

Early-indicator signals of groundwater contamination: the case of seawater encroachment

A. J. Melloul · L. C. Goldenberg

Abstract An early indication of groundwater contamination occurs when pollutant concentrations start to fluctuate and exceed background values of ambient fresh groundwater. An analysis of a characteristic situation of this type uses data from Israel's coastal phreatic granular aquifer. The pollutant is generally seawater, and the contamination process involves replacement of freshwater by encroaching sea- or other saltwater, a process augmented by human activity. The contamination process involves three stages: (1) groundwater composition remains relatively stable with small salinity content; (2) small salinity changes are perceptible with reversible fluctuations; and (3) salinity concentration increases at a sharply higher rate. The second stage is a useful early-indicator signal of contamination. Early-indicator signals of groundwater pollutant concentrations involve "minor" fluctuations in water chemistry at the advent of the contamination process. The intensity and magnitude of such a salinization/pollution process at any given location depends upon lithologic matrix, aquifer heterogeneity, and resultant flow domain characteristics, as well as contaminant properties. If such "signs" are detected at a sufficiently early stage, appropriate management steps may be taken to rectify further seawater and/or saltwater encroachment.

Key words Salt- and seawater encroachment · Groundwater contamination · Early-indicator signals

Introduction

Groundwater quality may deteriorate as a result of: (1) degradation of intrinsic chemical, biological or physical parameters of groundwater at a rate exceeding the self-cleaning capacity of the aquifer (Goldenberg and Melloul 1994a, c), and/or (2) excessive pumpage of water from the aquifer, creating undesirable groundwater gradients and balances (Gehrels and others 1994; Graham and Neff 1994; Melloul and Collin 1994).

Attention to early signals of groundwater quality degradation can prove an important contribution to the prevention of adverse environmental/hydrological changes. These early-indicator signals can be used to detect the point at which hydrological equilibrium is lost between recharge and discharge components, thus indicating groundwater safe yield (Todd 1961; Chow 1964). Today, due to the growth of industry, housing, construction, mining, and other changes in land-use, highly unfavorable and often irregular changes in the water properties have been detected. Thus, the inevitable result of inadequate water management poses a barrier to further economic development as well as a threat to a city's population (Pociask-Karteczka 1994). A better situation would now be the case, had appropriate attention been paid during the first stages of groundwater degradation. Sources of groundwater degradation may include infiltration of contaminated surface water, disposal sites of solid or liquid waste materials, agricultural activities (US EPA 1990; Melloul and Collin 1992), and deposition of airborne pollutants on the soil and their migration to groundwater (Melloul and Goldenberg 1994), etc. However, amongst the major potential sources of contamination to coastal aquifers are the sea and brines, the latter located mostly in the deeper horizons of the aquifer (Fink 1970; Melloul and Bachmat 1975; Vengosh and others 1991; Vengosh and Starinsky 1993). When these intrude into the aquifer they may contaminate significant quantities of fresh groundwater (US EPA 1990; Melloul and Gilad 1993). Near the sea, the driving force of saltwater intrusion is commonly excess pumpage, which lowers the local freshwater table, changing the direction and magnitude of groundwater gradients.

Before the onset of seawater (or other saline water) intrusion, certain initial signs of contamination appear, the early detection of which might prove complicated owing

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to uncertainty resulting from insufficient data and information regarding hydrogeologic parameters (Latinopoulos and others 1994). For one thing, pollution/salinization does not occur equally along the entire vertical water profile. Furthermore, movement of contaminants depends upon aquifer matrix properties, which are generally quite heterogeneous. For example, a situation might arise where the matrix comprises "interbedded red sandstones, silt stone, mud stone, anhydrite and halite etc." (Nance 1988). In such a situation, even with a constant gradient, velocities of contaminants would vary, and processes like filtration, sorption, capillary action, plant-root uptake, ion exchange, dispersion, oxidation dilution, and microbial degradation would contribute to heterogeneity in the pollution process. A reliable tracer would therefore be required in order to characterize the contamination/salinization process.

