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Formation of a salt plume in the Coastal Plain aquifer of Israel: the Be'er Toviyya region

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Abstract

The formation and development of a salt plume (salinity up to 800 mg Cl l^{-1}) in the inner part of the Coastal Plain aquifer of Israel is analyzed. Massive groundwater exploitation during the 1950s caused a large drop in the water level and formation of a hydrologic depression in the Be'er Toviyya–Kefar Warburg area. The depression reached a maximal depth during the late 1960s; thereafter a reduction in the rate of pumpage led to restoration of water levels and shallowing of the depression, until its complete disappearance towards the end of the 1980s. A spot of high salinity first appeared in 1956, following a deep drawdown in the water levels. This saline plume has been continuously expanding with increasing salinity concentrations ($200\text{--}800 \text{ mg Cl l}^{-1}$) in its center. The average rate of radial expansion was about 50 m year^{-1} . The expansion and salinization did not cease as the depression disappeared. Rather, equalization of water levels in wells situated within the plume area with those of situated along its margins resulted in the salinization of the latter within a period of 1 year.

Mass balances for water and chloride contents were made for the period 1967–1990. Taking into consideration the storage change, pumpage, natural replenishment and artificial recharge, the lateral inflow to the depression is estimated as $60 \times 10^6 \text{ m}^3$. Upon addition of the chloride balance, and taking into consideration the chloride concentrations of the surrounding fresh water and the apparent possible end-member of the saline source (based on geochemical considerations), the saline inflow is estimated as $(40\text{--}60) \times 10^6 \text{ m}^3$. These estimates indicate that a large amount of saline water penetrated into the aquifer, of about half of the natural replenishment of the study area, with an estimated salinity of $1900\text{--}2700 \text{ mg Cl l}^{-1}$.

It is suggested that the salt plume was formed as a result of a drop in water level combined with a flow of underlying saline water bodies from deeper strata. The chemical composition of the groundwater points to the existence of two saline water bodies of Ca-chloride composition

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and a marine Br/Cl ratio: (1) saline water with low Na/Cl (0.6), SO₄/Cl, and B/Cl ratio; (2) saline water with higher Na/Cl (> 0.6), SO₄/Cl, and B/Cl ratios. These chemical compositions resemble Ca-chloride saline waters found in other locations in the Coastal Plain aquifer and in underlying formations. The saline water bodies may occur in either pockets at the bottom of the aquifer or lumachelle and sandstone layers of high hydraulic conductivity in underlying sediments.

1. Introduction

Groundwater quality in the Coastal Plain aquifer of Israel has been impaired owing to salinization processes along the western and eastern margins, and also in internal areas of the aquifer, in the form of salt plumes. Salt plume phenomena are found in several areas in the Coastal Plain (Fig. 1), with salinities varying between 200 and 800 mg Cl l⁻¹. Some of these plumes have expanded over the years and have even merged with neighboring plumes, and some were obscured by saline waters that penetrated along the western margins of the aquifer. The salt plume in the Be'er Toviyya area (Fig. 1) is an example, however, of a saline plume growing and expanding in the aquifer with no apparent direct cause. Because of its distance from the coastline (about 12 km) and from neighboring plumes, the plume is isolated from other sources of salinization. This facilitates uncovering the sources of salinization and reconstructing the development of the salinity in this area. The salt plume in the Be'er Toviyya area is characterized by high salinity (800 mg Cl l⁻¹) and relatively broad geographic spread (about 9 km²; Fig. 2(A)). Yet the present water-level contour (Fig. 2(B)) does not show any anomaly in groundwater levels of the area.

Most of the studies of salt plumes in groundwater systems are related to artificial intrusion of saline water into aquifers and formation of a salt-water contaminant plume (e.g. Ellsworth and Jury, 1991; Ellsworth et al., 1991; Oostrom et al., 1991). Yet the formation of salt plumes in the coastal aquifer of Israel is not necessarily directly related to anthropogenic sources. The present study investigates the origin and formation of the salt plume in the Be'er Toviyya area in order to understand the formation mechanism of salt plumes in the aquifer in general. The study was based on the past changes in water levels, chloride concentrations, pumpage data and chemical data of wells from the study area. In addition, new chemical analyses of the groundwater in the area were carried out.

2. General description and hydrogeological background

The study area (Fig. 1) is located in the agricultural region of the Be'er Toviyya, Kefar Warburg, Avigedor, Kefar Ahim and Orot settlements and the municipal area of Qiryat Malakhi, some 12 km east the coastline. Cutting across in the northeast corner of the area is Nahal (creek) HaEla, which is fed by surface runoff, excess sewage effluents and by direct flow of industrial wastes. There are two industrial

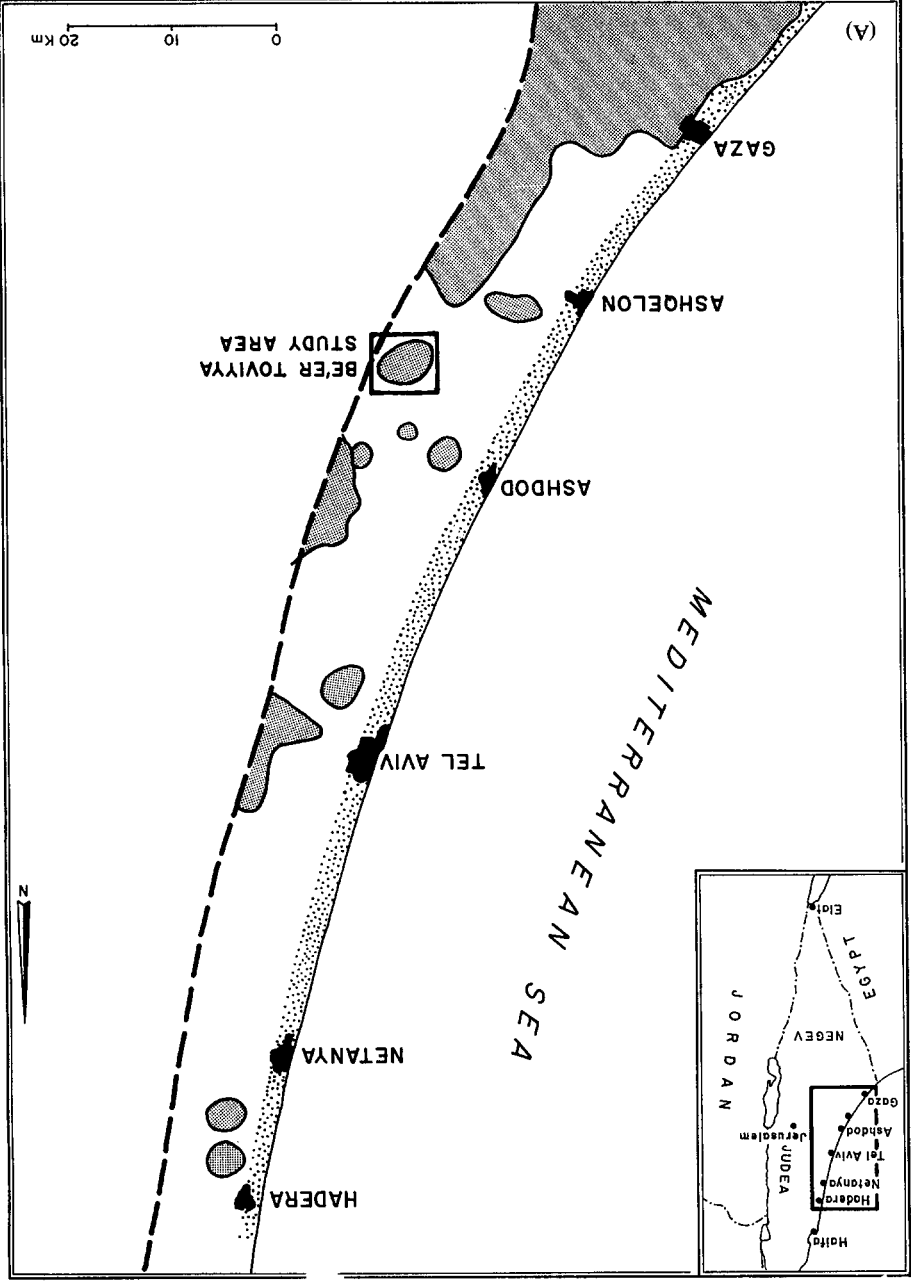
centers in the area that constitute a potential danger to the environment: the industrial area of Qiryat Malakhi, which is on the banks of Nahal HaEla (coordinates 1266/1275), and the salt ponds (coordinates 1263/1253) in the southern area (Fig. 1).

The Coastal Plain aquifer in Israel is a phreatic aquifer, located along the Mediterranean coast (Fig. 1). It varies in width from 7 km in the north to 20 km in the south; its thickness decreases eastwards from 200 m near the coastline to a few meters at the foothills of the Judea Mountains. The aquifer (the Kurkar Group) consists of interlayered sandstone, calcareous sandstone (arenites), siltstone, red loamy soils, and marine clays of Pleistocene age, which overlie impervious marine clays of Pliocene age (the Saqiye Group). Towards the west, up to 5–8 km from the coastline, intervening clay layers divide the aquifer into four to six subaquifers. In the central and eastern areas, the aquifer is uniform and phreatic. The calcareous sandstone (arenites or 'kurkar') is composed of several minerals: quartz, feldspar, calcite, aragonite and iron oxides (Gavish and Friedman, 1969). The diagenetic history of the kurkar rocks started from a mineralogical assemblage that is similar to carbonate sand which is deposited today in the littoral zone. Gavish and Friedman (1969) suggested the following diagenetic evolution: (1) transformation of high-Mg calcite to low-Mg calcite; (2) dissolution of aragonite biogenic skeletons and precipitation of druzey cement (low-Mg calcite).

A schematic cross-section from east to west in the study area (Fig. 3) exhibits a phreatic aquifer of approximately 140 m thickness, overlying impermeable layers of the Saqiye Group. The area is characterized by a soil cover of up to 20 m thickness in the upper part of the unsaturated zone and clay lenses of 5–10 m thickness at depths of 60–70 m. Analysis of the geological section in the area reveals that these lenses are not continuous, and most of the aquifer in the area consists of sandstone and kurkar. The geological evidence is based on data from water wells in the area; however, these wells are only up to 100 m deep and do not penetrate to the base of the aquifer or to the underlying rocks of the Saqiye Group. Therefore, the lithology and the geological structure at the base Kurkar Group–top Saqiye Group boundary in the area is not well known. In various areas of the aquifer some geological anomalies are known: deep depressions containing clays and sandstone occur within the Yafo Formation of the Saqiye Group (Vinokurov et al., 1991), and layers of sandstone and lumachelles (compact limestone composed of fossil mollusc shells) bearing saline water occur at shallow depths of the Saqiye layers (Muravinsky and Grossman, 1982; Dikenstein and Croker, 1983; Fligelman and Shomrony, 1983).

3. Sampling and analytical methods

The hydrological regime presented for the study area is based on changes in water levels, chloride content, and pumpage data of selected wells. The data are from files and the computer data base of the Hydrological Service of Israel. The levels described below represent minimum autumn levels of the particular years noted. The chemical data are based on water samples taken in summer 1991 and in winter 1992 which



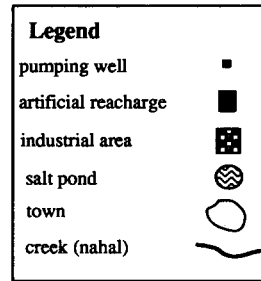
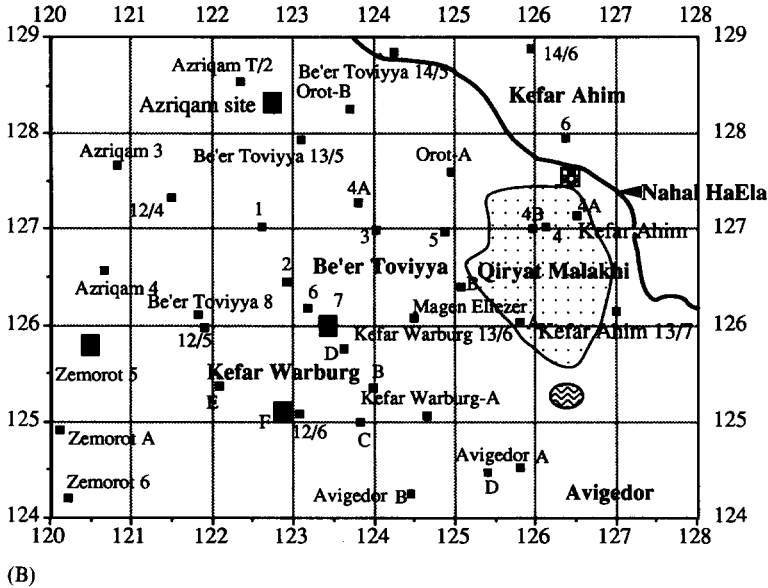


Fig. 1. (A) Location maps of Israel and the Coastal Plain Aquifer including the study area. Areas in the aquifer in which the chloride concentration exceeded 400 mg l^{-1} during 1992 are shown by shading. (B) Detailed map of the study area with locations of the investigated wells.

were analyzed in the laboratory of the Hydrological Service, and on historical data taken from the water-well archives. Potassium and sodium were analyzed by a Perkin Elmer atomic absorption (AA) instrument, calcium and magnesium by EDTA titration, chloride by titration with silver nitrate, sulfate by turbidimetry, bicarbonate by titration with HCl, and nitrate by Corning 476134 specific electrode, adjusted to chloride concentrations. Analytical error did not exceed 4%. Boron and bromide concentrations were determined by inductively coupled plasma (ICP) and X-ray fluorescence (XRF), respectively, at the laboratory of the Geological Survey of Israel. The sodium, calcium and magnesium values in several samples were also measured by AA at the Geological Survey. The data from both laboratories were in complete agreement with each other.

