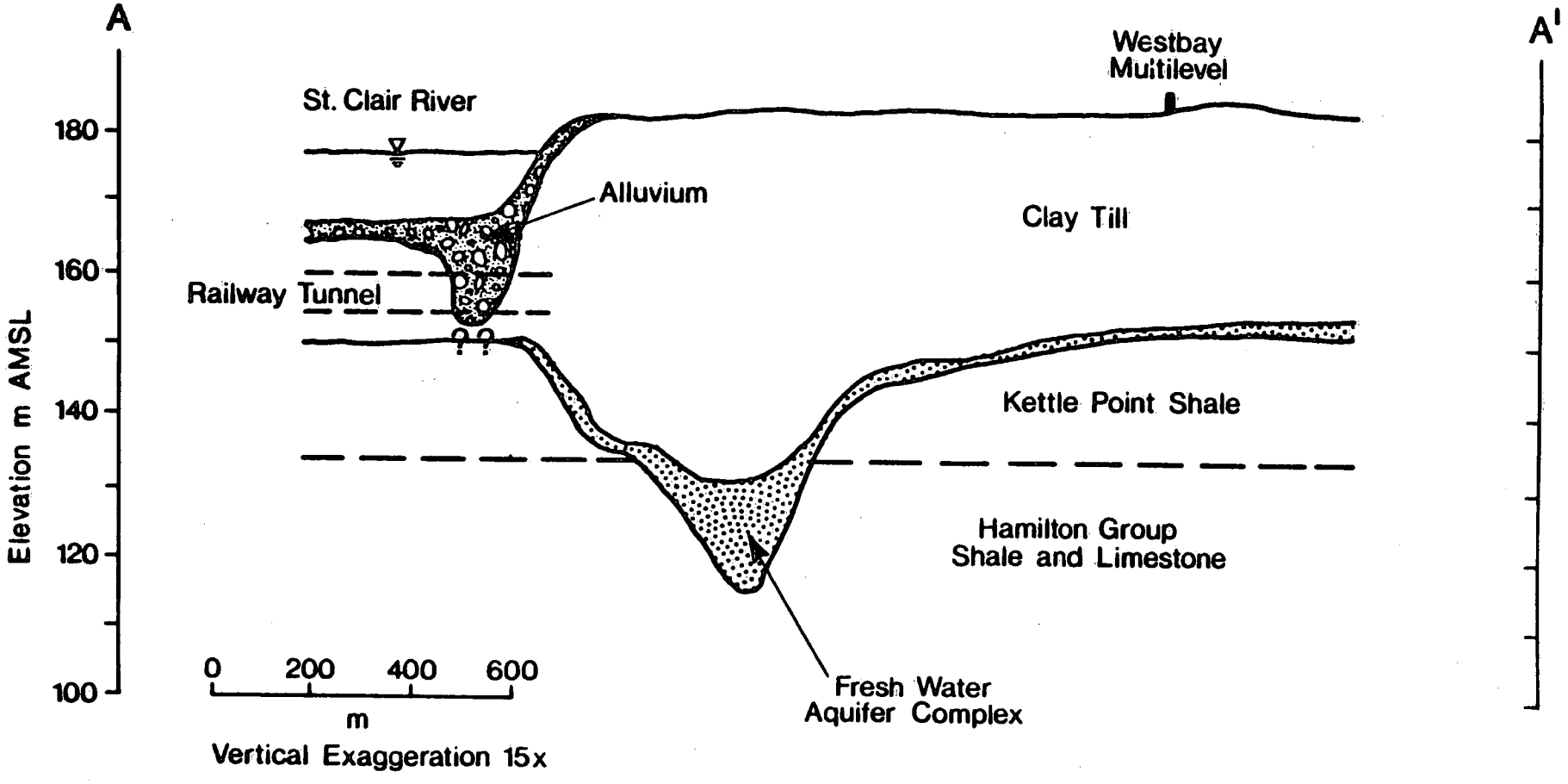
A
A'
Westbay
Muitilevel
St. Clair River
Alluvium
Clay Till
Railway Tunnel
Kettle Point Shale
Hamilton Group
Shale and Limestone
Fresh Water
Aquifer Complex
Elevation m AMSL
180
160
140
120
100
0
200
400
600
m
Vertical Exaggeration 15x

