

CHROMIUM POLLUTION EVALUATION IN GROUNDWATERS OF NORTH MEXICO CITY

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ABSTRACT

Groundwater pollution by soluble chromium species was surveyed throughout a one-year, monthly analysis of 38 wells at the "Municipio de Tultitlán", north of Mexico City. The chosen wells were located in the vicinity of the industrial disposal site of the former plant "Cromatos de México, S.A.", where 75,000 tons of waste material were inadequately confined. The study showed a considerable contamination of the wells closest to the site, due to leaching of chromate salts found in the disposed wastes. The high amount of water consumption by the surrounding industries, is preventing chromium dispersion.

RESUMEN

Se evaluó la contaminación por compuestos de cromo en aguas subterráneas por medio del análisis mensual, durante un año, de 38 pozos en el municipio de Tultitlán, Edo. de México, al norte de la Ciudad de México. Los pozos seleccionados se localizan en la vecindad del cementerio industrial de la ex-planta de Cromatos de México, S.A., donde se encuentran confinadas inadecuadamente 75,000 toneladas de residuos. El estudio muestra una contaminación considerable de los pozos más cercanos al cementerio, debida a la lixiviación de sales de cromato contenidas en los residuos del confinamiento. El alto gasto en el consumo de agua por parte de las industrias circundantes ha evitado la contaminación de mantos acuíferos más profundos y por lo tanto la dispersión del contaminante hacia zonas más lejanas.

INTRODUCTION

This study is part of a more extensive project with the objectives to: evaluate pollution by chromates, interpret geographical data (geomorphology, hydrology, types of soils, etc.) (Gutiérrez *et al.* 1986) and to propose stabilization and reuse of chromate waste material confined in an area north of Mexico City.

The objective of this study was to evaluate the degree and extension of pollution of groundwater wells by chromium compounds, in the vicinity of a disposal site that contains

the residues of a chromate plant in the north of Mexico City*. It is also a goal of this work to identify if the presence of chromium in the aquifers arises from chromate leaching from the waste disposal site (Gutiérrez *et al.* 1986) or from direct injections to the aquifers carried out during the active period of the plant.

The chromate plant was closed in 1978 due to the evident chromate contamination of air and soils of the surrounding areas. 75,000 tons of wastes were left in the open air next to the plant and considerable additional amounts of residues were used to fill cavities in dust roads at the area, due to their gravel appearance. In 1982 the dispersed wastes were gathered and confined into a poorly-constructed disposal facility consisting of a concrete "box" paved on the cover and sides, and built on the field next to the industry, where the residues lie directly on the soil. Nevertheless many of the residues used for filling of roads were left unconfined.

Yellow water observed in some wells close to the plant, led to the conclusion that aquifers were polluted with Cr(VI). In order to determine the intensity and origin of the contamination, as well as possible solutions, the present study was carried out.

The soils of the region are Vertisols (fine textured with $\text{pH} > 7$), and the geomorphology shows accumulation of hundreds of meters of lacustrine clay sediments, interstratified with deposits of fluvial sands and gravels, as well as lavas and sands of volcanic origin. Thus soil profile is very heterogeneous. Moreover, basaltic and andesitic lavas are found 50 m below the surface, increasing considerably the permeability of the subsoil at these depths (Valverde-Valverde *et al.* 1980).

Chromium is present in nature mainly in the form of highly insoluble Cr(III) minerals frequently in combination with iron, e.g. chromite. The other stable oxidation state of chromium is Cr(VI), which forms very soluble chromate compounds (exceptions are the Ag, Ba and Pb chromates). However, the occurrence of appreciable amounts of Cr(VI) in soil and water suggests the artificial addition of chromates (National Research Council of Canada 1976). The insoluble chromium (III) oxides dissolve at pH values below 6. On the other hand, the solubility of Cr(VI) compounds is high at all pH values and they are readily reduced to Cr(III) under acid conditions. High amounts of organic matter, however, have been reported to provide electrons for reduction even at neutral pH conditions. Humic and fulvic acids seem to play an important role in this process (Bartlett and Kimble 1976).

Chromium is generally classified as a beneficial element, but it is not clear whether it is essential or not to life. It helps the growth and reproduction processes in living organisms, especially to higher species (U.S. Environmental Protection Agency 1979) and it is found in animal and plant tissues in concentrations $< 1 \text{ mg/kg}$.

The trivalent form of chromium seems to be the biologically active one (Villalobos-Pietrini 1977). Deficiency of Cr(III) causes mainly a syndrome similar to diabetes, because its function is related to the tolerance factor of glucose (Mertz 1969). However, in excess,

*"Cromatos de México" produced daily, from 1958-1978, 12 tons of Na_2CrO_4 , 1 ton of K_2CrO_4 , approximately 8 tons of Na_2SO_4 and, with the latter, presumably equal molar amounts of dichromates from the reaction of chromates with sulfuric acid.