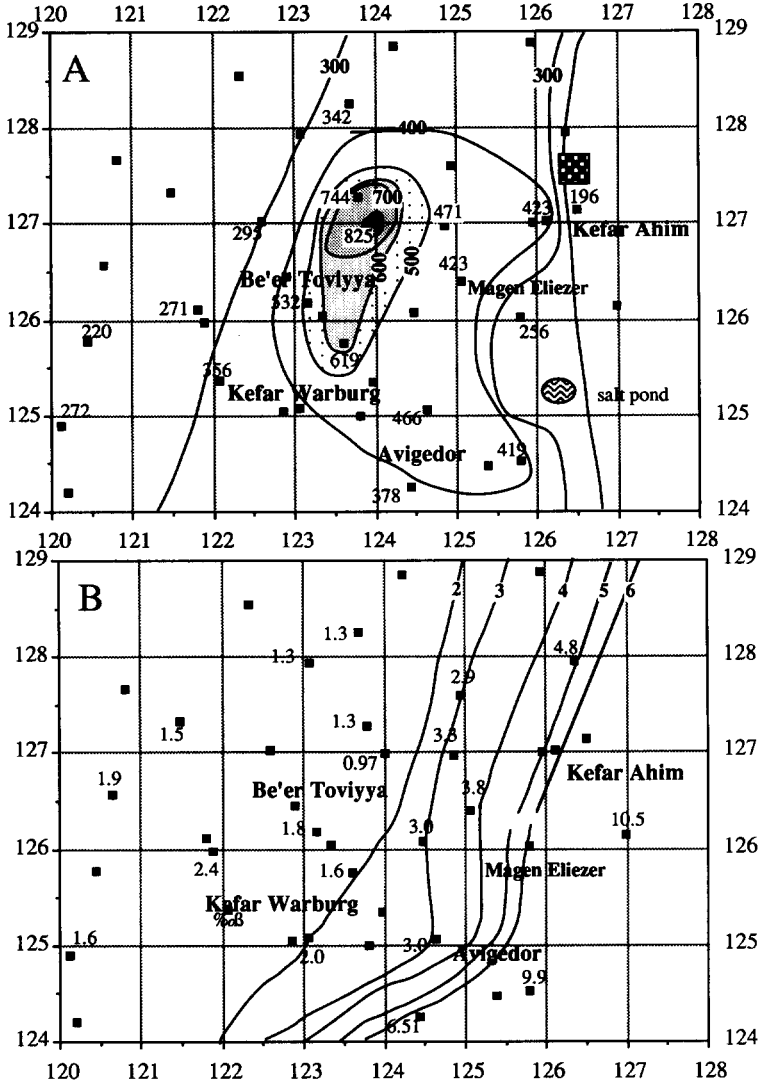


Fig. 2. (A) Map of chloride contents in the study area, summer 1990; (B) map of water-level elevations (relative to sea-level) in the study area, autumn 1991 (see Fig. 1 for the relevant wells).

4. Results

4.1. Water management

The total volumes of pumpage in all the wells of the study area (Table 1) indicate a general decrease in the amounts of groundwater exploited; from $7.5 \times 10^6 \text{ m}^3$ in 1970 to $3.5 \times 10^6 \text{ m}^3$ in 1990 (Fig. 4). During that period a total of $122 \times 10^6 \text{ m}^3$ was pumped in the study area, and the amount of the artificial recharge was

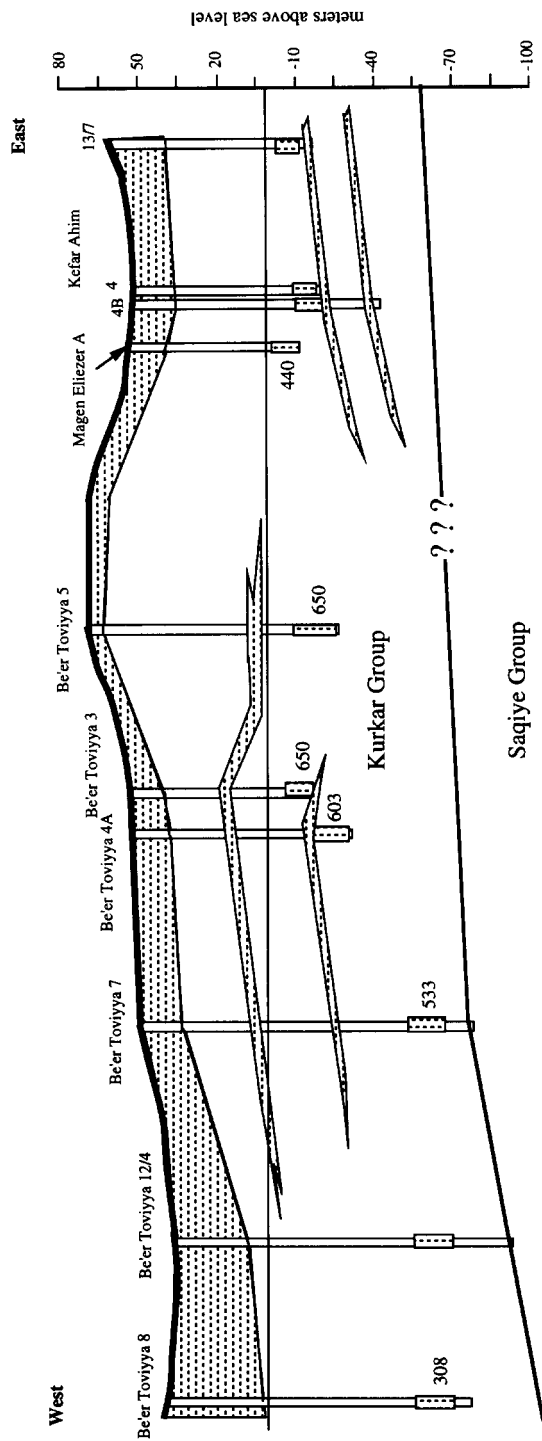


Fig. 3. Schematic hydrogeologic east-west cross-section of the Coastal Plain aquifer in the study area (see locations of the drillholes in Fig. 1(B)). Dashed lines represent foamy soils and clays; numbers are chloride concentrations measured in these wells during 1992.

Table 1
List of wells in the Be'er Toviyya area

| Well | No. | East | West |
|--------------------|----------|--------|--------|
| Be'er Toviyya 1 | 12712201 | 122.60 | 127.03 |
| Be'er Toviyya 2 | 12612201 | 122.90 | 126.45 |
| Be'er Toviyya 3 | 12612403 | 124.02 | 126.99 |
| Be'er Toviyya 4A | 12712302 | 123.78 | 127.27 |
| Be'er Toviyya 5 | 12612402 | 124.86 | 126.97 |
| Be'er Toviyya 6 | 12612301 | 123.16 | 126.18 |
| Be'er Toviyya 7 | 12612302 | 123.34 | 126.04 |
| Be'er Toviyya 8 | 12612101 | 121.80 | 126.11 |
| Be'er Toviyya 14/5 | 12812401 | 124.24 | 128.84 |
| Be'er Toviyya 12/4 | 12712101 | 121.48 | 127.32 |
| Be'er Toviyya 12/5 | 12512101 | 121.88 | 125.98 |
| Be'er Toviyya 13/5 | 12712301 | 123.07 | 127.94 |
| Orot A | 12712401 | 124.94 | 127.60 |
| Orot B | 12812301 | 123.68 | 128.26 |
| Kefar Warburg-A | 12512401 | 124.64 | 125.07 |
| Kefar Warburg-B | 12512303 | 123.97 | 125.35 |
| Kefar Warburg-C | 12412301 | 123.80 | 125.00 |
| Kefar Warburg-D | 12512302 | 123.60 | 125.76 |
| Kefar Warburg-E | 12512201 | 122.07 | 125.36 |
| Kefar Warburg-F | 12512202 | 122.86 | 125.04 |
| Kefar Warburg-12/6 | 12512301 | 123.06 | 125.08 |
| Kefar Warburg-13/6 | 12612401 | 124.48 | 126.07 |
| Avigedor A | 12412501 | 125.80 | 124.52 |
| Avigedor B | 12412401 | 124.43 | 124.26 |
| Avigedor D | 12412502 | 125.38 | 124.48 |
| Magen Eliezer A | 12612502 | 125.80 | 126.03 |
| Magen Eliezer B | 12612501 | 125.06 | 126.40 |
| Kefar Ahim 4 | 12712601 | 126.11 | 127.03 |
| Kefar Ahim 4A | 12712603 | 126.50 | 127.14 |
| Kefar Ahim 4B | 12712501 | 125.95 | 127.00 |
| Kefar Ahim 13/7 | 12612601 | 126.99 | 126.14 |
| Kefar Ahim 6 | 12712602 | 126.36 | 127.96 |
| Kefar Ahim 14/6 | 12812501 | 125.94 | 128.88 |
| Zemorot A | 12412001 | 120.13 | 124.89 |
| Zemorot 4 | 12412003 | 120.28 | 124.88 |
| Zemorot 6 | 12412002 | 120.21 | 124.20 |
| Zemorot 5 | 12512201 | 120.44 | 125.78 |
| Azriqam 3 | 12712001 | 120.81 | 127.66 |
| Azriqam 4 | 12612001 | 120.64 | 126.56 |
| Azriqam T/2 | 12812202 | 122.32 | 128.54 |

$14.7 \times 10^6 \text{ m}^3$. Analysis of exploitation data from the end of the 1950s and beginning of the 1960s (Tahal, unpublished report, 1964) indicates even greater exploitation rates during 1958–1962. In certain wells it reached up to $1.3 \times 10^6 \text{ m}^3 \text{ year}^{-1}$ (Kefar Warburg-A) and $1.9 \times 10^6 \text{ m}^3 \text{ year}^{-1}$ (Be'er Toviyya 6). A calculation of the total pumpage in 1960 (not all the wells were included owing to absence of data) yields an amount of $11.5 \times 10^6 \text{ m}^3 \text{ year}^{-1}$. As a result of the large rates of

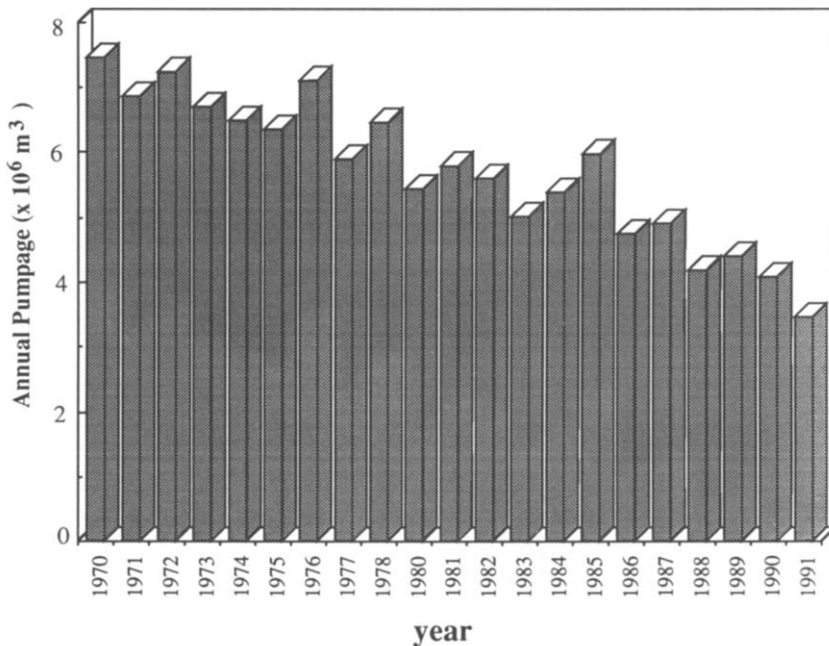


Fig. 4. Variations in the total amounts of pumpage from the wells in the study area over the last 20 years.

groundwater exploitation, an hydrologic depression developed around the Be'er Toviyya and Kefar Warburg area.

4.2. Water-level and chloride data

Hydrological analysis along the NW–SE and east–west cross-sections in the Be'er Toviyya area indicates a piezometric low in the center of the plume area which reached maximum low levels (–5 m) in 1967 (Figs. 5 and 6). Beginning in 1968, the trend of falling levels reversed and the water levels began to rise.

The water level in the Kefar Warburg-A well, located in the southeastern part of the study area, fell even lower, to –7 m (Figs. 5 and 6). However, this trend did not reverse itself, and the water levels remained low, around –6 m, for four more years, until 1971. A maximum low level was also evident in the Avigedor A borehole in the southern margins of the plume in 1971. Only after this year is a change in the trend recorded, and water levels began to rise. The data on chlorinity changes over time (Fig. 5). The NW–SE cross-section indicates that processes of salinization in the southern wells occurred only later.