This study intends to analyze the early signals before the onset of sea- or other saltwater intrusion into a coastal groundwater system, where there are hydrological management constraints to maintaining stable groundwater pumpage (Hydrological Service Situation Report 1994). The objectives of the study are: (1) to present a methodology to describe early-indicator signal phenomena of sea or brines water intrusion, (2) to propose explanations for processes which result in early-indicator signals, and (3) to make a case for groundwater quality monitoring even when concentration of contaminants in groundwater is not substantial.

Hydrological description of the study area

The coastal aquifer of Israel extends from the town of Hadera in the north to the Gaza Strip in the south, paralleling the coast for about 120 km (Fig. 1). The aquifer, a major component of Israel's national water supply system, is composed of Pleistocene dune sands and calcareous sandstones with intercalated clay layers. Within approximately 5 km from the sea, these clay layers separate the aquifer into three major sub-aquifers (Fig. 2). The phreatic portion of this aquifer is located beneath a highly populated area and supplies significant quantities of water for agriculture, domestic, and industrial purposes. The aquifer is recharged mainly by precipitation, artificial recharge from the National Water Carrier, and by water percolating from agricultural, industrial, and domestic sources (Melloul 1988; Melloul and Bibas 1990). During the past decade, until the 1991/1992 rainy year, the coastal aquifer was overexploited. Between 1970 and 1991 the hydrological situation involved a regime of relatively high pumpage that saw a decline in groundwater levels of about 5–10 cm per year, followed by increases in groundwater salinity (Hydrological Service Situation Report 1994). In proximity to the sea shore this has resulted in sea- (Fig. 3) and other saltwater intrusion (Vengosh and

others 1991). Inland it has resulted in a significant increase in pollution (Goldenberg and Melloul 1994b,c; Melloul and Collin 1994).

This study concentrates upon the upper sub-aquifer, in an area where seawater intrusion has already significantly affected groundwater quality (Melloul and Gilad 1993). The coastal aquifer of Israel has been used here to characterize the general pattern to be expected upon the onset of groundwater quality deterioration within any phreatic coastal aquifer. Data were collected by the Hydrological Service of Israel (IHS) between 1952 and 1992.

Methodology

This study investigates the pattern of groundwater salinity changes in an area contaminated by seawater intrusion. The study involved the following concepts:

1. Data utilized include a period of time preceding the onset of significant anthropogenic degradation of the aquifer water.
2. Water samples were taken from the aquifer at wells located less than 1500 m from the sea shore in an area in which probability of seawater intrusion is highest. In this case the area is limited by the relevant IHS field measurements taken between the coast and the saltwater/freshwater interface involving a network of observation wells (of around 5-cm diameter) in which electrical resistivity and water-table levels were used to assess seawater intrusion (Fig. 3). Most of the chemical samples were taken only from pumping wells located in this vicinity and with at least 20 years of observation.
3. The major chemical parameters chosen were chloride and nitrate, both routinely reported in the coastal aquifer data base of the Hydrological Service (Hydrological Service Situation Report 1994). Chloride was selected because it is a nonreactive constituent with the aquifer matrix material. Thus, the behavior of the Cl^- ion can be used as a good tracer to follow the processes of groundwater contamination/salinization, especially during the initial stages to monitor seawater/freshwater interface movement (White 1977; Konikow and Rodriguez 1993). Nitrates, although not a tracer parameter, are utilized to identify pollution in wells. Trends and levels of nitrates, when compared to those of chloride data, can be quite helpful in checking and emphasizing the existence of pollution in seawater intrusion areas. Other parameters such as sodium, calcium, sulfate, and chemical ratios like $r\text{Na}/r\text{Cl}$ and $r\text{Ca}/r\text{SO}_4$ ("r" meaning that chemical parameters are in milliequivalent per liter) taken from different places, are also helpful in characterizing and explaining seawater intrusion processes (Bachmat and Chetboun 1974; Yechieli and others 1992).