Figure 4

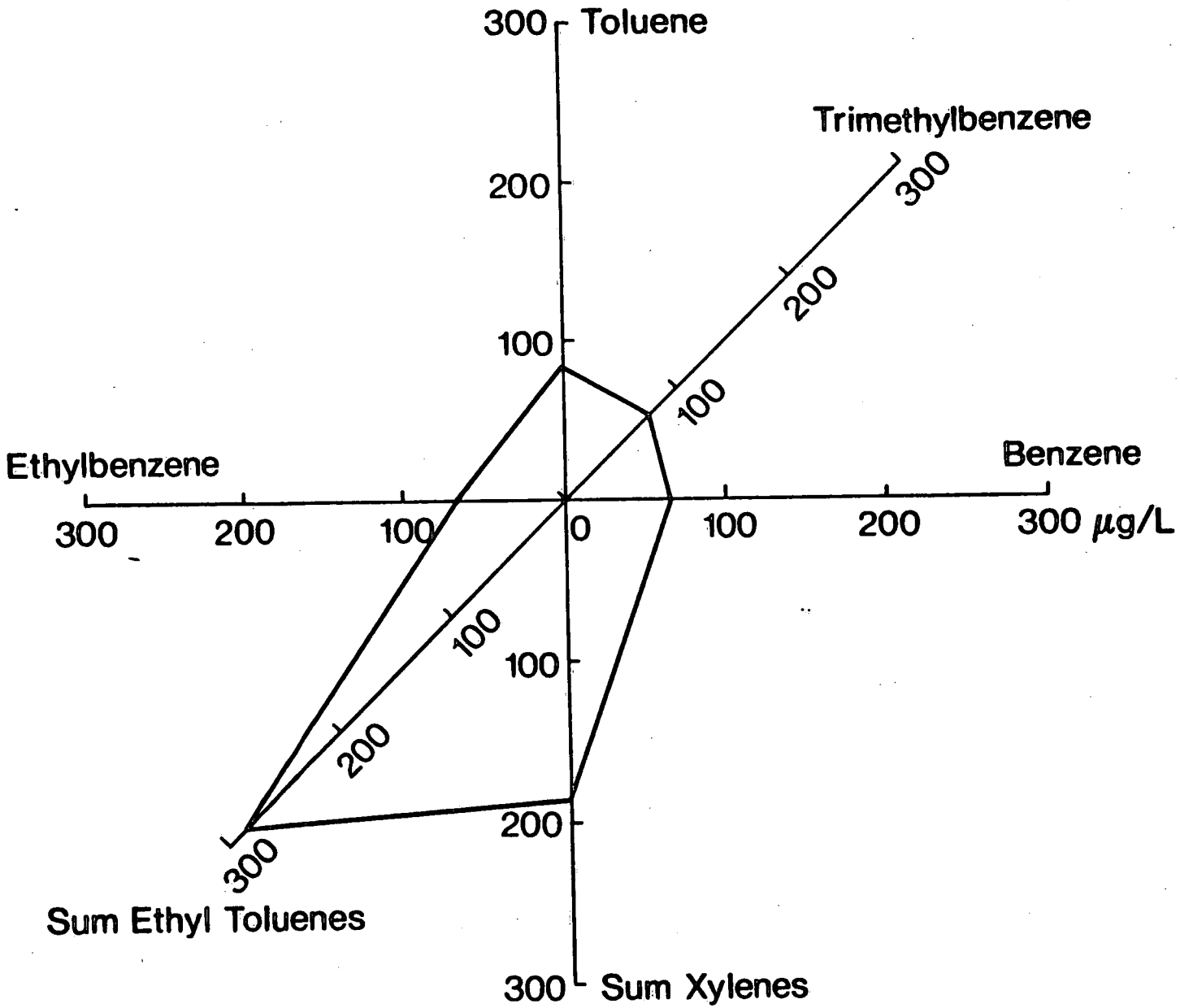
300
Toluene
200
100
Trimethylbenzene
300
200
100
Ethylbenzene
Benzene
300
200
100
0
100
200
300 µg/L
100
100
200
200
300
Sum Ethyl Toluenes
300
Sum Xylenes