In the Be'er Toviyya 2 well the increase in salinity beyond the background salinity level of about 200 mg l⁻¹ took place in two stages (Fig. 7): (a) in 1951, when the minimum water level was at an elevation of +9 m, there was a rapid increase in the concentration of chloride to 270 mg l⁻¹, thereafter, in the course of the next 4 years, the concentrations of chloride dropped back to the background level; (b) in 1956

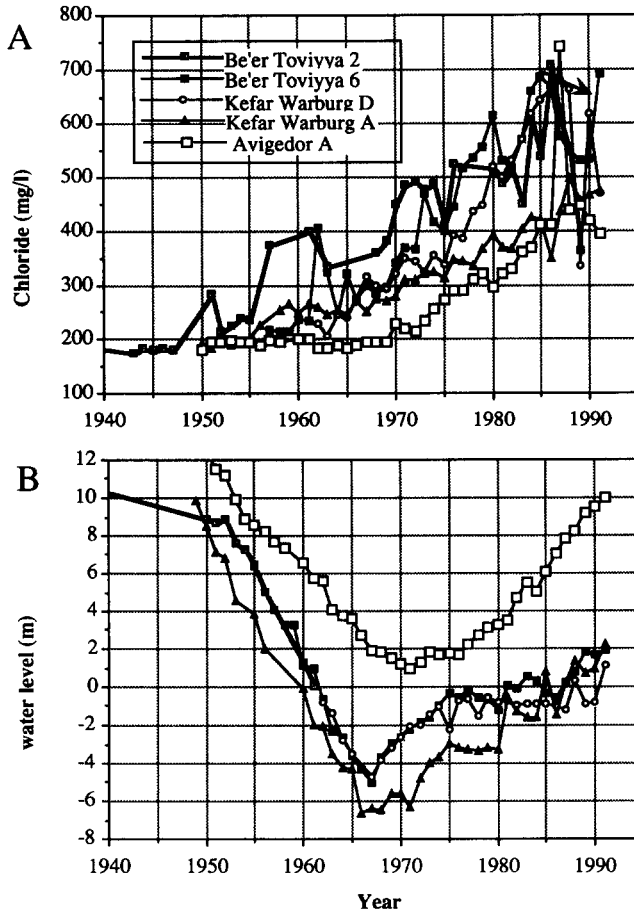


Fig. 5. Variations over time in chloride content (A) and water level (B) in wells along a NW–SE cross-section.

when the minimum water level dropped to an elevation of +5 m, a breakthrough occurred and the chloride concentrations rose to 375 mg l^{-1} . These processes took place while the water levels were falling at a slow rate (from 1932 to 1952) and at a very fast rate (1.1 m year^{-1}) (from 1953 to 1967).

In Be'er Toviyya 4A (Fig. 7), the increase in salinity (the 'breakthrough') took place in the years 1959–1961, during which the water levels dropped from an elevation of +5 m to +2 m. In the Be'er Toviyya 6, the increase in salinity (the breakthrough) took place rapidly during 1962, when the chloride concentrations increased from 270 mg l^{-1} to 400 mg l^{-1} . In Be'er Toviyya 3, the salinization rate was gradual, beginning in 1959, at a rate of $10 \text{ mg l}^{-1} \text{ year}^{-1}$. The moderate increase in salinity in this well differs from the breakthrough events in other wells. In Be'er Toviyya 5, farther east, the increase in salinity occurred suddenly in 1963 (Fig. 7). Within a year the chloride concentrations rose from 225 mg l^{-1} to 410 mg l^{-1} and water levels dropped from 0 to

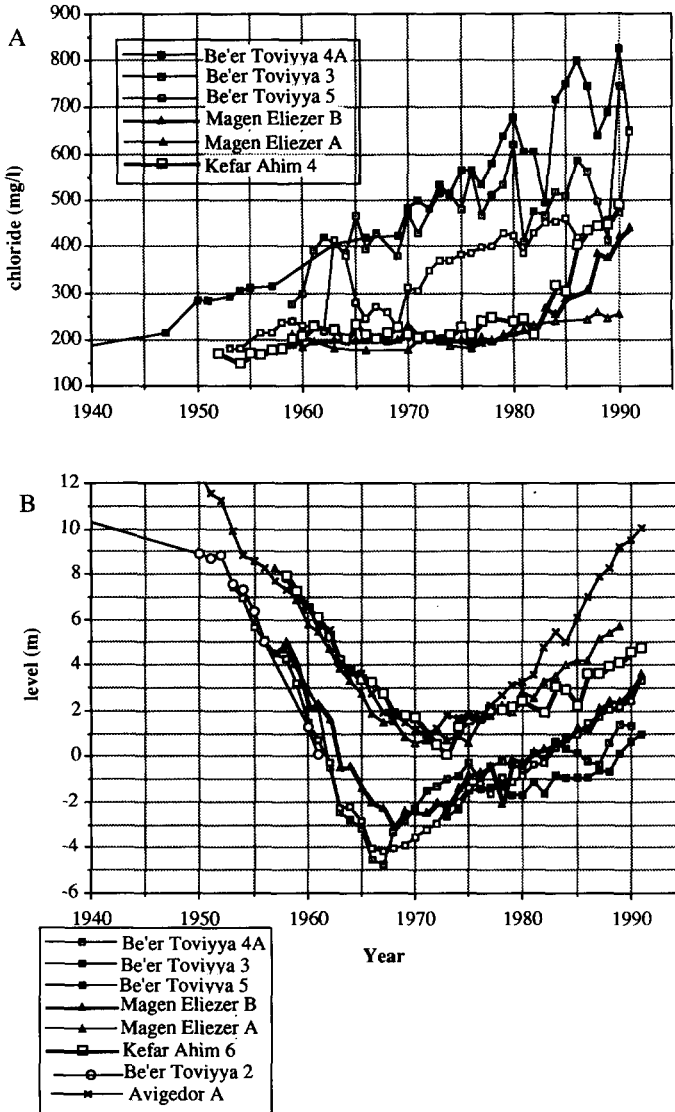


Fig. 6. Variations over time in chloride content (A) and water level (B) in wells along an east-west cross-section.

–2 m. In the following 6 years the chloride content decreased to around 250 mg l^{-1} , and only in 1970 did the salinity begin to increase again.

4.3. Chemical composition of the groundwater

Analysis of previous water well data from the study area (Table 2) indicates that fresh water, unaffected by salinization, had a different chemical composition from

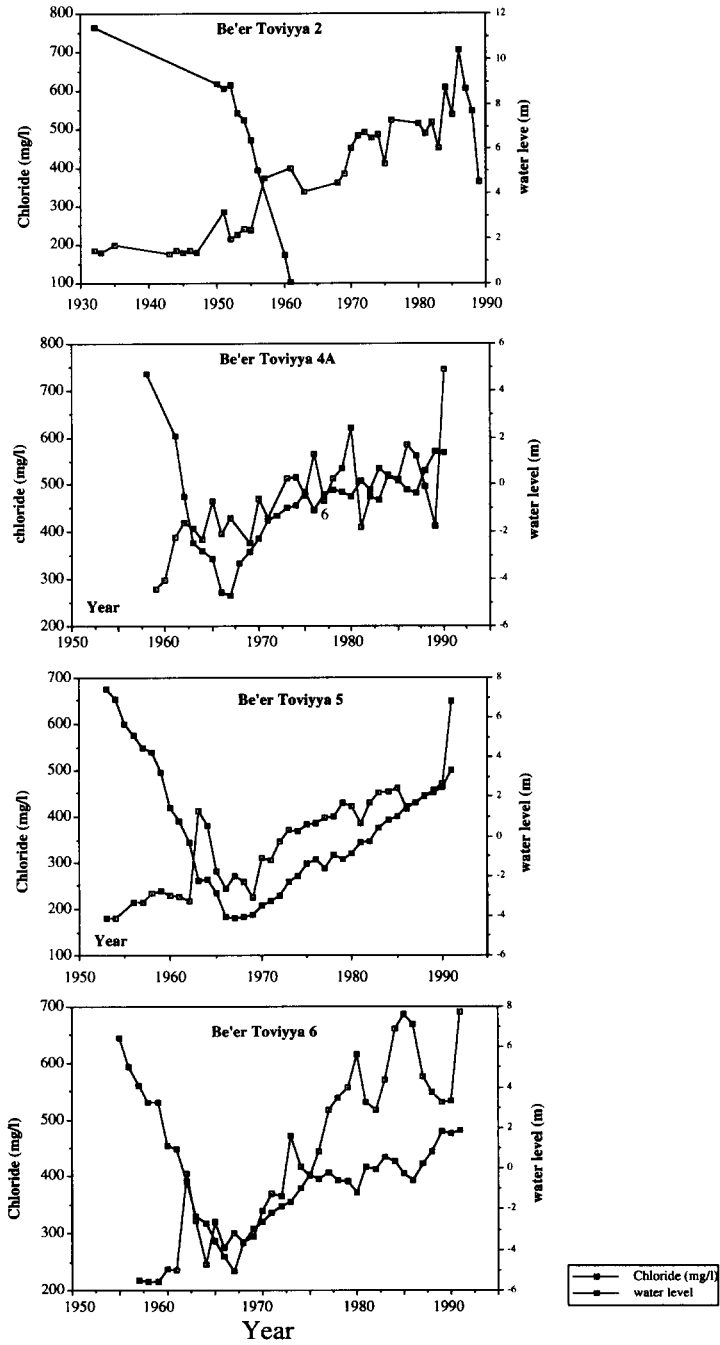


Fig. 7. Changes over time in chlorinity and water level in selected wells.

that of the brackish water. Although defined as fresh water, the chlorinity of groundwater from the Be'er Toviyya area was in the range of 170–280 mg l⁻¹, which was higher than that of groundwater in other places in the Coastal Plain aquifer, e.g. 50–100 mg Cl l⁻¹ (Vengosh and Starinsky, 1991). This water was characterized by Na/Cl ratios of 0.8–1.2 (in molar units) and Ca/Mg ratios ranging between 0.9 and 1.5.

The rise in chloride concentrations in the Be'er Toviyya area was accompanied by a rise in the total dissolved solids (TDS) in the groundwater. The following are the variations of the ion ratios (Table 3; Fig. 8):

Sodium: based the variations of Na/Cl ratios, three water groups are defined: (1) water with salinities lower than 300 mg Cl l⁻¹, characterized by an Na/Cl molar ratio higher than the marine ratio (0.86) and exceeding unity; (2) water with salinities around 300–500 mg Cl l⁻¹ with low Na/Cl ratios, reaching a minimal value of 0.6; (3) the most saline water, with salinities of 600–800 mg Cl l⁻¹, having Na/Cl ratios of around 0.8.

Magnesium: Mg/Cl molar ratios indicate a uniform decrease with the increase in salinity; the ratio in the most saline samples reaches 0.16, which is higher than the marine ratio of 0.095.

Calcium: the Ca/Cl molar ratios indicate notable enrichment of calcium relative to the marine ratio. The increase in salinity is accompanied by a decrease in the Ca/Cl ratio. The Ca/(SO₄ + HCO₃) ratio (*Q* value) rises with increasing salinity and stabilizes at around unity. This ratio is higher than the marine ratio and suggests a Ca-chloride signature of the salinity source.

Potassium: the K/Cl molar ratios decrease with increase of salinity and stabilize around 0.005, which is lower than the marine ratio (0.019).

Sulfate: the SO₄/Cl molar ratios (like Na/Cl ratios) point to three main water groups: (1) water with salinities lower than 300 mg Cl l⁻¹, characterized by high SO₄/Cl ratios; (2) water with salinities of 300–550 mg Cl l⁻¹ with low SO₄/Cl ratios; (3) the most saline water (600–800 mg Cl l⁻¹), characterized by a SO₄/Cl ratio of 0.05, which is close to the marine ratio.

Bromide: The fresh water yielded a wide range of Br/Cl molar ratios whereas the saline water has a ratio of about 1.5×10^{-3} , which is similar to the marine ratio.

Boron: the B/Cl molar ratios (similar to the Na/Cl and SO₄/Cl ratios) suggest three main water groups: (1) water with salinities lower than 250 mg Cl l⁻¹, characterized by a wide range of B/Cl ratios which are usually higher than the marine ratio; (2) water with salinities of 250–550 mg Cl l⁻¹, with low B/Cl ratios around the marine ratio (0.8×10^{-3}) and even lower; (3) the most saline water (600–800 mg Cl l⁻¹), characterized by B/Cl ratios around 2×10^{-3} , which are slightly higher than the marine ratio.

5. Discussion

5.1. The hydrological history of wells in the Be'er Toviyya area

Since the 1970s the most outstanding phenomena in the Be'er Toviyya area are the

Table 2

Chemical data of groundwater in the Be'er Toviyya area (data from the Hydrological Service archives;