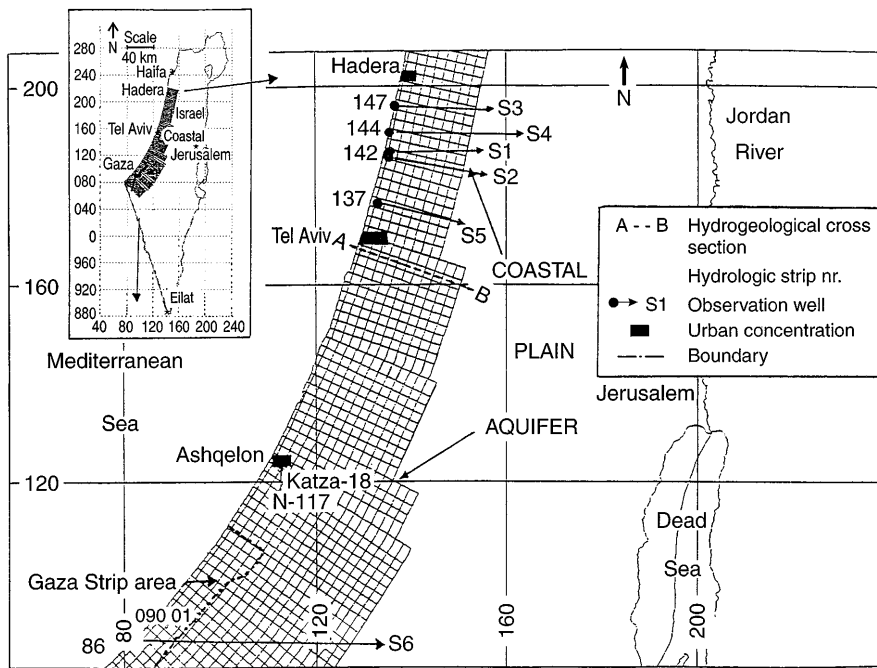


Fig. 1
Location map: the study area

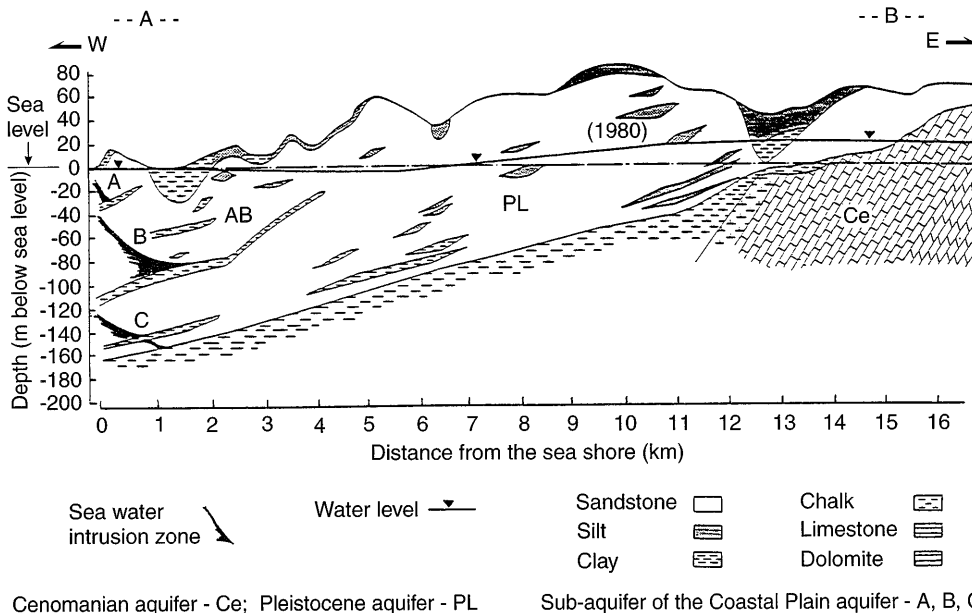


Fig. 2
A typical hydrogeological cross-section (A-B) through the coastal aquifer of Israel

4. Certain laboratory results have been utilized in this study for better understanding the problem of aquifer heterogeneity and its relationship with groundwater quality changes. The research involved laboratory assessment of field data dealing with seawater intrusion into groundwater, changes in permeability, and the formation of preferred pathways in a porous media aquifer. This has been enabled by characterization of chemical and physical parameters (electrical conductivity (EC), Cl^- and SO_4^{2-} concentrations, and grain size distribution and mineralogic composition of the samples) of interstitial water profiles, taken from cores in the Israeli coastal aquifer. These cores representing

the geology of the unsaturated zone of the coastal aquifer at various depths were analyzed as explained by Goldenberg and others (1986, 1993) and Goldenberg and Melloul (1994a).