Figure 5

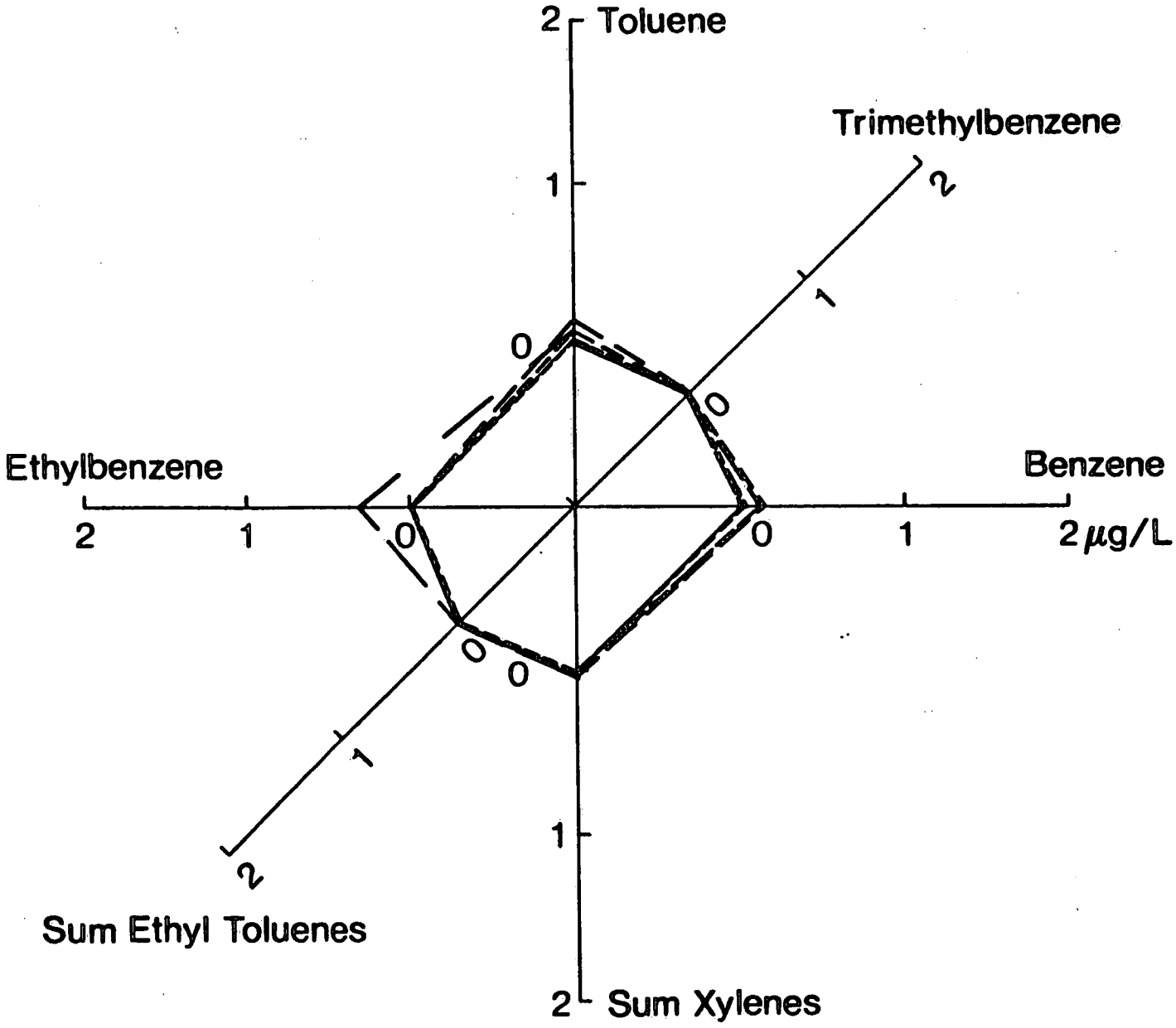
2 Toluene
Trimethylbenzene
1
2
1
0
Ethylbenzene
Benzene
2
1
0
0
1
2 µg/L
0
0
1
2
Sum Ethyl Toluenes
1
2 Sum Xylenes

Figure 6