Cr(III) may be toxic because of its ability to form a variety of coordination compounds with the biomolecules, including DNA and RNA, interfering with their normal functions. Evidence of the carcinogenic and mutagenic effects of Cr has been documented (Villalobos-Pietrini 1977, U.S. Environmental Protection Agency 1979, Gómez-Arroyo *et al.* 1981, 1989, Gómez-Arroyo and Villalobos-Pietrini 1983, Villalobos-Pietrini *et al.* 1986).

The interaction of Cr(VI) with biological material is never beneficial because of its highly oxidizing nature and its great mobility.

Thus, this pollutant will be found, under natural conditions in water, either as dissolved Cr(VI) species and/or as poorly soluble Cr(III) oxide deposited in the sediments, unless slightly acid conditions and the presence of high amounts of organic ligands enhance its dissolution. Therefore, Cr(VI) is potentially more dangerous to living organisms than Cr(III), because of its high solubility.

Due to the variation in natural conditions that determine the oxidation state of Cr, it is necessary to consider the hazardous limits of chromium in water as the total Cr contents.

The highest permissible level of total soluble Cr concentration in drinking water has been set to 0.05 mg/L (National Research Council of Canada 1976, U.S. Environmental Protection Agency 1979, Galvao and Corey 1987). However in rivers Cr(VI) concentrations should not exceed 1.10×10^{-6} mg/L, and the Cr(III) limits are given by formulas that involve the hardness of water (U.S. Environmental Protection Agency 1979).

MATERIALS AND METHODS

Three factors were considered to classify the wells to be sampled in the area: 1. distance from the chromate disposal site, 2. orientation of the main underground drain fluxes (from west to east), and 3. type of soils. Accordingly the area was divided into three groups, in which the number of wells to be monitored was set proportional to the total amount of wells in each area. Wells within each group were chosen by a random sampling method without replacement, influenced by operative limitations (e.g. accessibility to the wells). Details on the sampling design were discussed by Ducoing and Trejo (1987).

Forty four wells were selected (Figure 1), from which 15 are federal and the remaining are private owned or belong to industries in the area of "Lechería". Table I lists properties of the wells for which information was available. Sampling was carried out once a month for a total period of one year (nov. 1984 to nov. 1985).

Water samples were collected in 1 L polyethylene bottles, previously washed with 2M HNO_3 and rinsed with distilled water. Samples were acidified with concentrated nitric acid to a concentration of 0.75M HNO_3 , to avoid precipitation and adsorption of the metal ions on the bottle-walls. A subsample was kept unacidified for pH, electrical conductivity and sodium concentration determinations. The following analyses were carried out:

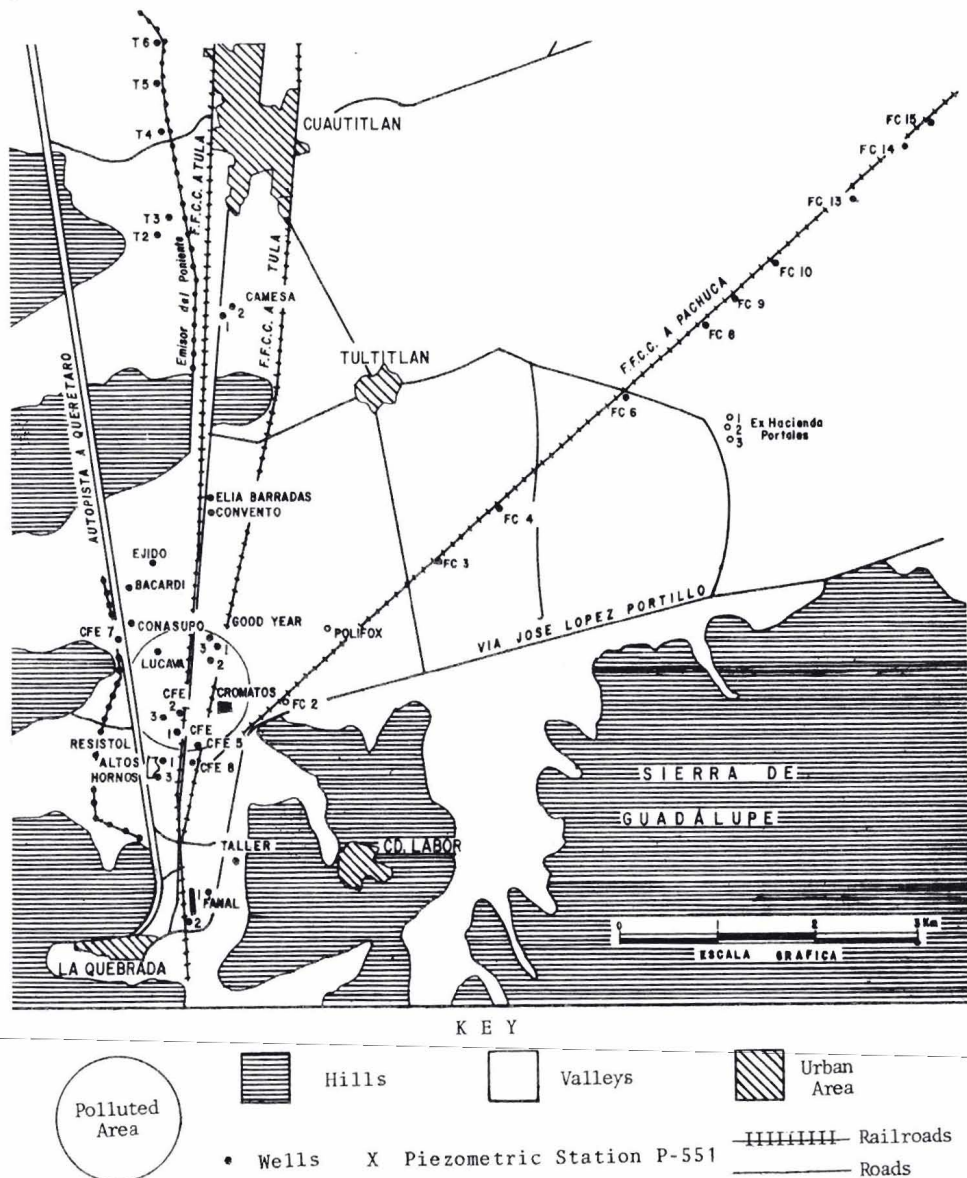


Fig. 1. Location of the sampled groundwater wells in the study area, piezometric station P-551 and chromate disposal site.

Cr(VI) by spectrophotometry: the following additions were done sequentially to 100 mL of Cr(VI) standards (0.010-0.200 mg/L Cr made from a stock solution of $K_2Cr_2O_7$ and acidified with HNO_3 to 0.075M) and water samples in duplicate (diluted if necessary): 1 mL of 1.2M H_2SO_4 and 1 mL of 1, 5-diphenylcarbazide reagent (0.2% in acetone). Color development was completed within 10 minutes and measured in a ZEISS PM2K spec-

TABLE I. SELECTED PARAMETERS OF SOME SAMPLED WELLS (NOV/1984)*

Well No.	Static Level (m)	Dynamic Level (m)	Consumption Per Month (m ³)	Consumption Flux (L/s)
CFE 1	49	55	46258	43.9
CFE 2	54	65	77129	53.0
CFE 3	52	56	75160	35.7
CFE 5	53	73	119336	47.9
CFE 7	64	91	77923	33.5
CFE 8	84	104	127970	59.7
GY 1	---	---	38453	22.0
GY 2	---	---	8010	25.6
GY 3	---	---	36183	25.8
FC 2	20	32	---	---
FC 3	45	82	---	33
FC 4	45	90	---	23
FC 6	36	90	---	35
FC 8	42	65	---	37
FC 9	33	39	---	18
FC 10	40	90	---	38
FC 12	35	50	---	27
FC 14	36	---	---	---
FC 15	35	50	---	54
EX-P 3	---	---	---	20
T 2	63	78	---	30
T 3	58	84	---	68
T 4	54	80	---	30
T 5	69	113	---	54
T 6	63	113	---	38
CAM 1	---	---	---	2
CAM 2	---	---	---	2
BAC 1	46	---	---	13

*Data from wells CFE and GY were averaged in the period November 1984 to November 1985.

(Key to the abbreviations appears in table III)

trophotometer at a wavelength of 540 nm. Measurements were carried out in triplicate (Villalobos-Peñalosa *et al.* 1987).

Total Cr by Atomic Absorption: 1 mL of 30% H₂O₂ was added to 100 mL water sample (in duplicate) and HNO₃-acidified standards (0.20-2.00 mg/L Cr(VI)). The solutions were mixed and aspirated into a VARIAN A440 atomic absorption spectrophotometer at: 357.9 nm wavelength, 0.2 nm slit width, 7 mA lamp current, air/acetylene flame. Measurements were made in triplicate.

Total Mn and Fe by Atomic Absorption: determinations of these cations were carried out using the methods described by Varian (1979) on the same equipment as for the total Cr determinations.