5. Analyses include chlorographs of different wells to describe the different stages in the process of groundwater contamination.

6. Finally, analyses involved a combination of chlorographs, nitrographs, chemical ratios, and laboratory results to explain and assess a conceptual model of early-indicator signals of sea- and/or saltwater intrusion in wells located at different distances from each other in Israel and abroad.

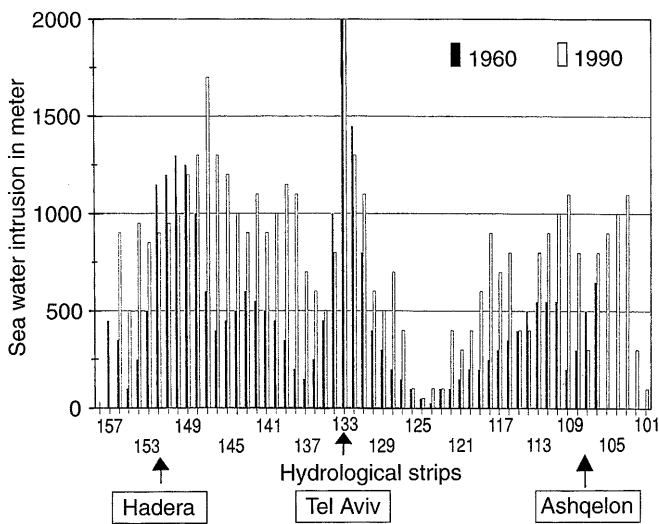


Fig. 3 Sea water intrusion into the coastal aquifer of Israel in 1960 and 1990

Findings

The key findings of this study are presented in Fig. 4–7 and in Table 1, illustrating the behavior of Cl⁻ and other chemical parameters. Figures 8 and 9 illustrate the problem of heterogeneity, into saline intrusion and changes in poros media aquifer matrix characteristics in a coastal aquifer as shown by laboratory results from the research of Goldenberg and others (1986, 1993) and Goldenberg and Melloul (1994a).

Figure 4 presents chlorographs of wells S1, S2, S4, S5, S6, S10, S11, and S12, – all the wells involved in this study. They are located along the coastal aquifer of Israel and

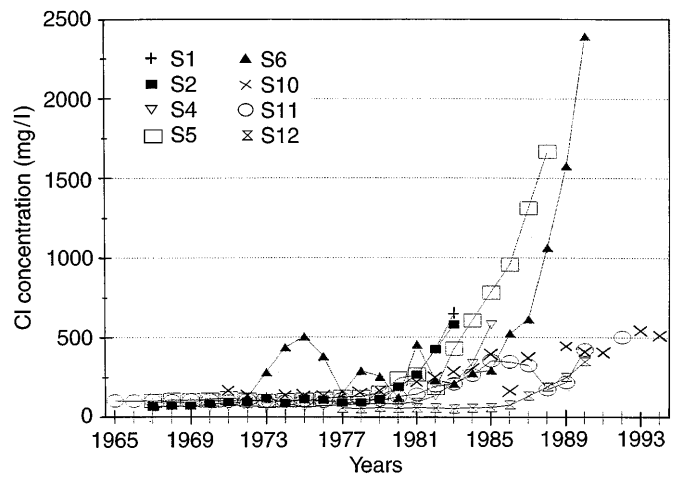


Fig. 4 Chlorographs illustrating the dynamics of seawater intrusion into various wells along the length of the coastal aquifer

the Gaza Strip, about 1500 m from the sea shore, in areas where seawater intrusion is expected. The figure indicates that in most cases, the chlorographs involve three separate portions or stages. The first stage entails initial well data history, generally characterized by the lowest levels and almost nonperceptible fluctuations in chloride concentrations. The second stage is characterized by higher concentrations and more prominent fluctuations than the previous stage. During this stage, Cl⁻ values are still low but fluctuate from 50% to 100% around an approximate mean value. The third stage commences when Cl⁻ values begin to increase continuously and rapidly (at a rate of around 30 mg/l of chlorides per year). Of key interest in this investigation is the second stage, which includes a fluctuation period which can take between 5 and 10 years before the onset of a sharp increase in ground-