| Well | No. | Year | Cl (mg l ⁻¹) | TDS (mg l ⁻¹) | Cl (mequiv l ⁻¹) | SO ₄ (mequiv l ⁻¹) | HCO ₃ (mequiv l ⁻¹) | Na (mequiv l ⁻¹) | K (mequiv l ⁻¹) |
|--------------------|----------|------|-----------------------------|------------------------------|---------------------------------|--|---|---------------------------------|--------------------------------|
| Be'er Toviyya 1 | 12712201 | 1963 | 212.7 | 758.4 | 6.00 | 0.82 | 4.10 | 5.39 | 0.072 |
| Be'er Toviyya 1 | 12712201 | 1973 | 191.0 | 665.0 | 5.40 | 0.80 | 6.00 | 5.00 | 0.07 |
| Be'er Toviyya 2 | 12612201 | 1963 | 336.1 | 1081.6 | 9.48 | 0.93 | 5.59 | 6.85 | 0.084 |
| Be'er Toviyya 2 | 12612201 | 1973 | 478.6 | 1334.7 | 13.50 | 1.40 | 5.60 | 8.40 | 0.079 |
| Be'er Toviyya 3 | 12612403 | 1963 | 404.2 | 1173.3 | 11.40 | 1.20 | 4.90 | 9.13 | 0.079 |
| Be'er Toviyya 3 | 12612403 | 1973 | 514.0 | 1390.8 | 14.50 | 1.30 | 5.60 | 9.00 | 0.080 |
| Be'er Toviyya 4A | 12712302 | 1963 | 404.0 | 1215.3 | 11.40 | 1.37 | 5.15 | 8.93 | 0.079 |
| Be'er Toviyya 4A | 12712302 | 1973 | 514.0 | 1379.0 | 14.50 | 1.20 | 5.39 | 9.00 | 0.080 |
| Be'er Toviyya 5 | 12612402 | 1963 | 231.2 | 910.5 | 6.52 | 0.82 | 5.35 | 5.72 | 0.064 |
| Be'er Toviyya 5 | 12612402 | 1973 | 301.3 | 1055.5 | 8.50 | 0.80 | 5.90 | 5.73 | 0.060 |
| Be'er Toviyya 6 | 12612301 | 1963 | 248.2 | 887.9 | 7.00 | 0.84 | 5.00 | 5.70 | 0.069 |
| Be'er Toviyya 6 | 12612301 | 1973 | 407.7 | 1188.4 | 11.50 | 1.20 | 5.30 | 7.50 | 0.080 |
| Be'er Toviyya 7 | 12612302 | 1967 | 279.0 | 1022.5 | 7.87 | 0.87 | 5.59 | 5.99 | 0.100 |
| Magen Eliezer A | 12612502 | 1963 | 177.3 | 861.4 | 5.00 | 0.85 | 5.80 | 5.12 | 0.059 |
| Magen Eliezer B | 12612501 | 1973 | 195.0 | 923.6 | 5.50 | 0.90 | 6.20 | 4.50 | 0.060 |
| Kefar Warburg-B | 12512303 | 1963 | 276.6 | 986.8 | 7.80 | 0.83 | 5.69 | 6.27 | 0.074 |
| Kefar Warburg-E | 12512201 | 1963 | 202.8 | 841.0 | 5.72 | 0.70 | 5.59 | 5.86 | 0.077 |
| Kefar Warburg-E | 12512201 | 1973 | 237.5 | 917.0 | 6.70 | 0.70 | 5.70 | 6.40 | 0.070 |
| Kefar Warburg-C | 12412301 | 1963 | 191.5 | 854.3 | 5.40 | 0.73 | 5.95 | 6.22 | 0.079 |
| Kefar Warburg-D | 12512302 | 1973 | 358.0 | 957.4 | 10.10 | 0.80 | 4.10 | 7.00 | 0.069 |
| Kefar Warburg-F | 12512202 | 1967 | 243.0 | 892.8 | 6.85 | 0.79 | 5.44 | 6.96 | 0.113 |
| Zemorot A | 12412001 | 1963 | 180.1 | 811.3 | 5.08 | 0.70 | 6.00 | 5.76 | 0.090 |
| Zemorot A | 12412001 | 1973 | 262.3 | 1000.9 | 7.40 | 1.30 | 5.80 | 7.10 | 0.079 |
| Kefar Ahim 4 | 12712601 | 1957 | 164.5 | 464 | 4.64 | 0.70 | 6.30 | 5.28 | |
| Kefar Ahim 4 | 12712601 | 1963 | 219.8 | 929.6 | 6.20 | 0.84 | 6.15 | 5.44 | 0.072 |
| Kefar Ahim 4 | 12712601 | 1963 | 203.5 | 929.5 | 5.74 | 0.92 | 6.30 | 5.25 | 0.064 |
| Kefar Ahim 4 | 12712601 | 1967 | 201.0 | 567 | 5.67 | 0.86 | 6.17 | 4.87 | |
| Kefar Ahim 4 | 12712601 | 1971 | 188.0 | 783.0 | 5.30 | 0.80 | 6.40 | 4.70 | 0.05 |
| Kefar Ahim 4B | 12712501 | 1982 | 212.0 | 933.6 | 5.98 | 1.23 | 6.31 | 5.83 | 0.092 |
| Kefar Ahim 14/6 | 12812501 | 1959 | 245.0 | 691 | 6.91 | 1.36 | | 7.28 | |
| Be'er Toviyya 12/4 | 12712101 | 1963 | 304.9 | 860 | 8.60 | | 2.81 | 8.14 | 0.118 |
| Be'er Toviyya 14/5 | 12812401 | 1959 | 113.5 | 320 | 3.20 | 0.51 | | 3.35 | |
| Be'er Toviyya 14/5 | 12812401 | 1963 | 209.9 | 592 | 5.92 | 0.72 | 6.62 | 6.83 | 0.146 |
| Azriqam 4 | 12612001 | 1967 | 182.0 | 513 | 5.13 | 0.71 | 6.30 | 5.33 | |
| Be'er Toviyya 12/5 | 12512101 | 1958 | 194.3 | 548 | 5.48 | 0.56 | 1.60 | 4.59 | |
| Be'er Toviyya 12/5 | 12512101 | 1963 | 305.0 | 860 | 8.60 | | 2.70 | 8.06 | 0.087 |
| Be'er Toviyya 13/5 | 12712301 | 1963 | 224.0 | 843.8 | 6.32 | 0.91 | 5.30 | 5.46 | 0.164 |
| Avigedor A | 12412501 | 1973 | 216.3 | 989.4 | 6.10 | 1.10 | 6.50 | 5.60 | 0.060 |
| Avigedor B | 12412401 | 1963 | 255.0 | 955.8 | 7.19 | 0.81 | 5.64 | 7.99 | 0.077 |
| Avigedor D | 12412502 | 1973 | 258.8 | 928.8 | 7.30 | 0.70 | 5.30 | 8.40 | 0.060 |
| Avigedor B | 12412401 | 1963 | 198.6 | 898.5 | 5.60 | 0.86 | 5.98 | 5.35 | 0.066 |
| Orot B | 12812301 | 1973 | 258.8 | 1044.3 | 7.30 | 0.88 | 6.30 | 6.80 | 0.060 |

decrease in the volume of pumpage and the concurrent rise in water levels and in salinity. The rise in water levels that commenced at the beginning of the 1970s resulted from the decline in the amount of water exploitation. The rise of piezometric levels, however, did not cause a decline in chloride concentrations, nor did it reduce the expansion of the saline plume. The first appearance of salinity in the area and the primary formation of the Be'er Toviyya plume are related to the drop in water levels. The initial undisturbed water head in the area was more than 10 m above sea-level. Water exploitation in that area began during the 1930s. The rate of pumpage was accelerated in the 1950s. This caused a steep decline in water levels. From the beginning of the 1950s the water level in the Be'er Toviyya 2 well, for example,

1957–1973) Q , ratio of $\text{Ca}/(\text{HCO}_3 + \text{SO}_4)$ in the equivalent units; TDI, total dissolved ions.

| Ca (mequiv l ⁻¹) | Mg (mequiv l ⁻¹) | NO ₃ (mequiv l ⁻¹) | TDI (mequiv l ⁻¹) | Na/Cl (mequiv l ⁻¹) | Q (mequiv l ⁻¹) | SO ₄ /Cl (mol l ⁻¹) | K/Cl (mol l ⁻¹) | CaCl (mol l ⁻¹) | Mg/Cl (mol l ⁻¹) | Ca.Mg (mequiv l ⁻¹) |
|---------------------------------|---------------------------------|--|----------------------------------|------------------------------------|----------------------------------|---|--------------------------------|--------------------------------|---------------------------------|------------------------------------|
| 2.48 | 3.60 | 0.59 | 23.04 | 0.898 | 0.504 | 0.069 | 0.012 | 0.206 | 0.300 | 0.69 |
| 4.40 | 3.40 | 0.84 | 25.91 | 1.122 | 0.628 | 0.198 | 0.075 | 2.027 | 1.027 | 1.974 |
| 5.46 | 4.26 | 0.61 | 33.27 | 0.723 | 0.836 | 0.049 | 0.009 | 0.288 | 0.225 | 1.28 |
| 6.71 | 5.60 | 0.78 | 42.07 | 0.622 | 0.958 | 0.052 | 0.006 | 0.248 | 0.208 | 1.20 |
| 4.87 | 4.26 | 0.81 | 36.65 | 0.800 | 0.800 | 0.053 | 0.007 | 0.214 | 0.187 | 1.14 |
| 6.80 | 5.80 | 0.90 | 43.99 | 0.621 | 0.986 | 0.045 | 0.006 | 0.235 | 0.200 | 1.17 |
| 5.30 | 4.62 | 0.98 | 37.82 | 0.784 | 0.814 | 0.060 | 0.007 | 0.233 | 0.203 | 1.15 |
| 6.60 | 6.00 | 1.02 | 43.80 | 0.621 | 1.002 | 0.041 | 0.006 | 0.228 | 0.207 | 1.10 |
| 4.32 | 3.52 | 0.81 | 27.13 | 0.876 | 0.700 | 0.063 | 0.010 | 0.331 | 0.270 | 1.23 |
| 5.30 | 4.60 | 0.96 | 31.86 | 0.675 | 0.791 | 0.047 | 0.007 | 0.312 | 0.271 | 1.15 |
| 4.04 | 3.64 | 0.58 | 26.86 | 0.814 | 0.692 | 0.060 | 0.010 | 0.289 | 0.260 | 1.11 |
| 5.60 | 5.50 | 0.73 | 37.41 | 0.652 | 0.862 | 0.052 | 0.007 | 0.244 | 0.239 | 1.02 |
| 4.54 | 7.55 | 0.59 | 33.09 | 0.761 | 0.703 | 0.055 | 0.013 | 0.288 | 0.479 | 0.60 |
| 4.04 | 3.26 | 0.80 | 24.91 | 1.023 | 0.608 | 0.085 | 0.012 | 0.404 | 0.326 | 1.24 |
| 5.00 | 3.80 | 0.89 | 26.85 | 0.819 | 0.705 | 0.082 | 0.011 | 0.455 | 0.346 | 1.32 |
| 4.57 | 4.02 | 0.58 | 29.84 | 0.803 | 0.702 | 0.053 | 0.010 | 0.293 | 0.258 | 1.14 |
| 3.34 | 3.12 | 0.34 | 24.75 | 1.025 | 0.530 | 0.061 | 0.013 | 0.292 | 0.273 | 1.07 |
| 3.40 | 3.70 | 0.57 | 27.24 | 0.955 | 0.531 | 0.052 | 0.010 | 0.254 | 0.276 | 0.92 |
| 3.08 | 3.05 | 0.32 | 24.84 | 1.152 | 0.461 | 0.067 | 0.015 | 0.285 | 0.283 | 1.01 |
| 4.00 | 3.90 | 0.32 | 30.29 | 0.693 | 0.816 | 0.040 | 0.007 | 0.198 | 0.193 | 1.03 |
| 3.00 | 3.10 | 0.29 | 26.54 | 1.015 | 0.481 | 0.058 | 0.016 | 0.219 | 0.226 | 0.97 |
| 2.63 | 3.37 | 0.03 | 23.66 | 1.134 | 0.392 | 0.069 | 0.018 | 0.259 | 0.332 | 0.78 |
| 3.50 | 4.40 | 0.52 | 30.10 | 0.960 | 0.493 | 0.088 | 0.011 | 0.237 | 0.298 | 0.80 |
| 4.51 | 3.59 | | 25.02 | 1.138 | 0.644 | 0.076 | | 0.486 | 0.387 | 1.26 |
| 4.65 | 3.55 | 0.49 | 27.38 | 0.877 | 0.665 | 0.068 | 0.012 | 0.375 | 0.286 | 1.31 |
| 4.62 | 3.55 | 0.63 | 27.07 | 0.915 | 0.641 | 0.080 | 0.011 | 0.402 | 0.309 | 1.30 |
| 4.66 | 4.01 | 0.58 | 26.81 | 0.859 | 0.664 | 0.075 | | 0.411 | 0.353 | 1.16 |
| 4.70 | 3.60 | 0.46 | 26.01 | 1.389 | 0.365 | 0.161 | 0.022 | 0.513 | 0.561 | 0.915 |
| 4.79 | 3.37 | 0.05 | 27.66 | 0.975 | 0.636 | 0.103 | 0.015 | 0.401 | 0.282 | 1.42 |
| | | | 15.55 | 1.054 | | 0.099 | | | | |
| 3.12 | | | 22.79 | 0.947 | | | 0.014 | 0.181 | | |
| | | | 7.07 | 1.048 | | 0.080 | | | | |
| 3.52 | 2.80 | | 26.56 | 1.154 | 0.479 | 0.061 | 0.025 | 0.297 | 0.236 | 1.26 |
| 3.80 | 3.66 | 0.64 | 25.57 | 1.038 | 0.542 | 0.069 | | 0.370 | 0.357 | 1.04 |
| | | | 12.23 | 0.838 | | 0.051 | | | | |
| 3.05 | | | 22.50 | 0.937 | | | 0.010 | 0.177 | | |
| 4.08 | 2.92 | 0.07 | 25.21 | 0.864 | 0.658 | 0.072 | 0.026 | 0.323 | 0.231 | 1.40 |
| 4.40 | 4.60 | 0.78 | 29.15 | 0.918 | 0.579 | 0.090 | 0.010 | 0.361 | 0.377 | 0.96 |
| 3.13 | 3.03 | 0.51 | 28.38 | 1.110 | 0.485 | 0.057 | 0.011 | 0.218 | 0.211 | 1.03 |
| 2.75 | 2.50 | 0.52 | 27.53 | 1.151 | 0.459 | 0.048 | 0.008 | 0.188 | 0.171 | 1.10 |
| 4.17 | 3.57 | 0.66 | 26.27 | 0.956 | 0.610 | 0.076 | 0.012 | 0.372 | 0.319 | 1.17 |
| 4.60 | 4.00 | 0.96 | 30.90 | 0.931 | 0.640 | 0.060 | 0.008 | 0.315 | 0.274 | 1.15 |

dropped at a rate of 1.1 m year^{-1} (Fig. 7), coinciding with an increase in salinity from 1956, when the water level was around +5 m. The total drop for 1950 to 1967 was about 15–16 m.

In contrast, the increase in salinity does not coincide with the lowest point of the hydrologic depression in the area. In autumn 1967 the center (–6 m) of the hydrologic depression was located in the southern part of the Kefar Warburg-A well area. However, the salt plume originated about 2 km NW of the depression's center (Fig. 11). This indicates a NW location of the primary source of salinity, in the vicinity of the Be'er Toviyya 2, 3, 4A, 5 and 6 wells (see Fig. 1).