Sodium: analysis was carried out using a CORNING 400 flame photometer and standards from 1 to 10 mg/L according to Rodier (1980).

Electrical Conductivity: a COLE - PARMER 148100 conductivity meter was used, calibrated with a 0.005M KCl solution (720 μ mhos/cm at 20°C) (Rodier 1980).

pH: a CONDUCTRONIC PH20 pH meter equipped with a SARGENT - WELCH 530072 electrode were used and calibration was carried out with SIGMA buffer solutions of pH 7 and 9.

Piezometric Analysis: data were obtained from 12 multipiezometers installed in the study area by the Secretaría de Agricultura y Recursos Hidráulicos (1978). The hydraulic potentials were analyzed for a period of 4.5 years, providing information on the water flux in time and space.

RESULTS AND DISCUSSION

No statistical difference was obtained in the determination of Cr by the atomic absorption and the spectrophotometric methods (see the confidence limits listed at the bottom of Table II), hence no Cr(III) was detected. This was expected from the high pH values measured. No interference from Fe(III) was observed in the polluted samples, due to the low values found for this component (Villalobos-Peñalosa *et al.* 1987).

The average values of the monthly results of pH, electrical conductivity, Na^+ , Fe(III), Mn(II) and Cr(VI) concentrations from the analyses of 1 year sampling are shown in tables II and III.

Wells were considered polluted when they showed Cr(VI) concentrations above the permissible sanitary limit of 0.05 mg/L ($\approx 10^{-6}$ M) at least once during the year sampled. Of the total 44 groundwater wells sampled, 10 wells were polluted with Cr(VI): CFE 1, 2, 3, 5, and 8; Good Year 1, 2, and 3; Lucava 1 and Resistol 2. 7 wells showed intermediate concentrations between the detection limit and the permissible limit: CFE 7; Resistol 1; Altos Hornos 1 and 2; FC 15; T4 and Ex-Hacienda Portales 3. The remaining wells showed chromium (VI) concentrations below the detection limit (0.008 mg/L by colorimetry).

Polluted wells are overexploited and belong to the four industries closest to the disposal site (Fig. 1). The wells occupy an area not larger than 1300 m in radius, west to the waste disposal site. In general, Cr(VI) concentrations show considerable fluctuations with time (see standard deviations in Table II), and no similar patterns or tendencies are observed between each well (results not shown). The individual concentrations were analyzed and statistically related to different parameters, using a linear correlation model: static and dynamic levels of the water in the wells, water consumption (see Table I), discharge, distance from the disposal site and rainfall.

The following results were obtained:

Well Good Year 2 showed considerably higher Cr(VI) concentrations than the other polluted wells. This well is located closest to the disposal site. A direct relationship is observed between the amount of rainfall in the considered time-period and the Cr(VI)

TABLE II. AVERAGE METAL CONCENTRATIONS OF WELLS SAMPLED OVER THE PERIOD NOV/1984 TO NOV/1985

Well No.	Fe (III) (mg/L)	Mn(II) (mg/L)	Cr(VI) (mg/L)			
			Colorimetry		Atomic Absorption	
			C.V.		C.V.	
FC 2	0.26	nd	nd	----	nd	----
FC 3	nd	nd	nd	----	nd	----
FC 4	nd	nd	nd	----	nd	----
FC 6	nd	0.5	nd	----	nd	----
FC 8	nd	0.2	nd	----	nd	----
FC 9	nd	0.3	nd	----	nd	----
FC 10	nd	0.2	nd	----	nd	----
FC 12	nd	0.4	nd	----	nd	----
FC 14	0.30	nd	nd	----	nd	----
FC 15	nd	0.1	nd	----	nd	----
EX-P 1	2.69	1.1	nd	----	nd	----
EX-P 2	0.05	0.5	nd	----	nd	----
EX-P 3	1.22	1.6	nd	----	nd	----
T 2	nd	nd	nd	----	nd	----
T 3	nd	nd	nd	----	nd	----
T 4	nd	nd	nd	----	nd	----
T 5	nd	nd	nd	----	nd	----
T 6	nd	nd	nd	----	nd	----
CAM 1	nd	nd	nd	----	nd	----
CAM 2	nd	nd	nd	----	nd	----
FAN 1	nd	nd	nd	----	nd	----
FAN 2	nd	nd	nd	----	nd	----
BAC 1	nd	nd	nd	----	nd	----
CON 1	nd	nd	nd	----	nd	----
AHMSA 1	1.46	0.1	nd	----	nd	----
AHMSA 3	nd	nd	nd	----	nd	----
CFE 1	nd	nd	1.50 ± 0.12	0.52	1.62	0.55
CFE 2	nd	nd	1.22 ± 0.1	0.53	1.21	0.51
CFE 3	0.41	nd	0.043	1.52	nd	----
CFE 5	nd	nd	0.91 ± 0.07	0.54	0.84	0.45
CFE 7	nd	nd	nd	----	nd	----
CFE 8	0.31	nd	0.053	1.31	nd	----
GY 1	0.09	nd	0.24 ± 0.02	2.60	0.25	2.50
GY 2	0.33	nd	6.0 ± 0.5	0.30	8.00	0.33
GY 3	nd	nd	0.287	0.19	0.31	0.25
LUC 1	0.16	nd	0.071	0.80	nd	----
RES 1	nd	nd	nd	----	nd	----
RES 2	nd	nd	0.041	0.15	nd	----