Table 1

Chemical data illustrating different types of water. (1): Bachmat and Chetboun (1974), (2): Yechieli and others (1992), (3): Melloul and Collin (1992), (4), (5): Suleiman (1995), (6), (7), (8), (9): representative data from groundwater wells in seawater in-

trusion areas of the Israel coastal aquifer: groundwater salinity levels (in milligrams per liter of chlorides): a=fresh (<400), b=low salinity (<1000); c=brackish (<6000); d=saline (>6000)

chemical parameters	Medi- terranean Sea	Dead Sea brine	Rainfall in eastern coastal aquifer	types of groundwater in					
				seawater intrusion area of coastal wells in Libya		seawater intrusion area of coastal wells in Israel			
				1976 (4)	1993 (5)	a (6)	b (7)	c (8)	d (9)
rNa [Meq/L]	538	1744	0.2	3.17	143.5	3.1	5.7	105	283
rCl [Meq/L]	620	6344	0.26	3.0	169.5	2.7	8.7	140	343
rCa [Meq/L]	25	860	0.45	2.1	19.2	1.3	2.5	17.4	33
rSO ₄ [Meq/L]	65	10	0.24	0.83	25.7	0.08	0.06	7.3	18
rNa/rCl	0.87	0.27	1.1	1.05	0.85	1.15	0.66	0.75	0.82
rCa/rSO ₄	0.4	86	1.8	2.5	0.75	16	41	2.4	1.8

water salinity. However, because there are significant differences in Cl⁻ concentrations between the first and last stage of these graphs (with Cl⁻ values varying from 50 to around 2500 mg/l), fluctuations in the intermediate stage are not very distinct.

Figure 5 focusses only on chlorographs of wells S1 and S5, located around 10 km from one another, and S6, located in the Gaza Strip area, around 100 km from S5, along a north-to-south transept of the coastal aquifer. This figure indicates that despite the varied distances between these wells, their chlorographs indicate similar behavior, involving three distinct chlorograph stages. Of key interest is the fluctuation stage of the chlorograph of well S6, involving somewhat higher Cl⁻ levels than the other wells. High values have been explained in Melloul and Collin (1994) as resulting from stressed and polluted conditions existing in the aquifer. In this area, however, significant similarity can be noted in the third stage chlorograph breakthrough curve, characterized by a sharp increase in Cl⁻ content of wells located in the northern and southern portions of the coastal aquifer.

Figure 6 represents a typical chlorograph curve for well S1 which characterizes and summarizes the exchange process of freshwater by saline water. In this well, the three stages of the chlorograph (A, B, and C) are quite apparent. During stage B, fluctuations at low contamination levels of 60–130 mg/l occurred between 1969 and 1978. The first distinct fluctuation appears when Cl⁻ content subsequently increased to around 120 mg/l by 1970–1971. During the following year, Cl⁻ concentration dropped below the original low values to around 60 mg/l in 1972, only to increase again to around 130 mg/l in 1973, leading to another distinct fluctuation. Two years later, further fluctuation appeared after a decrease to nearly 70 mg/l in 1975 and a subsequent increase to around 130 mg/l, at which level it remained almost constant for 2 years. Such low Cl⁻ concentration fluctuations eventually ceased in 1979, when Cl⁻ started to increase continuously. Initially, in this third stage (stage C) the in-

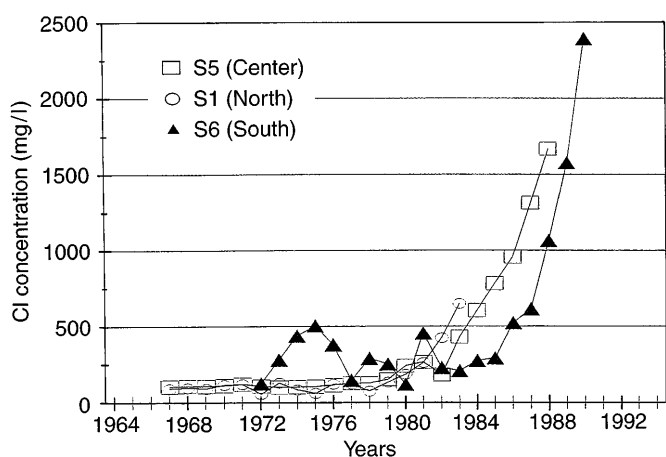


Fig. 5

Chlorograph illustrating the dynamics of seawater intrusion into wells located in the northern, central, and southern portions of the coastal aquifer (including the Gaza Strip area)