The decline in pumping that commenced at the beginning of the 1970s caused a shallowing of the hydrologic depression until it completely disappeared toward the

Table 3

Chemical data of groundwater and sources of contamination in the Be'er Toviyya area

| Well | No. | Date | Ca (mg l ⁻¹) | Mg (mg l ⁻¹) | Na (mg l ⁻¹) | K (mg l ⁻¹) | Sr (mg l ⁻¹) | Cl (mg l ⁻¹) | SO ₄ (mg l ⁻¹) | HCO ₃ (mg l ⁻¹) | NO ₃ (mg l ⁻¹) |
|------------------------------|----------|---------|-----------------------------|-----------------------------|-----------------------------|----------------------------|-----------------------------|-----------------------------|--|---|--|
| Be'er Toviyya 3 | 12612403 | 10/7/91 | 108 | 79 | 340 | 4 | | 652 | 98 | 207 | 76 |
| Be'er Toviyya 3 | 12612403 | 11/3/92 | 140 | 74 | 379 | 4.5 | 2.04 | 713 | 105 | 322 | 87 |
| Be'er Toviyya 4A | 12712302 | 10/7/91 | 95 | 73 | 310 | 3.8 | | 603 | 88 | 176 | 75 |
| Be'er Toviyya 5 | 12612402 | 10/7/91 | 107 | 79 | 330 | 4 | | 649 | 78 | 260 | 76 |
| Be'er Toviyya 6 | 12612301 | 10/7/91 | 92 | 78 | 350 | 4 | | 698 | 105 | 151 | 71 |
| Be'er Toviyya 7 | 12612302 | 16/7/91 | 102 | 65 | 250 | 3.8 | | 533 | 55 | 260 | 27 |
| Be'er Toviyya 8 | 12612101 | 10/7/91 | 35 | 36 | 207 | 3.2 | | 308 | 48 | 220 | 40 |
| Avigedor A | 12412501 | 10/7/91 | 54 | 66 | 186 | 3 | | 393 | 48 | 158 | 38 |
| Avigedor A | 12412501 | 11/3/92 | 128 | 54 | 173 | 3.6 | 1.7 | 348 | 58 | 361 | 52 |
| Avigedor D | 12412502 | 10/7/91 | 56 | 41 | 243 | 2.8 | | 336 | 48 | 298 | 32 |
| Kefar Warburg-A | 12512401 | 9/7/91 | 66 | 65 | 236 | 3.2 | | 475 | 55 | 168 | 43 |
| Kefar Warburg-A | 12512401 | 11/3/92 | 124 | 79 | 236 | 3.8 | 1.93 | 476 | 66 | 339 | 55 |
| Kefar Warburg-D | 12512302 | 9/7/91 | 88 | 64 | 236 | 3.2 | | 470 | 63 | 224 | 43 |
| Kefar Warburg-E | 12512201 | 9/7/91 | 68 | 53 | 228 | 4 | | 356 | 50 | 307 | 39 |
| Magen Eliezer B | 12612501 | 10/7/91 | 93 | 67 | 175 | 2.8 | | 440 | 53 | 161 | 43 |
| Magen Eliezer B | 12612501 | 11/3/92 | 111 | 62 | 176 | 3.2 | 1.86 | 404 | 54 | 349 | 56 |
| Orot B | 12812301 | 10/7/91 | 95 | 56 | 186 | 2.7 | | 379 | 46 | 254 | 35 |
| <i>Anthropogenic sources</i> | | | | | | | | | | | |
| Nahal HaEla (Pecker plant) | | 11/3/92 | 147 | 48 | 123 | 5 | 1.23 | 449 | 66 | 93 | 61 |
| Nahal HaEla (Felicia plant) | | 11/3/92 | 128 | 46 | 127 | 6 | 1.21 | 282 | 74 | 334 | 52 |
| Salt pond | | 11/3/92 | 82 | 61 | 8650 | 93 | 1.21 | 13078 | 200 | 161 | 145 |
| Felicia waste effluent | | 11/3/92 | 176 | 48 | 1950 | | 0.77 | 1544 | 2773 | 17 | |

RE, reaction error, i.e. $(\Sigma\text{cations} - \Sigma\text{anions})/(\Sigma\text{cations} + \Sigma\text{anions})$ in equivalent units; Q , ratio of

end of the 1980s. This shallowing led to equalization of the water levels, whereby the water levels in the saline areas in the center of the plume equilibrated with those in its margins. We suggest that this led to spatial expansion of the saline plume.

The appearance and times of salinity breakthroughs in the study area can be seen in Figs. 5–7. The salinity was first apparent in the Be'er Toviyya 2, 3, and 4A wells at the end of the 1950s, and in the Be'er Toviyya 6 and 5 wells at the beginning of the 1960s. The salinization trend and buildup of the salt plume was in a SE direction, as a result of continuous water gradients in that direction, during the years 1971–1981.

The evolution of the salt plume can be reconstructed by following the migration of chloride concentrations of about 400 mg l⁻¹ over time and space (Fig. 9): the occurrence of 400 mg l⁻¹ of chloride first appeared in the late 1950s and early 1960s in wells Be'er Toviyya 2, 3, 4 A, 5 and 6. During the 1970s the plume spread mainly SE. During the 1980s the SE trend of the plume's expansion continued, but the plume also expanded towards the east, north and SW. At the same time, the absolute salinity increased, and reached a concentration of 800 mg Cl l⁻¹ at the beginning of the 1990s (Figs. 9 and 10). During the 1970s the rate of the plume's SE expansion is estimated at about 50 m year⁻¹; in the 1980s it more than quadrupled to around 200 m year⁻¹, and locally up to 670 m year⁻¹. The rate of the plume's expansion, as noted above, is related to the groundwater gradients between the center of the saline plume and its margins. Once the piezometric levels of the saline area within the center of the salt plume exceeded those of its margin, the wells situated along the plume margins were contaminated within a period of 1 year. As water exploitation decreased, the water levels rosed until the hydrological depression diminished towards the end of the

| Br (mg l ⁻¹) | B (mg l ⁻¹) | Ca (mequiv l ⁻¹) | Mg (mequiv l ⁻¹) | Na (mequiv l ⁻¹) | K (mequiv l ⁻¹) | Sr (mequiv l ⁻¹) | Cl (mequiv l ⁻¹) | SO ₄ (mequiv l ⁻¹) | HCO ₃ (mequiv l ⁻¹) | NO ₃ (mequiv l ⁻¹) |
|-----------------------------|----------------------------|---------------------------------|---------------------------------|---------------------------------|--------------------------------|---------------------------------|---------------------------------|--|---|--|
| 2.4 | 0.364 | 5.39 | 6.50 | 14.79 | 0.10 | | 18.39 | 2.03 | 3.39 | 1.23 |
| | 0.501 | 6.99 | 6.09 | 16.49 | 0.12 | 0.05 | 20.11 | 2.19 | 5.28 | 1.40 |
| 2.5 | 0.338 | 4.74 | 6.01 | 13.48 | 0.10 | | 17.01 | 1.82 | 2.89 | 1.21 |
| 1.8 | 0.355 | 5.34 | 6.50 | 14.35 | 0.10 | | 18.31 | 1.61 | 4.26 | 1.23 |
| 2 | 0.389 | 4.59 | 6.42 | 15.22 | 0.10 | | 19.69 | 2.19 | 2.48 | 1.15 |
| 1.6 | 0.307 | 5.09 | 5.35 | 10.87 | 0.10 | | 15.03 | 1.15 | 4.26 | 0.44 |
| 0.2 | | 1.75 | 2.96 | 9.00 | 0.08 | | 8.69 | 0.99 | 3.61 | 0.65 |
| 0.6 | | 2.70 | 5.43 | 8.09 | 0.08 | | 11.09 | 0.99 | 2.59 | 0.61 |
| | 0.284 | 6.39 | 4.44 | 7.53 | 0.09 | 0.04 | 9.82 | 1.21 | 5.92 | 0.84 |
| 2.1 | | 2.80 | 3.37 | 10.57 | 0.07 | | 9.48 | 0.99 | 4.89 | 0.52 |
| 1.1 | | 3.30 | 5.35 | 10.27 | 0.08 | | 13.40 | 1.15 | 2.75 | 0.69 |
| | 0.437 | 6.19 | 6.50 | 10.27 | 0.10 | 0.04 | 13.43 | 1.37 | 5.56 | 0.89 |
| 1.1 | | 4.39 | 5.27 | 10.27 | 0.08 | | 13.26 | 1.30 | 3.67 | 0.69 |
| 1 | | 3.40 | 4.36 | 9.92 | 0.10 | | 10.04 | 1.04 | 5.03 | 0.63 |
| 1.1 | | 4.64 | 5.51 | 7.61 | 0.07 | | 12.41 | 1.09 | 2.64 | 0.69 |
| | 0.291 | 5.54 | 5.10 | 7.66 | 0.08 | 0.04 | 11.40 | 1.12 | 5.72 | 0.90 |
| 1.1 | | 4.74 | 4.61 | 8.09 | 0.07 | | 10.69 | 0.96 | 4.16 | 0.56 |
| | 0.237 | 7.34 | 3.95 | 5.35 | 0.13 | 0.03 | 12.66 | 1.37 | 1.52 | 0.98 |
| | 0.260 | 6.39 | 3.79 | 5.52 | 0.15 | 0.03 | 7.95 | 1.54 | 5.48 | 0.84 |
| | 2.244 | 4.09 | 5.02 | 376.27 | 2.38 | 0.03 | 368.88 | 4.16 | 2.64 | 2.34 |
| | 0.994 | 8.79 | 3.95 | 84.82 | 0.00 | 0.02 | 43.55 | 57.74 | 0.27 | 0.00 |

Ca/(HCO₃ + SO₄) in equivalent units; M, molar.

(continued on next page)

1980s. The changes in water heads allowed the expansion of the saline plume. In conclusion, two phenomena occurred: (a) a rise in the salinity of the groundwater in the area of the initial salinity onset; (b) spatial expansion of the plume as a result of reduction of water exploitation and changes in water levels.

5.2. Focal points of anthropogenic contamination

Two foci of contamination were discerned in the area; salt ponds and effluents from the Qiryat Malakhi industrial area (Fig. 1). Both constitute potential dangers of groundwater contamination and salinization in the Be'er Toviyya area. The two foci have different chemical signatures that can serve as indicators for sources of contaminants in the aquifer.

The salt ponds in the SE margins of the plume (Fig. 1) drain saline effluents from plants in the Qiryat Malakhi area. These ponds are new; they were built during the mid-1980s as a measure to overcome leakage from the old salt ponds, which were located 300 m to the NE. As a result of leakage of the old salt ponds, increasing moisture and damage were reported in the neighboring garages. No specific monitoring of the spread of the saline water was done, however. As evaporite salts were deposited in the evaporation ponds, the chemical composition of the brines in the ponds could have been developed as follows:

(A) intensified evaporation in summer caused deposition of salts, mainly halite. Accordingly, the predominant ions in the residual brines were chloride and

Table 3 Continued

| Br (mequiv l ⁻¹) | B (mequiv l ⁻¹) | RE (%) | Na/Cl (M) | Q (mequiv l ⁻¹) | Mg/Cl (M) | Ca/Mg (M) | CaCl (M) | K/Cl (M) | SO ₄ Cl (M) | Br/Cl (M) | B/Cl (M) |
|---------------------------------|--------------------------------|-----------|--------------|--------------------------------|--------------|--------------|-------------|-------------|---------------------------|--------------|-------------|
| 0.03 | 0.03 | 3.4 | 0.804 | 0.994 | 0.177 | 0.830 | 0.147 | 0.006 | 0.055 | 1.63E - 03 | 1.83E - 03 |
| | 0.05 | 1.2 | 0.820 | 0.937 | 0.151 | 1.148 | 0.174 | 0.006 | 0.054 | | 2.31E - 03 |
| 0.03 | 0.03 | 3.0 | 0.793 | 1.008 | 0.177 | 0.790 | 0.139 | 0.006 | 0.054 | 1.84E - 03 | 1.84E - 03 |
| 0.02 | 0.03 | 1.7 | 0.784 | 0.909 | 0.178 | 0.822 | 0.146 | 0.006 | 0.044 | 1.23E - 03 | 1.80E - 03 |
| 0.03 | 0.04 | 1.6 | 0.773 | 0.986 | 0.163 | 0.716 | 0.117 | 0.005 | 0.056 | 1.27E - 03 | 1.83E - 03 |
| 0.02 | 0.03 | 1.3 | 0.723 | 0.942 | 0.178 | 0.952 | 0.169 | 0.006 | 0.038 | 1.33E - 03 | 1.89E - 03 |
| 0.00 | | -0.5 | 1.036 | 0.380 | 0.170 | 0.590 | 0.101 | 0.009 | 0.057 | 2.88E - 04 | |
| 0.01 | | 3.2 | 0.730 | 0.753 | 0.245 | 0.497 | 0.122 | 0.007 | 0.045 | 6.77E - 04 | |
| | 0.03 | 1.9 | 0.767 | 0.897 | 0.226 | 1.439 | 0.326 | 0.009 | 0.062 | | 2.68E - 03 |
| 0.03 | | 2.9 | 1.115 | 0.476 | 0.178 | 0.829 | 0.148 | 0.008 | 0.052 | 2.77E - 03 | |
| 0.01 | | 2.7 | 0.766 | 0.845 | 0.200 | 0.616 | 0.123 | 0.006 | 0.043 | 1.03E - 03 | |
| | 0.04 | 4.1 | 0.765 | 0.893 | 0.242 | 0.953 | 0.231 | 0.007 | 0.051 | | 3.01E - 03 |
| 0.01 | | 2.8 | 0.774 | 0.884 | 0.199 | 0.834 | 0.166 | 0.006 | 0.049 | 1.04E - 03 | |
| 0.01 | | 3.0 | 0.988 | 0.559 | 0.217 | 0.779 | 0.169 | 0.010 | 0.052 | 1.25E - 03 | |
| 0.01 | | 2.9 | 0.613 | 1.244 | 0.222 | 0.842 | 0.187 | 0.006 | 0.044 | 1.11E - 03 | |
| | 0.03 | -2.0 | 0.672 | 0.810 | 0.224 | 1.087 | 0.243 | 0.007 | 0.049 | | 2.36E - 03 |
| 0.01 | | 3.4 | 0.757 | 0.926 | 0.216 | 1.030 | 0.222 | 0.006 | 0.045 | 1.29E - 03 | |
| | 0.02 | 0.7 | 0.422 | 2.532 | 0.156 | 1.859 | 0.290 | 0.010 | 0.054 | | 1.73E - 03 |
| | 0.02 | 0.1 | 0.695 | 0.911 | 0.238 | 1.689 | 0.402 | 0.019 | 0.097 | | 3.03E - 03 |
| | 0.21 | 1.3 | 1.020 | 0.602 | 0.007 | 0.816 | 0.006 | 0.006 | 0.006 | | 3.63E - 04 |
| | 0.09 | -2.0 | 1.948 | 0.152 | 0.045 | 2.225 | 0.101 | | 0.663 | | 2.11E - 03 |

magnesium. The Na/Cl ratio in the residual brine was dependent on the Na/Cl ratio of the effluents that were discharged into the ponds. A Na/Cl ratio of the effluents of less than unity would result in much lower Na/Cl ratio in the residual brine (following removal of both chloride and sodium ions). Contrarily, a Na/Cl ratio in the effluent source of more than unity would have resulted in a much higher Na/Cl ratio in the residual brine. Therefore, it can only be estimated that during the summer seasons leakage of the salt ponds would result in penetration of hypersaline brines with an Mg-chloride composition.