c.v. = coefficient of variation (= standard deviation/mean)

nd = not detected

DETECTION LIMITS ARE:

Cr: 0.008 mg/L FOR COLORIMETRY, 0.09 mg/L FOR ATOMIC ABSORPTION

Fe: 0.05 mg/L, Mn: 0.1 mg/L

CONFIDENCE LIMITS FOR INDIVIDUAL MEASUREMENTS (90% LEVEL):

COLORIMETRY = ±0.008 mg/L (EXCEPT WHERE INDICATED)

ATOMIC ABSORPTION = ± 0.06 mg/L (8.0 mg/L ± 0.7)

(Key to the Abbreviations appear in Table III)

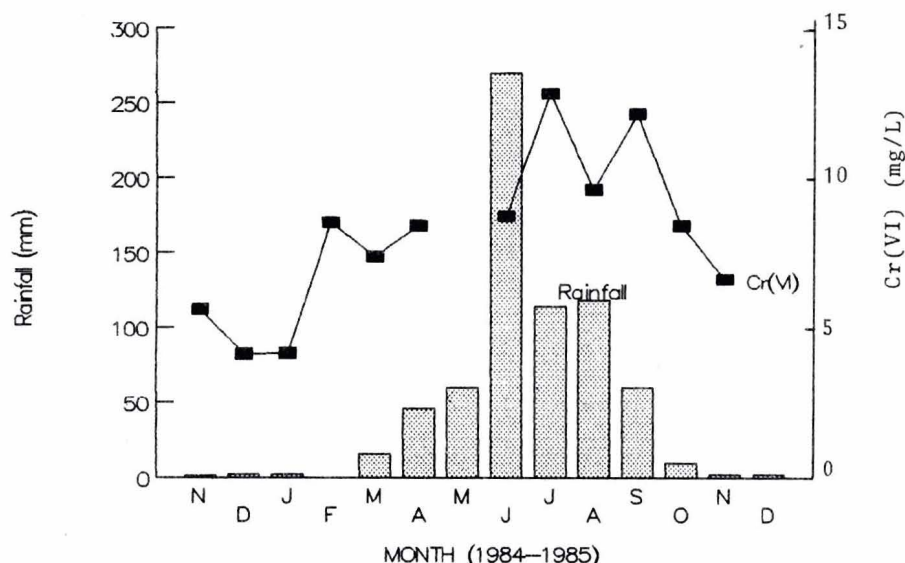


Fig. 2. Monthly Cr(VI) concentrations in well Good Year 2 and amount of rainfall from November 1984 to December 1985.

concentration. Figure 2 shows that during the rainy season (month to month), the maximum Cr(VI) concentrations were detected.

This trend was not observed for the other polluted wells. However an average of the Cr(VI) concentrations for each well during the wet and dry seasons showed higher Cr(VI) concentrations in the wet season (except in well Good Year 3) (Figure 3). Nevertheless, this relationship is only statistically significant for well Good Year 2.

A decrease in Cr content could be observed as the distance from the disposal site increased. This relationship, however, was not linear ($r = -0.56$, $t = 0.66$ at 0.05 level).

The highest Cr(VI) concentrations were found in the industries with high water consumption. This relationship is also non-linear.

For a same well the variation of pH with time was not very high (variation coefficients range from 1% to 10%), while the sodium concentrations and the electrical conductivity fluctuated largely (variation coefficients reached values up to 72% for both parameters). Averages are presented in table III.