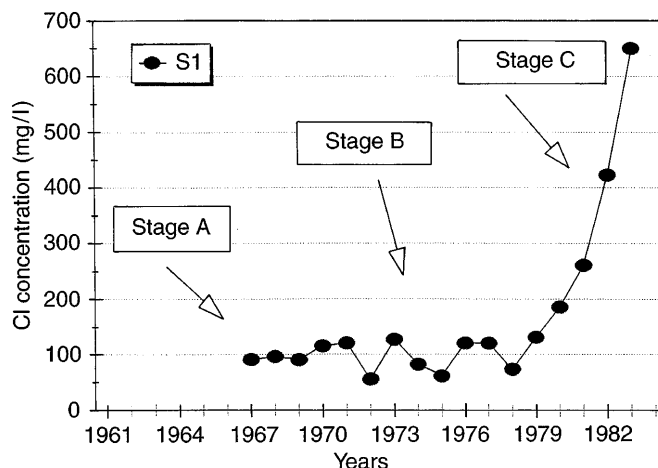


Fig. 6

Characteristic chlorograph illustrating the dynamics of sea-water intrusion into well S1

crease was at a rate of 70 mg/l per year for 2 years. Later, the increase rose to a rate of more than 100 mg/l per year. This figure indicates that distinct fluctuations occurred over a period of about 10 years before the sharp and ongoing increase in groundwater salinity.

Figure 7 shows nitrate data over a period of time from 1968 and from 1974 to 1994 for wells S1, S2, S4, S10, S11, and S12. This figure indicates that from 1977 to 1992, nitrates did not exhibit any substantial increase, whilst for the same wells, chlorides values increased sharply (Fig. 4). Figures 8 and 9 show the heterogeneity of groundwater chemical properties in the interstitial water of the aquifer, geographically and in profile, as well as changes in the properties of the matrix aquifer, based upon cores of two shallow coastal aquifer wells, as adopted from Goldenberg and Melloul (1994a).

Figure 8 shows the salt composition of interstitial water in the characteristic groundwater profile of well Katza-18,

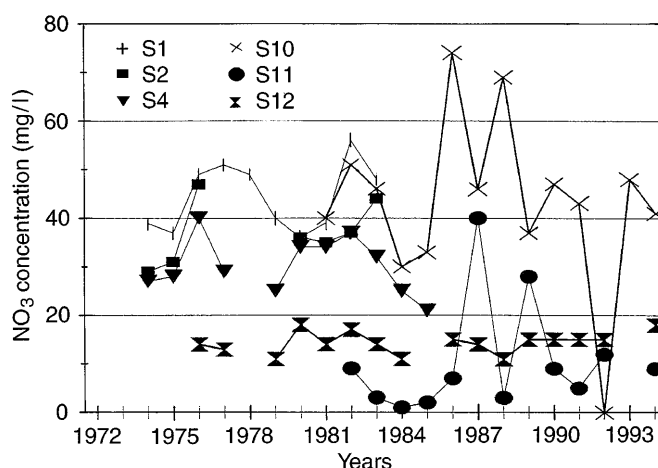


Fig. 7

Nitrographs of wells S1, S2, S4, S10, S11, and S12 located in seawater intrusion areas