(B) Rainfall and dilution of the pond waters during winter resulted in dissolution of the salts in the ponds. The chemical composition of a single sample from the salt pond taken in winter 1992 (Table 3) indicates a chlorinity of 13 000 mg l⁻¹, a predominance of sodium and chloride ions, and a Na/Cl ratio of about unity. This composition points to dissolution of salts, mainly of halite. Thus, during winter, flooding and leakage of the ponds would result in penetration of saline waters having an Na-chloride composition.

The Qiryat Malakhi industrial area in the north of the study area (coordinates 1266/1275) includes the Felicia and Pecker plants. During the past decades these two plants discharged their effluents directly into Nahal HaEla. The Felicia plant began operating in 1958 and the Pecker plant in 1974. The Felicia plant discharged effluents with high contents of chlorides (up to 13 000 mg l⁻¹) and chromates (as high as 2000 mg l⁻¹) directly into Nahal HaEla. Data on the effluents of the Pecker steel plant are even more critical: chloride content of 118 000 mg l⁻¹; zinc, 43 600 mg l⁻¹; lead, 65 mg l⁻¹; chromium, 21 mg l⁻¹; aluminium, 100 mg l⁻¹. These data (E. Lyninberg, personal communication, 1992) are partial and do not constitute a full and

continuous record of the quality of effluents in the industrial area of Qiryat Malakhi. However, the data show that for recent decades Nahal HaEla drained effluents with high contents of poisonous metals whose concentrations were higher by several orders of magnitudes than those prescribed by the regulations of the Ministry of Health for drinking water. However, it is still unknown whether these contaminants penetrated into the regional groundwater system.

The chemical composition of a single sample taken from the effluents of the Felicia plant (Table 3) shows a chloride concentration of $13\,000\text{ mg l}^{-1}$ and predominance of sodium, chloride and sulfate ions. Though it is only one single sample, the Na/Cl (more than unity) and high SO_4/Cl ratios of the effluents are different from those of the saline groundwaters in the area (Table 3).

Moreover, the establishment of Felicia and Pecker plants (in 1958 and 1974, respectively) took place only after the rise in salinity, which began in 1956. In addition, the initial focus of the saline plume started around wells Be'er Toviyya 2 and 3, whereas other wells located closer to Nahal HaEla were contaminated only in later stages. Thus salinization of groundwater in the Be'er Toviyya area was not the result of surficial anthropogenic activity.

The occurrence and distribution of nitrate concentrations in groundwater of the area might be indicative of the migration of contaminants. The data on nitrates are only partial and are only from 1980. At the beginning of the 1980s, a plume of high nitrate concentration (over 50 mg l^{-1}) appeared in the northern part of the study area (Fig. 12) at well Orot A, downslope of the water-level gradient relative to the Qiryat Malakhi contamination focal point. During the 1980s the nitrate plume spread southward, although only traces were still evident in the center of the area during summer 1990. Apparently, the source of high nitrate concentrations came from the northern part of the area and could be related to discharge of surplus urban sewage and industrial effluents. The effluents of the Felicia plant are also enriched in ammonia (up to 900 mg l^{-1}), which could oxidize to nitrate in the subsurface. If, indeed, the anomalous source of nitrates in the groundwater is related to industrial effluents, which are enriched in poisonous metals and have been flowing into Nahal HaEla in recent decades, groundwaters in that area could be contaminated by these dangerous industrial materials. In conclusion, although it seems that the saline plume was not a direct result of anthropogenic contamination, the high nitrate concentrations suggests superposition of anthropogenic contamination on the natural salinization process.

5.3. *The chemical characteristics of the salinity sources*

The variations in the chemical compositions with increasing salinity of the Be'er Toviyya groundwater may reflect contribution from two saline sources. Both are characterized by low Mg/Cl and K/Cl ratios, a $\text{Ca}/(\text{SO}_4 + \text{HCO}_3)$ ratio around unity (i.e. Ca-chloride sources), and a marine Br/Cl ratio. The two sources differ in their salinities, Na/Cl, SO_4/Cl , and B/Cl ratios. These differences may imply that the two saline sources were formed by different mechanisms:

- (1) salinity source A is characterized by a chloride concentration of $300\text{--}500\text{ mg l}^{-1}$

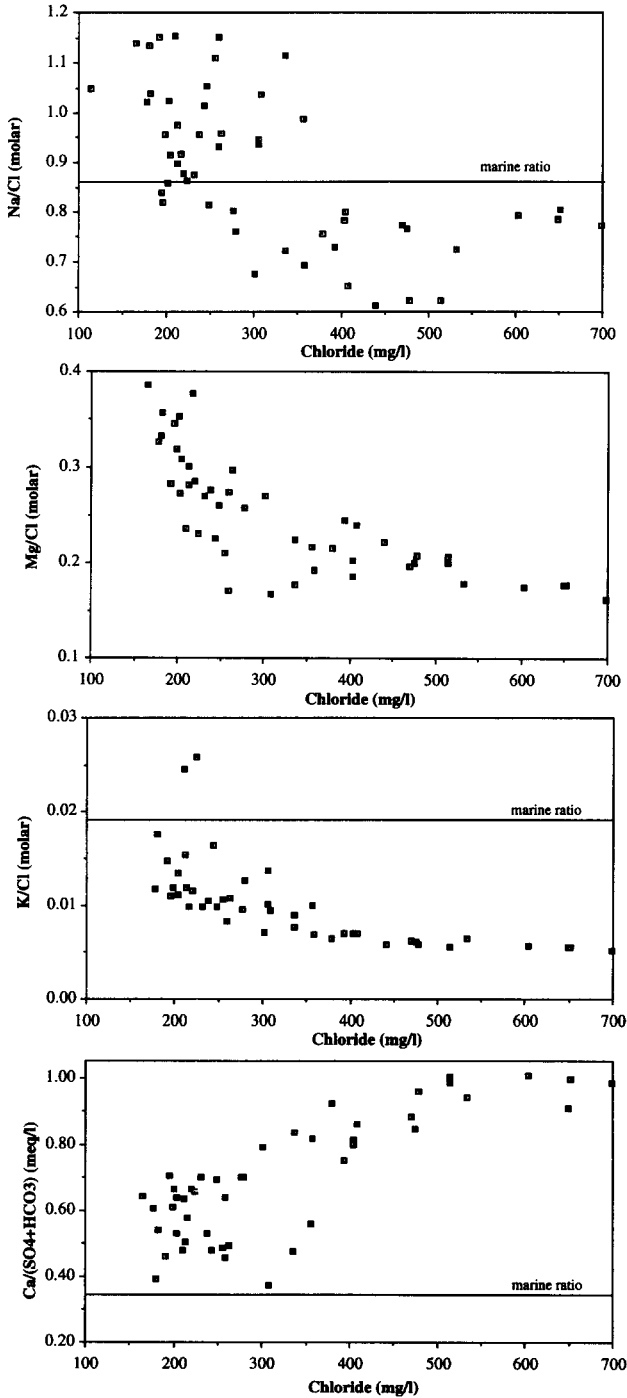


Fig. 8. Chloride concentrations vs. various ion ratios in groundwater.

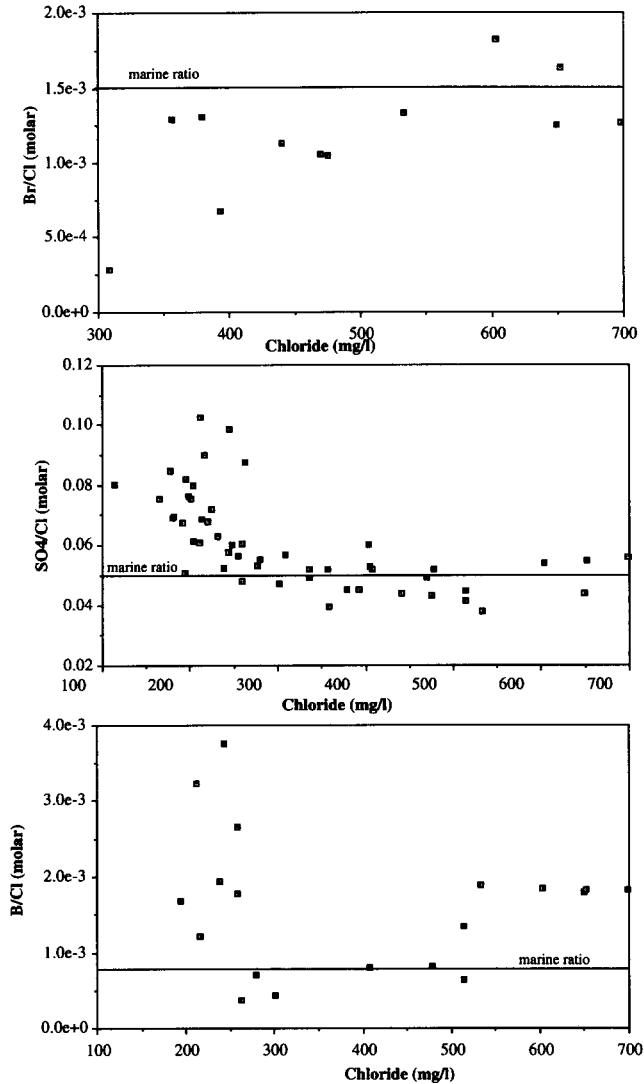


Fig. 8. Continued.

and relatively low Na/Cl, SO₄/Cl and B/Cl ratios (Fig. 8). These low ratios and Ca/(SO₄ + HCO₃) > 1 are typical of Ca-chloride brines that have undergone evaporation processes, deposition of evaporite minerals (gypsum and halite), dolomitization and sulfate reduction.

(2) Salinity source B is characterized by a relatively high chloride concentration ranging between 500 and 800 mg l⁻¹ and relatively high Na/Cl (0.8), SO₄/Cl (0.05) and B/Cl ratios (2×10^{-3} ; Fig. 8). The SO₄/Cl ratios are close to the marine ratio, whereas the B/Cl ratios are slightly higher. These saline waters are also characterized

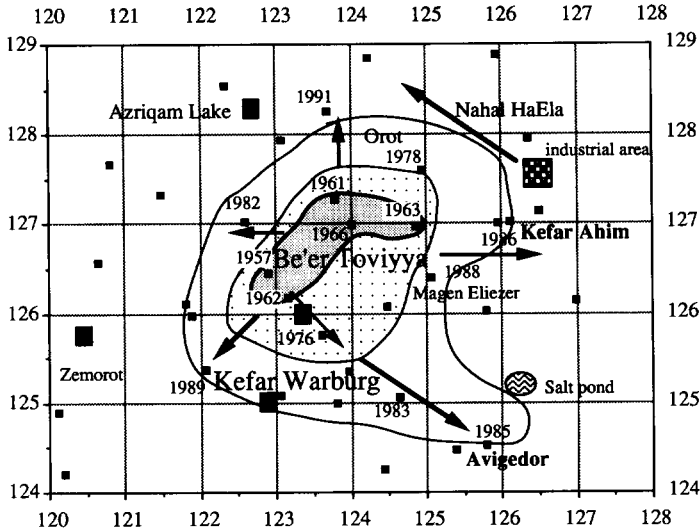


Fig. 9. Location map of first occurrences of chloride concentration of about 400 mg l^{-1} or above in groundwater in the Be'er Toviyya area.

by a $\text{Ca}/(\text{SO}_4 + \text{HCO}_3)$ ratio around unity and a marine Br/Cl ratio. This chemical composition is similar to that of seawater that has undergone partial evaporation but did not reach halite saturation, as indicated by the marine Na/Cl ratio. The apparent evaporation process occurred in a lagoon environment, and the evaporated seawater participated in dolomitization processes and thus became Ca-chloride with a marine Na/Cl ratio.

These hydrochemical features of the two saline sources are similar to the two types of saline water found in other locations at the Coastal Plain aquifer. Source A resembles Ca-chloride saline water (salinity of $1000\text{--}33\,000 \text{ mg Cl l}^{-1}$) with low Na/Cl ratios found along the interface zone at the western margin of the Coastal Plain aquifer (Vengosh et al., 1991). Source B resembles brines (up to $60\,000 \text{ mg Cl l}^{-1}$) with a marine Na/Cl ratio found in the deep subaquifer of the Gaza Strip (Fink, 1970; Vengosh et al., 1991). In light of the geochemical resemblance of the saline groundwater from the interface zone to the brines in the Bardawil Lagoon area, Vengosh et al. (1991) suggested that the Ca-chloride waters were formed in a coastal-sabkha environment, cut off from the main water body of the lagoon, whereas the Gaza brines derived from the lagoon waters themselves. Accordingly, it is not surprising to find in the Be'er Toviyya area the two water bodies together: remnants of both the lagoonal and the sabkha waters which were entrapped in close proximity.