Information from other studies (Secretaría de Agricultura y Recursos Hidráulicos 1961, 1978) was available on the concentration of Cr in the wells of Good Year from

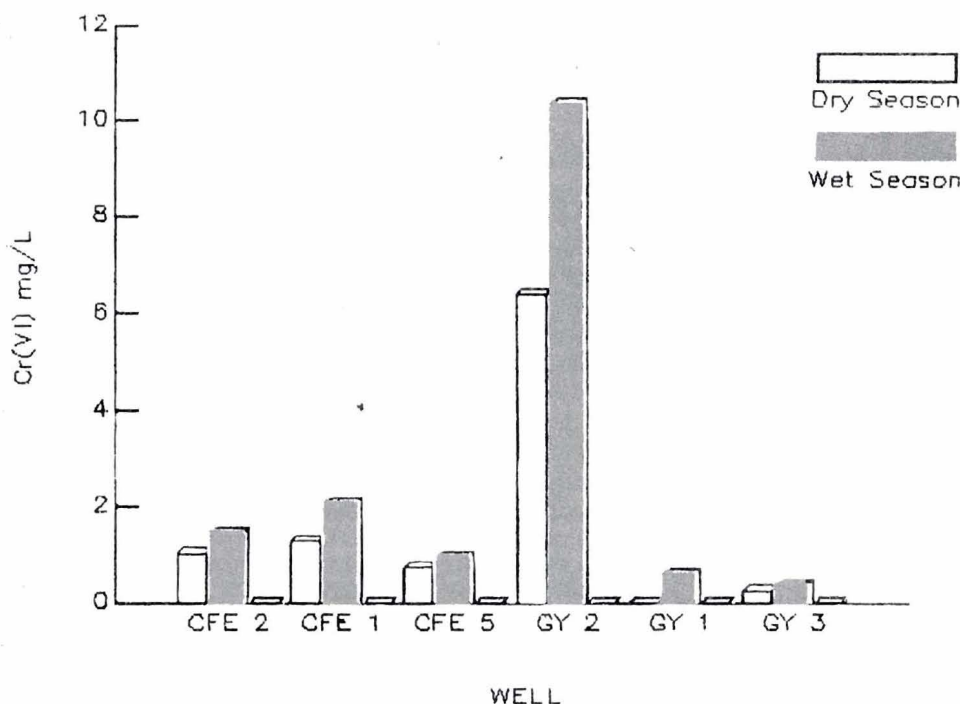


Fig. 3. Average Cr(VI) concentrations in the dry and wet season for the most contaminated wells
 Dry season: months 11, 12, 1, 2, 3, 4
 Wet season: months 6, 7, 8, 9, 10

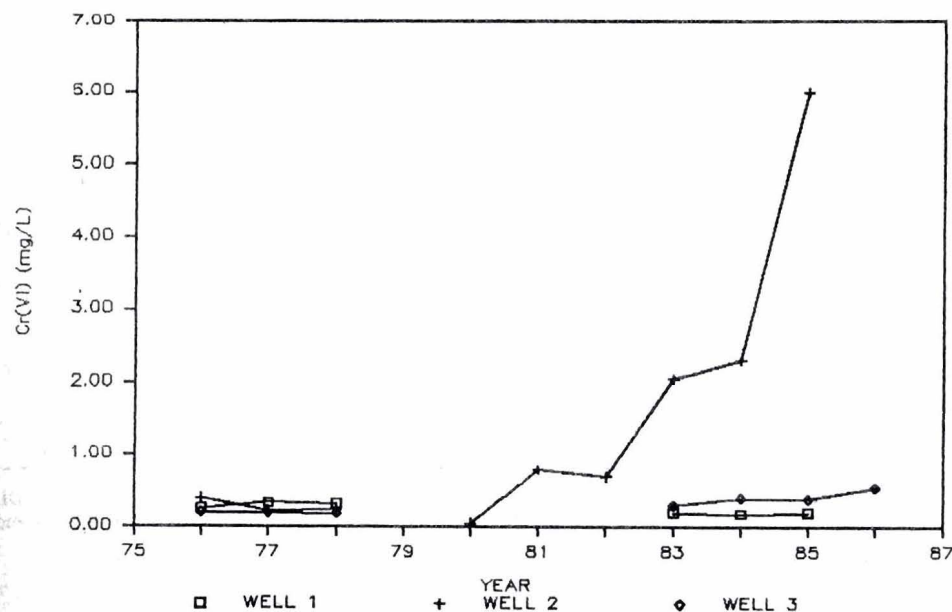


Fig. 4. Average annual Cr(VI) concentrations in the Good Year wells during the period 1976-1986.