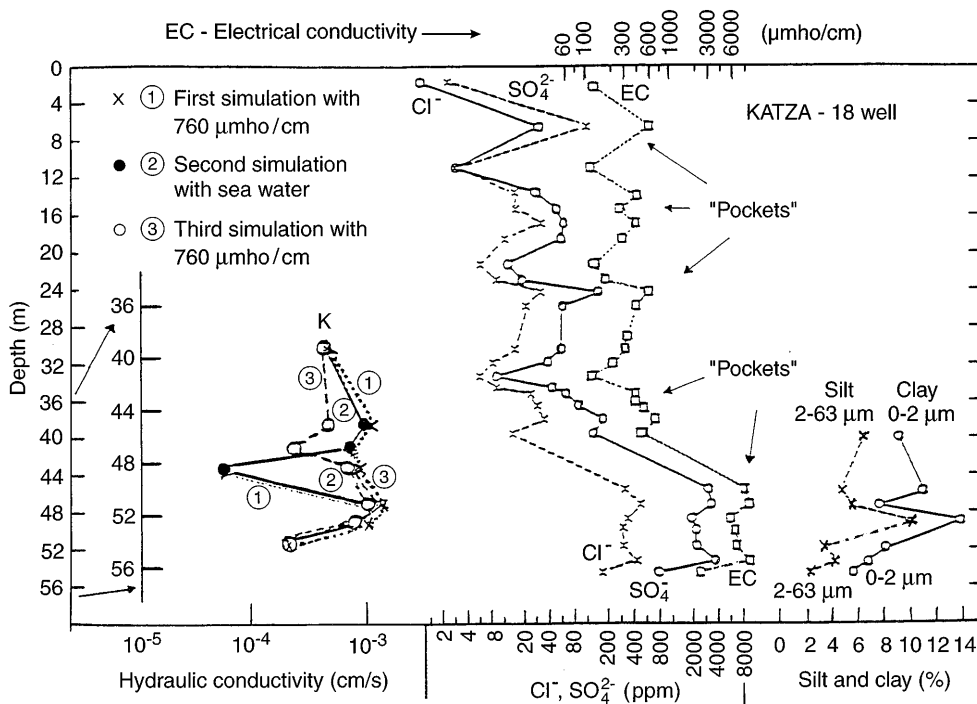


Fig. 8 Chemical, matrix and hydraulic properties of the Katza-18 core (taken from Goldenberg and Melloul 1994a)

near Ashqelon (Fig. 1). There, the aquifer consists mainly of sand, with around 4% silt (2–63 μm) and 6% clay (<2 μm). This figure also illustrates value changes in electrical conductivity (EC), chlorides (Cl^-) and sulfates (SO_4^{2-}) along the vertical water profile. Along this profile at a depth of between 32 and 55 m, some intermediate “pockets” characterized by more saline water may be observed. A general continuous trend of increase in EC with depth can be noted from around 100 $\mu\text{mho/cm}$ ($Z=2$ m) to about 6000 $\mu\text{mho/cm}$ ($Z=55$ m). This is also illustrated by Cl^- concentration which increases about two and half orders of magnitude, from 2 to 400 ppm (at $Z=55$ m). SO_4^{2-} behaves similarly to EC and Cl^- . Changes in hydraulic conductivity (K) of the aquifer are notable during laboratory experiments simulating infiltration of water having various properties. The simulation using relatively freshwater was followed by a run using saline water, resulting in a significant alteration in hydraulic conductivity (K). For instance, at $Z=46$ m and at $Z=48$ m (Fig. 8), permeability was reduced by about one order of magnitude (from initial characteristic values of about 10^{-3} to 10^{-4} cm/s). These laboratory simulations, related to specific clay and silt content in the profile of well Katza-18, show that the change in K can prove either reversible (e.g., the sample at $Z=48$ m), or irreversible (e.g., at $Z=45$ and 46 m).

Figure 9, representing well N-117, located only about 40 m from Katza 18, shows that despite the small distance between the two wells there are differing and varied “pockets” of saline groundwater. These pockets differ in magnitude. However, as in both wells, high values of SO_4^{2-} may be the result of gypsum in the local aquifer matrix.

In Table 1, typical chemical data are listed for the various types of water (surface and groundwater) which can be