The chemical composition of Source B is also similar to deep brines from the underlying Mavki'im (Upper Miocene) and Heletz (Lower Cretaceous) formations, at about $500\text{--}1500 \text{ m}$ depth in the southern Coastal Plain of Israel (Starinsky, 1974; Starinsky et al., 1983; Bein and Sofer, 1987). These 'C-brines' (Starinsky, 1974) are characterized by a salinity of up to $55\,000 \text{ mg Cl l}^{-1}$, marine Na/Cl ratio, Ca-chloride

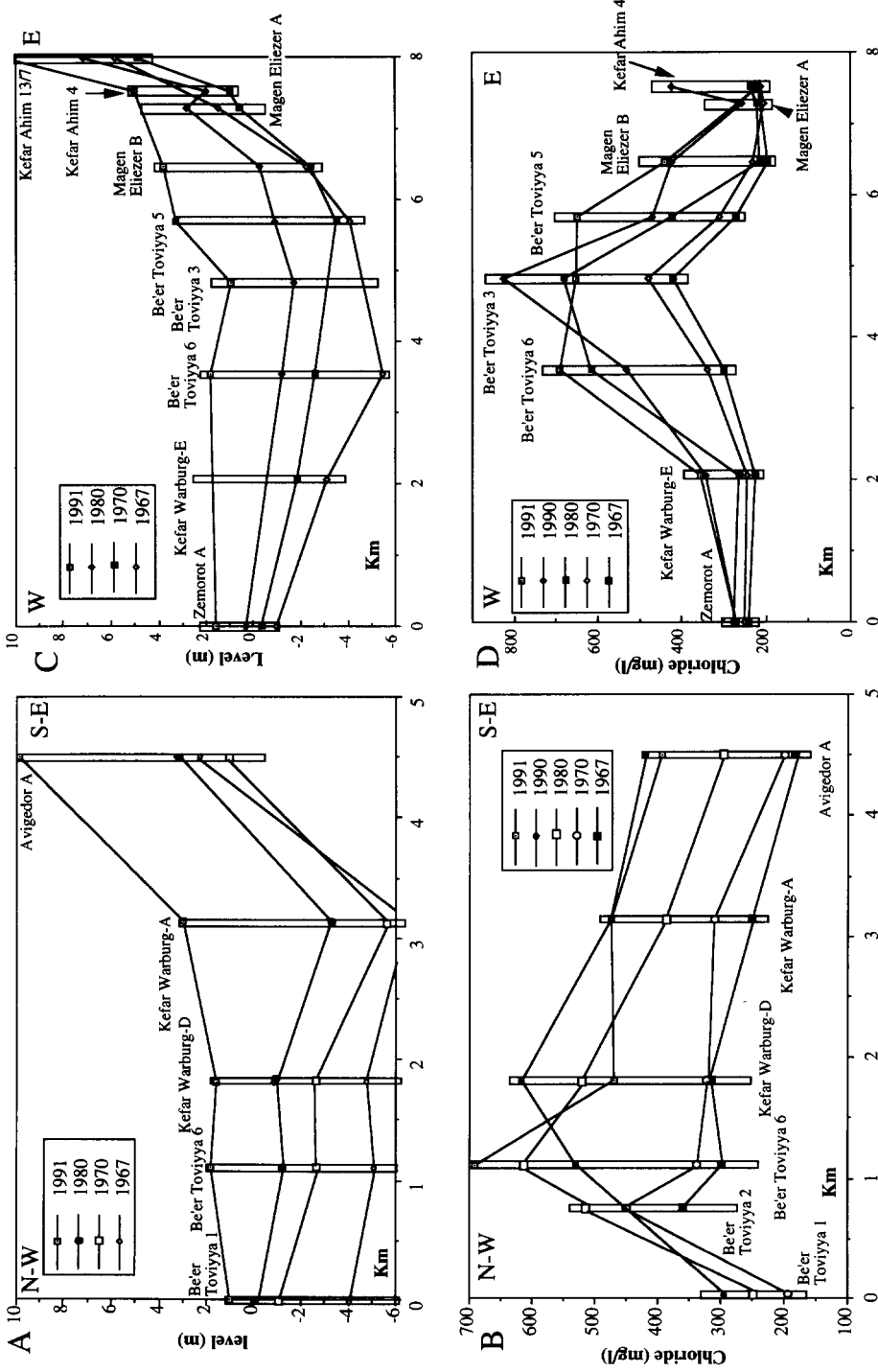


Fig. 10. (A) Schematic NW–SE cross-sections of water levels during 1967–1991; (B) schematic NW–SE cross-sections of chloride contents during 1967–1991; (C) schematic west–east cross-sections of water levels during 1967–1991; (D) schematic west–east cross-sections of chloride contents during 1967–1991.

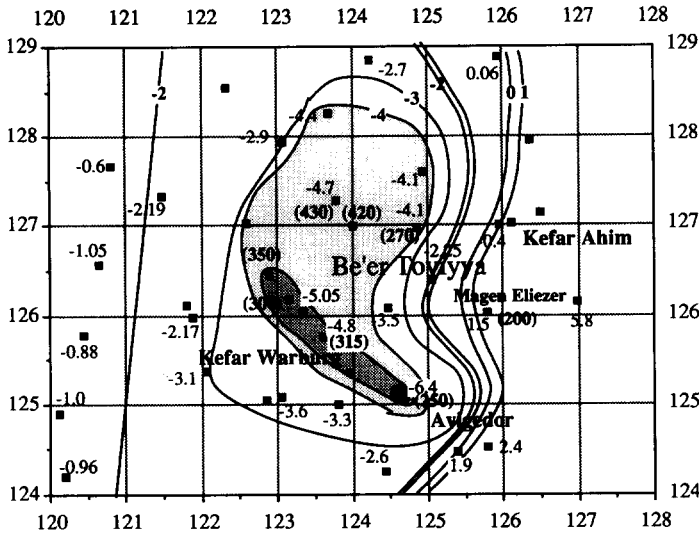


Fig. 11. Map of water-level elevations, autumn 1967, in relation to location of salinity onset and the high-salinity area of the plume (chloride concentrations written in bold within parenthesis).

signature, marine Br/Cl ratios, and high (greater than seawater) B/Cl ratios (Starinsky, 1974; Starinsky et al., 1983; Vengosh, 1990). Thus Source B potentially can be derived from the underlying deep C-brines.

From the chemical differences between the two salinity sources, their chlorinities can be estimated on the basis of the variations of Na/Cl ratios. Assuming that the concentrations of sodium (Na_{mix}) and chloride (Cl_{mix}) of the brackish water around 500 mg Cl l^{-1} are products of a mixture between fresh (Na_{fresh} and Cl_{fresh}) and saline water, and a conservative behavior of Cl and Na, the mixture equation for chloride will be

$$\text{Cl}_{\text{fresh}}F + \text{Cl}_{\text{salt}}(1 - F) = \text{Cl}_{\text{mix}} \quad (1)$$

where Cl_{salt} is the chloride concentration (in millimol units) of the saline end-member and F is the freshwater fraction in the mixture. Similarly, the mixture equation for sodium will be

$$\text{Na}_{\text{fresh}}F + \text{Na}_{\text{salt}}(1 - F) = \text{Na}_{\text{mix}} \quad (2)$$

The Na/Cl ratios of End-member A can be estimated in light of the Na/Cl, B/Cl, SO_4/Cl ratios and $\text{Ca}/(\text{SO}_4 + \text{HCO}_3) > 1$ that resemble Ca-chloride waters from the interface zone (Vengosh et al., 1991). Such waters are characterized by low Na/Cl ratios in the range of 0.3–0.8. Hence both mixture equations can be solved by assuming that $X = \text{Na}/\text{Cl}_{\text{salt}}$, where X is the estimated range of Na/Cl ratios in saline end-member water. The solution of the two equations sets the value of the lower limit of the Na/Cl salt ratio (0.45). The calculated chloride concentrations of End-member A (with an estimated Na/Cl ratio in the range 0.45–0.60) range between 570 and 2700 mg l^{-1} . Above the chlorinity of 500 mg l^{-1} a significant change in the chemical composition is evident: the Na/Cl, B/Cl and SO_4/Cl ratios increase. This change in

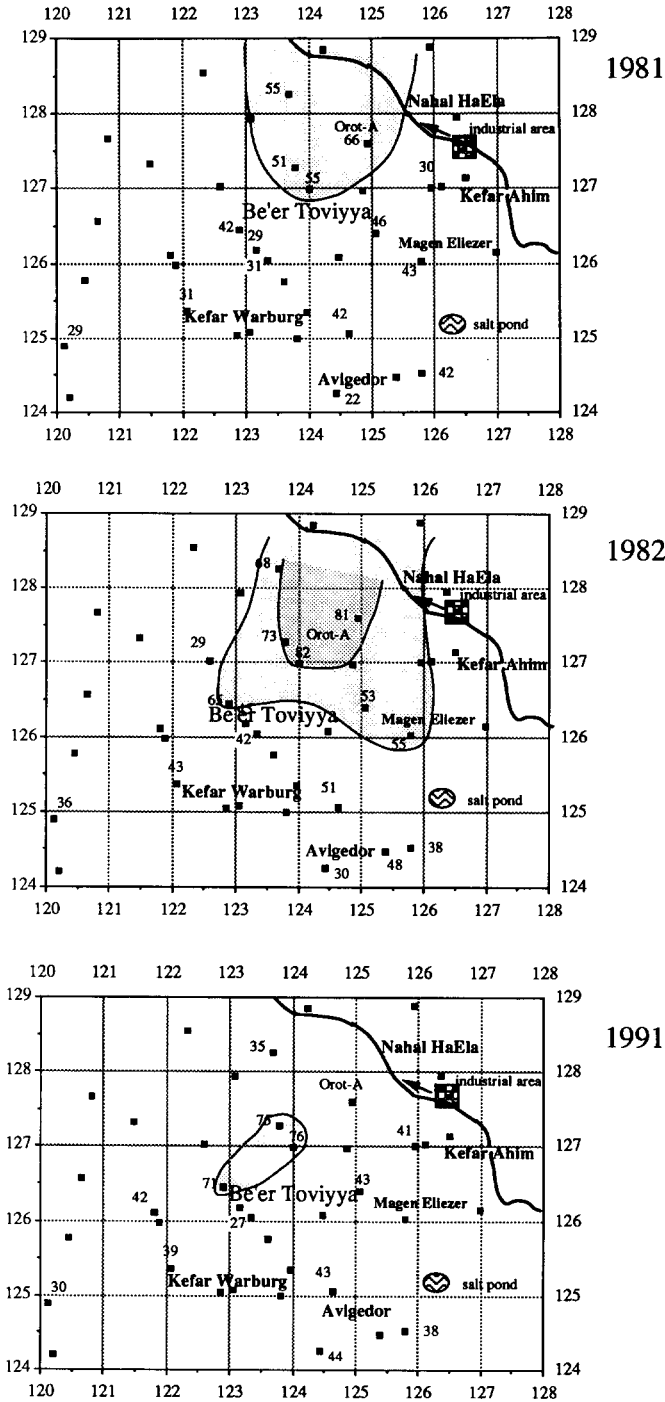


Fig. 12. Maps showing nitrate concentrations (mg l^{-1}) in 1981, 1982 and 1991.

composition is explained by contribution from End-member B. According to the chemical relations this end-member could be seawater or evaporated seawater below the degree of halite saturation. According to this assumption, the brackish waters (more than 500 g l^{-1}) in Be'er Toviyya are products of water contaminated by both End-member A (up to 500 mg l^{-1}) and saline water of End-member B. The rise in Na/Cl ratios with salinity indicates that the source of salinity B has a higher Na/Cl ratio than 0.6. Also, in this case, both mixture equations can be applied under the assumption that $\text{Na/Cl}_{\text{salt}}$ is greater than 0.6. The solution of the two equations sets the value of the upper limit of $\text{Na/Cl}_{\text{salt}}$ at 1.2. The calculated chloride concentrations of End-member B (with an estimated Na/Cl ratio in the range 0.8–1.2) range between 800 and 3350 mg l^{-1} .

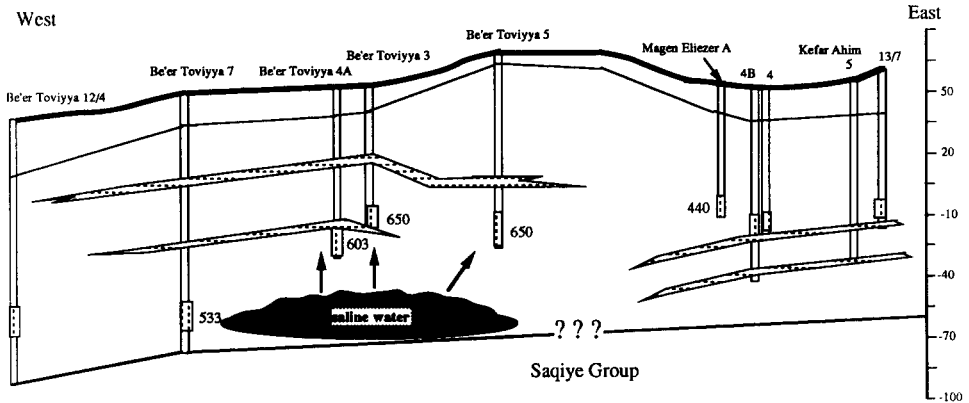
5.4. Possible sources of the saline water and estimations of its volume

The salt plume in Be'er Toviyya formed initially as a point source in the field of wells Be'er Toviyya 2, 3, 4A, 5 and 6 (see salinity evolution events; Fig. 9) which does not coincide with the piezometric lowest level of the hydrologic depression in Kefar Warburg-A area. Long-term data show that salinity in the initial point source continued to be higher relative to other wells in the vicinity, thus testifying to continued contribution of salts from the same point source. Despite the perennial trend of increasing salinity, changes in salinity were not continuous, but occurred as sharp fluctuations in the chloride concentrations (Figs. 5 and 6). These changes might indicate heterogeneity of several water bodies in the aquifer. No connection was found between these chloride fluctuations and time of sampling, e.g. winter vs. summer.