TABLE III. AVERAGE CHEMICAL ANALYSES OF WELLS SAMPLED OVER THE PERIOD NOV/1984 TO NOV/1985
(Routine Parameters)

Well No.	EC mmhos/cm		Na ⁺ (mg/L)		pH	
	C.V.		C.V.		C.V.	
FC 2	0.83	0.21	81	0.20	7.81	0.07
FC 3	0.65	0.29	76	0.48	7.92	0.06
FC 4	0.45	0.10	53	0.09	7.68	0.06
FC 6	0.59	0.23	64	0.31	7.62	0.07
FC 8	0.57	0.22	64	0.20	7.52	0.03
FC 9	0.48	0.11	47	0.46	7.47	0.03
FC 10	0.59	0.08	61	0.10	7.49	0.03
FC 12	0.62	0.07	63	0.08	7.84	0.05
FC 14	0.63	0.01	65	0.01	8.02	0.06
FC 15	0.68	0.01	87	0.05	7.95	0.05
EX-P 1	1.71	0.12	321	0.18	8.02	0.05
EX-P 2	1.64	0.04	348	0.18	8.24	0.05
EX-P 3	1.25	0.18	186	0.37	7.89	0.07
T 2	0.47	0.10	45	0.45	7.61	0.06
T 3	0.37	0.07	42	0.06	7.61	0.06
T 4	0.35	0.05	31	0.45	7.35	0.04
T 5	0.40	0.31	37	0.43	7.75	0.05
T 6	0.28	0.02	17	0.72	7.32	0.01
CAM 1	1.95	0.72	123	0.40	7.47	0.04
CAM 2	1.96	0.73	129	0.53	7.46	0.04
FAN 1	0.51	0.04	111	0.50	7.84	0.07
FAN 2	0.65	0.05	106	0.24	7.97	0.06
BAC 1	0.57	0.06	58	0.10	8.07	0.06
CON 1	0.46	0.05	51	0.07	8.01	0.05
AHMSA 1	1.01	0.34	101	0.36	7.85	0.08
AHMSA 3	0.60	0.04	79	0.39	7.82	0.06
CFE 1	0.67	0.12	88	0.18	7.71	0.07
CFE 2	0.61	0.21	80	0.28	7.86	0.05
CFE 3	0.44	0.08	57	0.15	7.98	0.05
CFE 5	1.01	0.11	136	0.42	7.31	0.10
CFE 7	0.42	0.04	47	0.11	8.05	0.06
CFE 8	0.76	0.10	112	0.10	7.78	0.07
GY 1	0.54	0.10	60	0.18	8.03	0.06
GY 2	0.60	0.08	62	0.14	8.04	0.05
GY 3	0.61	0.08	64	0.18	7.90	0.06
LUC 1	0.48	0.05	58	0.07	8.43	0.02
RES 1	0.48	0.08	65	0.07	8.17	0.03
RES 2	0.57	0.10	81	0.07	8.18	0.03

Key: EX-P = Ex Hacienda Portales, CAM = Camesa, FAN = FANAL, BAC = BACARDI, CON = CONASUPO, AHMSA = ALTOS HORNOS, GY = GOOD YEAR, LUC = LUCAVA and RES = RESISTOL.

C.V. = Coefficient of Variation (= standard deviation/mean)

1976-1986. These results are presented in figure 4, and they show that the concentration of Cr(VI) in water from well 2 increased considerably, while in the other two wells it remained around approximately constant values.

The low concentrations of Fe and Mn showed that water from the wells was not polluted by metallic sources such as the pumping equipment, etc. Therefore the possibility of Cr contamination from these same sources was discarded. The three wells that showed relatively high concentrations of Fe and Mn (i.e., Ex-Hacienda Portales 1 and 3, and Altos Hornos 1), did not show Cr(VI) levels above the permissible limit of 0.05 mg/L.

The results of the piezometric study in station P-551 are illustrated in figure 5. This station is located closest to the disposal site. The hydraulic head decreased within depths from 35 to 100 m, indicating a soil water flow in downward direction. The difference in hydraulic heads has increased over the 4.5 year period considered, at a rate of 0.4m per year, at 50 m depth, and 3.8 m per year, at 100 m depth. Moreover, in 1981 a water flux inversion occurred at 67-70 m depth and the piezometer at 67 m depth dried up in 1986. These effects are the consequences of an excessive water consumption from the wells by the surrounding industries at 70 m depth, preventing soluble chromate migration to deeper horizons and aquifers, where so far no chromate has been detected.