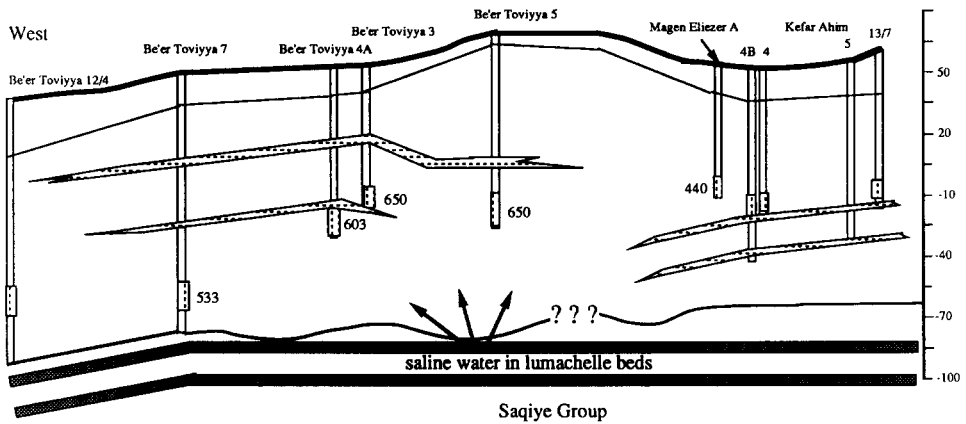
As surficial anthropogenic sources for the formation of the salt plume are ruled out (see above), saline water could have been derived from two possible sources (Fig. 13): (1) saline waters from the bottom of the aquifer, with no hydrological separation from the main body of fresh water above it; (2) saline waters from outside of the aquifer, such as shallow permeable layers within the underlying Saqiye Group.

In some locations of the Coastal Plain aquifer, saline water bodies were found at the bottom of the aquifer. These saline waters escaped flushing possibly owing to density stratification and also because of a decrease in hydraulic conductivity and relatively high clay content at the base of the aquifer. According to this model, the rapid drop in water levels that started during the 1950s following intensified pumping, caused a drop in the freshwater head, a break in the hydraulic equilibrium and an upward flow of the saline water to the pumping field (Fig. 13). In well Be'er Toviyya 2 the breakthrough of salinity occurred in 1956 when the autumn level of the water table was +5 m. However, during 1932 the water level was at +12 m (see Fig. 7), i.e. a drop of 7 m.

The other source of salinization could be saline water from outside the aquifer, possibly from the conductive layers in the Saqiye Group. The Yafo Formation, which constitutes the base of the coastal aquifer, is built of a thick section of marine clay. Oil and gas boreholes, drilled in the 1980s, revealed lumachelle and sandstone beds at shallow depths within the upper section of Yafo Formation. These conductive layers contain high-pressure gas and saline water of $2000\text{--}35\,000 \text{ mg Cl l}^{-1}$ (Muravinsky and



model a - saline water at the bottom of the coastal plain aquifer;



model b - saline water in shallow sandstone layers in the Saqiye Group.

Fig. 13. Models for saline water sources of the Be'er Toviyya plume. Model A: saline water at the bottom of the Coastal Plain aquifer; Model B: saline water in shallow sandstone layers in the Saqiye Group.

Grossman, 1982; Dikenstein and Croker, 1983; Fligelman and Shomrony, 1983). In some locations the hamra-clays beds, separating the Coastal Plain aquifer from the saline water-bearing layers with high hydraulic conductivity, are only a few meters thick (e.g. Ga'ash, 5 m; Fligelman and Shomrony, 1983). In the Ashdod area at 368–371 m depth, under conditions of equilibrium, the gas pressure reached values of 530 psig, i.e. the head of the saline water entrapped in these layers was around 370 m. Such conductive layers, containing highly pressurized saline water, were also found in the Bene Darom area (Dikenstein and Croker, 1983) near Ashdod, which is about 5 km from the Be'er Toviyya study area (Fig. 1).

Vinokurov et al. (1991) reported deep narrow depressions built of clays and sandstone at the top of the Yafu Formation of the Saqiye Group in the Petah Tiqwa area. These depressions, which contain saline water, might also constitute a

source of saline water to the Coastal Plain aquifer. Discontinuity of the separating clay beds, which are only few meters thick in several places, or the changing hydraulic conductivity within the Saqiye Group, could have resulted in leakage of saline waters that are under pressure and high heads (up to 370 m) relative to the groundwater in the coastal aquifer (in the given area, about 70 m). If a hydraulic connection exists between the Coastal Plain aquifer and these conductive layers, saline water could possibly have flowed to the aquifer. A scenario such as this means that an 'endless' source of saline water may penetrate into the aquifer by underground seepage as a result of a drop in the freshwater head and a break in the previous hydraulic equilibrium. This model is strengthened by the fact that salinization in the Be'er Toviyya area continued during the last 25 years despite the continuous rise of water levels owing to the reduction in pumpage.

If the source of the saline water of the Be'er Toviyya plume is indeed outside the aquifer, the amount of chloride, the volume of saline water penetrating into the aquifer, and the rate of supply of saline water can be estimated by mass balance calculations. The water balance follows the equation

$$\Delta S = R + A + V_1 - P \quad (3)$$

where ΔS is the storage change, R is the natural recharge and replenishment, A is the artificial water recharge, V_1 is the net lateral flow to the study area, and P is pumpage. Integrating the differences in water levels between 1967 (the lowest level of the hydrologic depression; Fig. 11) and 1990 (its disappearance) yields a total storage change (ΔS) of $+40 \times 10^6 \text{ m}^3$. The natural replenishment (about 150 mm year^{-1}) in the area of the saline plume ($30 \times 10^6 \text{ m}^2$) contributed $4.5 \times 10^6 \text{ m}^3 \text{ year}^{-1}$, i.e. $103 \times 10^6 \text{ m}^3$ (R) during the interval 1967–1990. The artificial recharge (A) is summarized from the actual records as $15 \times 10^6 \text{ m}^3$, whereas the total pumpage (P) for wells in the study area during the period 1967–1990 was $138 \times 10^6 \text{ m}^3$. Applying these figures in Eq. (3) yields $60 \times 10^6 \text{ m}^3$ for the net lateral flow (V_1) into the study area.

The chloride mass balance follows the equation

$$\Delta \text{Cl} = \sum \text{Cl}_{\text{input}} - \text{Cl}_{\text{output}} \quad (4)$$

where ΔCl is the difference in chloride storage, $\sum \text{Cl}_{\text{input}}$ is the total input, composed of replenishment and lateral flow, and $\text{Cl}_{\text{output}}$ is pumpage (P). By assuming (1) vertical uniformity in chloride concentrations, and (2) horizontal uniformity between isochloride lines, which are always equal to the lower values, and by integration of isochloride line above 400 mg l^{-1} , we estimate the chloride amount in the saline plume area at 1990 (Fig. 2). The estimation for 1990 yields an amount of about $80 \times 10^6 \text{ kg}$ chloride, whereas integration of the saline plume area for 1967 (using 300 mg l^{-1} isochloride lines) yields only $6 \times 10^6 \text{ kg}$ chloride. Thus ΔCl (Eq. (4)) is estimated as $74 \times 10^6 \text{ kg}$ chloride. Integration of the chlorides in pumpage during the interval 1967–1990 yields $43 \times 10^6 \text{ kg}$, thus $\sum \text{Cl}_{\text{input}} = 117 \times 10^6 \text{ kg}$ chloride ($43 + 74$, Eq. (4)).

Assuming that chloride content in precipitation is negligible, the chloride input is

composed of

$$\sum \text{Cl}_{\text{input}} = (VC)_{\text{saline}} + (VC)_{\text{background}} + (VC)_{\text{A}} \quad (5)$$

where $(VC)_{\text{saline}}$ is the saline flow, $(VC)_{\text{background}}$ is the lateral aquifer background flow, and $(VC)_{\text{A}}$ is the artificial recharge. The chloride concentration in artificial recharge is about 250 mg l^{-1} , thus the integrated chloride mass during 1967–1990 is $4 \times 10^6 \text{ kg}$. The total lateral flow (V_1) (Eq. (4)) is estimated as $60 \times 10^6 \text{ m}^3$, and is composed of

$$V_1 = V_{\text{saline}} + V_{\text{background}} \quad (6)$$

Applying the estimated concentration of a possible saline end-member of $2700 \text{ mg Cl l}^{-1}$, which is the upper concentration limit based on Na/Cl ratio variations (Eqs. (1) and (2)) and other chemical parameters (see above), we use both Eqs. (5) and (6) to calculate the volume of the saline input, V_{saline} . The result is a saline inflow of $40.4 \times 10^6 \text{ m}^3$. Next, we estimate the lower limit of the concentration of the saline end-member. For that purpose we assume that the entire net lateral flow originated from the saline source, i.e. $V_{\text{background}} = 0$ and $V_{\text{saline}} = 60 \times 10^6 \text{ m}^3$. Upon application of Eqs. (5) and (6) for this case we obtain a chloride concentration of about 1900 mg l^{-1} for the lower concentration limit of the saline end-member. Consequently, these estimations show that a relatively large volume of saline water flowed into the hydrological depression of Be'er Toviyya during the period 1967–1990 with an estimated flux of $(1.8\text{--}2.6) \times 10^6 \text{ m}^3 \text{ year}^{-1}$, which is about half of the natural replenishment.

In conclusion, the restoration of the water levels during the interval 1967–1990 was a result of the water balance in which input (replenishment, artificial recharge, lateral and saline flows; about $178 \times 10^6 \text{ m}^3$) exceeded output (pumpage, $138 \times 10^6 \text{ m}^3$). The saline water composes a relatively large fraction of the input component, i.e. $(40\text{--}60) \times 10^6 \text{ m}^3$. The salinity of the end-member saline source is estimated as being $1900\text{--}2700 \text{ mg Cl l}^{-1}$.

6. Summary and conclusions

(1) In the Be'er Toviyya area a salt plume has been developed that reached a salinity of up to 800 mg Cl l^{-1} . Several wells located in the area of the plume record exponential increase in salinity. Assuming that the observed trends in the Be'er Toviyya area will continue, a chloride concentration of up to 1000 mg l^{-1} is predicted for the year 2000.

(2) Formation of the salt plume in the Be'er Toviyya area took place in two main stages: (a) onset of saline water in the central area between the wells Be'er Toviyya 2, 3, 4A, 5 and 6; (b) spatial spread of the saline water towards the SE, east and SW. The spread of salinity is the result of a combination of points of recharge of the Azriqam area in the NW and the recharge wells in the west (Zemorot, Be'er Toviyya 7, and Kefar Warburg-F), and differential pumping in time and space that consequently developed groundwater gradients to the SW.

(3) Intensified pumping beginning in the 1950s caused a decline in water levels and the formation of a hydrologic depression in the Be'er Toviyya area. This depression reached its deepest level during 1967. The configuration of the depression resulted in the development of the water-level gradient to the SW and flow of saline water in that direction. Decrease in pumpage that began in the early 1970s resulted in a restoration of the water levels and a shallowing of the hydrologic depression, until it completely disappeared by the late 1980s. Equalization of the water levels within and on the margin of the salt plume also accounted for the spatial spread of the salt plume. As a result of the restoration of the water levels, the water tables in several wells reached, or came close, to their original levels before pumping (Avigedor A, Fig. 5). However, despite the rise in water levels, the trend of increasing salinity in the initial area continued.

(4) Although there are several focal points of surficial anthropogenic contamination in the Be'er Toviyya area (the industrial area of Qiryat Malakhi and the salt ponds) which have high salinization potentials, the Be'er Toviyya salt plume is not a result of contamination from a surficial anthropogenic source. This is clear from the time of the plume's formation: the first onset of salinity occurred in Be'er Toviyya 2 in 1956, whereas the Felicia and Pecker plants were founded in 1958 and 1974, respectively. The center of salinity that formed in the middle and late 1950s retained high salinity relative to the surrounding area during all the years the plume developed, which indicates continuation of salinization from sources that have no connection to the source of anthropogenic contamination.

(5) The salt plume in the Be'er Toviyya area formed following the drop in water levels and flow of saline water bodies from underlying layers. The chemical composition of the saline waters indicates the presence of two saline water bodies of Ca-chloride composition: (a) saline water with low Na/Cl ratios (0.45–0.60) and estimated chloride concentrations of 570–2700 mg l⁻¹; (b) saline water with higher (more than 0.60) Na/Cl ratios and possible chloride concentrations of up to 3350 mg l⁻¹.

(6) High contents of nitrates in the groundwater in the area, above 50 mg l⁻¹, imply infiltration and penetration of contaminants in the Nahal HaEla area. The combination of high concentration of poisonous metals in the industrial effluents in Nahal HaEla and the presence of a hydrologic depression for about 30 years could have caused flow and concentration of contaminants in the aquifer in western and southern directions relative to the focal point of the contamination of Nahal HaEla (Fig. 1). The dumping of industrial effluents containing high levels of contaminants into Nahal HaEla calls for establishment of a monitoring system for groundwater in the vicinity.

(7) Decrease in water exploitation and shallowing of the hydrologic depression did not prevent the continued rise in salinity and expansion of the saline plume. Thus it seems that the salinization process was irreversible, at least within the time period 1956–1992. Water and chloride mass balance calculations show that the volume of the saline water that penetrated into the aquifer between 1967 and 1990 was about $(40.4–60) \times 10^6$ m³, with an estimated flux of $(1.8–2.6) \times 10^6$ m³ year⁻¹. These values indicating that a large amount of saline water penetrated into the aquifer as a result of the deep drawdown in water level (15 m) and formation of the hydrological

depression. It also implies derivation from a large saline source. The saline water comprises a relatively large fraction of the input component, i.e. $(40\text{--}60) \times 10^6 \text{ m}^3$. The chlorinity of the saline source is estimated as 1900 mg l^{-1} (based on mass balance calculations) to 2700 mg l^{-1} (based on chemical variations).

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