CONCLUSIONS

The data obtained in this study confirm that the most probable source of chromium found in the wells is the disposal site, due to the following observations:

- i. The polluted wells are the closest to the confinement and are those of highest consumption.
- ii. The chromium contents of the well Good Year 2, which is closest to the disposal site, have increased considerably with time. This suggests that there is a continuous source that replenishes the aquifer. Considering the disposal site an abundant chromate-waste deposit, it may constitute a more plausible contaminating source than, for example, injections of liquid wastes directly to the industrial well, that were supposedly carried out during the active period of the plant. Anyway, if this was the pollution source, Cr(VI) concentrations would tend to diminish gradually, especially after such a long time period since the industry was closed.
- iii. The piezometric study showed that water flows downward through the clayey profile of the valley. Additional evidence is shown by a geoelectric study carried out by González-Morán *et al.* (1989) in the disposal site, which confirms that Cr(VI) moves towards the subsoil (Gutiérrez-Ruiz *et al.* 1988).

The flow of polluted water through the clayey subsoil to the aquifers is taking place at a much faster rate than would be expected. This is due to the overexploitation of the surrounding wells, which has caused the clays to acquire a secondary porosity. The groundwater table has decreased more than 30 meters in 20 years (Gutiérrez-Ruiz *et al.* 1986), and since the industries using the wells lie to the west of the disposal site (Fig. 1), the local flows are inverted with respect to the general pattern (west to east) (Gutiérrez-

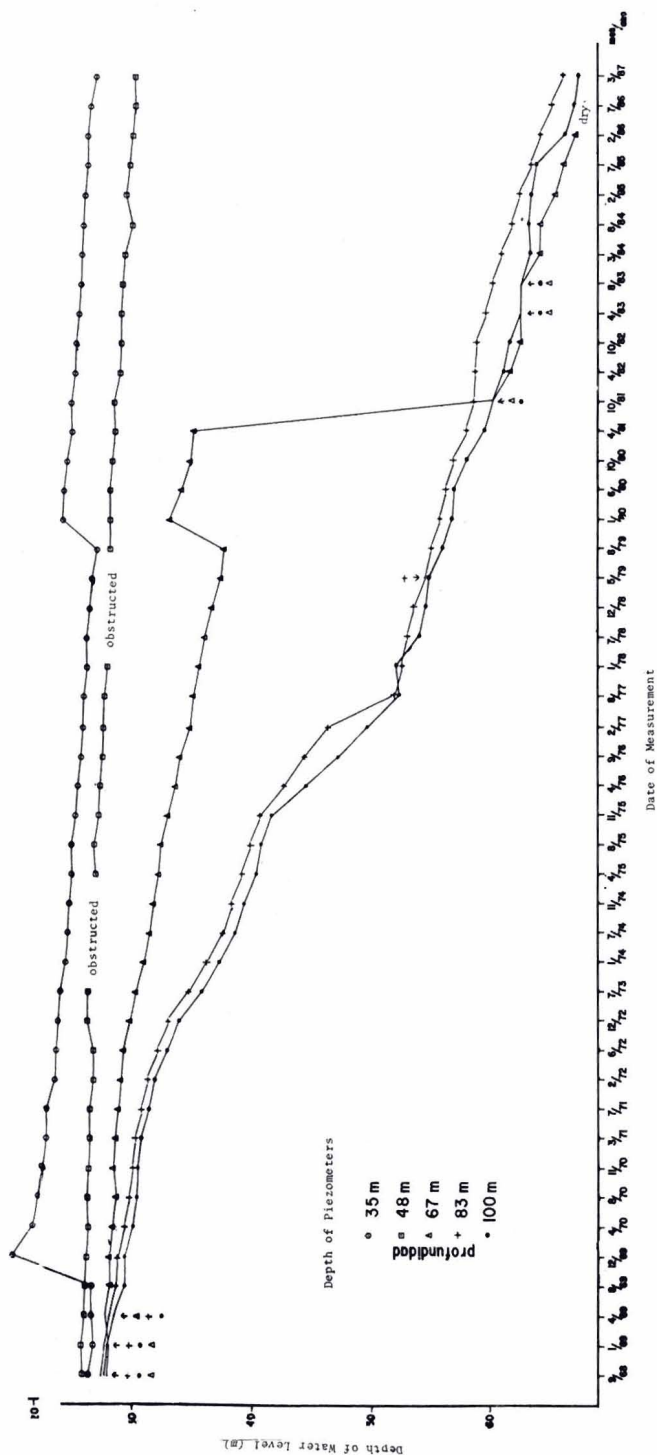


Fig. 5. Depth of water levels in the piezometers of station P-551 in Tultitlan State of Mexico (from September 1968 to March 1987).

Ruiz *et al.* 1986). Cr(VI) is thus moving from the disposal site downwards through the very heterogeneous and anisotropic soil profile, and towards the industries above mentioned (from east to west).

Overexploitation of the wells has prevented chromium dispersion. Until now the polluted zone is restricted to a very localized area. In case the Cr(VI) reaches depths over 50 m, dispersion will increase considerably because of fractures and sandy material found at these depths.

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