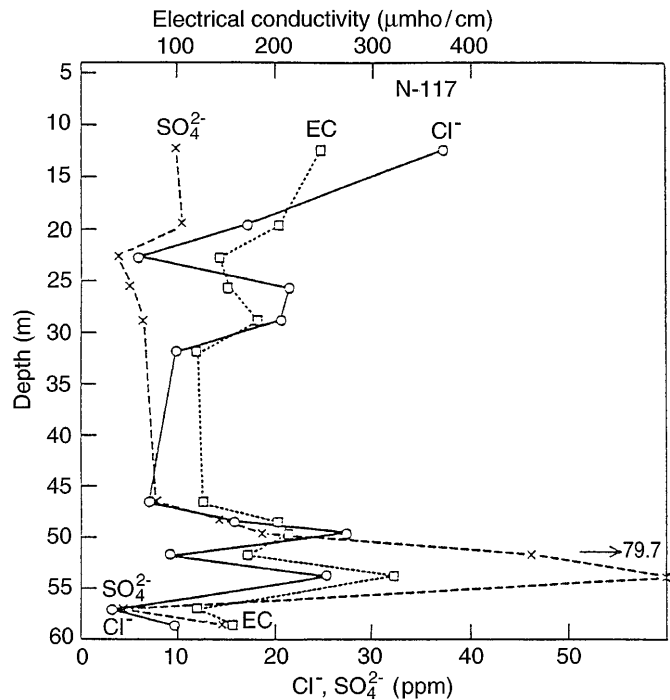


Fig. 9 Chemical characteristics of the N-117 core (taken from Goldenberg and Melloul 1994a)

found in this area. The chemical parameters shown are $r\text{Na}^+$, $r\text{Cl}^-$, $r\text{Ca}^{2+}$, and $r\text{SO}_4^{2-}$, as well as the characteristic ratios $r\text{Na}/r\text{Cl}$ and $r\text{Ca}/r\text{SO}_4$. Each water type has a specific genetic history. Mediterranean seawater is representative of seawater intrusion; rainfall over the eastern portion of the coastal aquifer is related to freshwater into the aquifer; Dead Seawater is representative of very saline

or brine properties; and (a), (b), (c) and (d) in the table represent respectively, characteristic fresh, low salinity, brackish, and very saline groundwater found in the study area (Fink 1970; Melloul and Bachmat 1975; Vengosh and others 1991; Vengosh and Starinsky 1993). Two key points are evident from Table 1. One is that water of relatively low salinity (with Cl^- lower than 400 mg/l) characterizing the fluctuation stage exhibits certain groundwater chemical properties which appear very similar to highly saline water such as brines (e.g., $r\text{Na}/r\text{Cl}=0.66$). The second point is that water with Cl concentrations higher than 6000 mg/l show a chemical resemblance to intruding Mediterranean seawater (e.g., $r\text{Na}/r\text{Cl} = 0.82$). Other data presented in this table are taken from groundwater wells located in seawater intrusion areas of the Suani wellfield, at Tripoli, Libya from Suleiman (1995) which show a similarity between the Libyan water and that of the coastal plain aquifer along the eastern portion of the Mediterranean sea.

Discussions

The key issues of this study include: (1) the fact that near the sea shore contamination of aquifers results mainly from seawater and/or very salty water, (2) the variability in groundwater quality is due to the heterogeneity of the liquids and the aquifer media coming in contact in this area of study, and (3) an explanation of the process of the appearance of early signals.

Contamination of coastal aquifers by seawater and/or saltwater

This study indicates that near the sea shore, the main source of contamination of groundwater in the vicinity of the sampling locations is due to ancient and/or recent seawater intrusion. This is supported by five simultaneous factors:

1. The location of the study wells is inside an area where seawater intrusion has been detected (Fig. 3)
2. The appearance of Cl^- concentrations greater than 1000 mg/l and trends greater than 30 mg/l/year, indicated in Figs 4–6, demonstrates that the main source of contamination must be a massive and continuous source of saline water such as seawater and/or brines.
3. Chlorographs of wells in areas influenced by seawater all along the coastal aquifer depict similar chlorograph behavior despite the distance separating the wells (Fig. 5). Similarities were also noted between chlorograph curves of wells located in the seawater intrusion area of the Suani wellfield of Tripoli, Libya (Suleiman 1995) and those of the coastal aquifer of Israel (Table 1). It is therefore reasonable to presume that this process may characterize other seawater intrusion areas throughout the world.
4. The presence of sea- and/or saltwater can be indicated by chemical parameters like: Na^+ , Cl^- , Ca^{2+} , SO_4^{2-} ,

and by ratios like: $r\text{Na}/r\text{Cl}$ and $r\text{Ca}/r\text{SO}_4$, related to the various types of water encountered in the area (Table 1). In this table, low saline water (with Cl^- lower than 400 mg/l), shows chemical ratio values indicating a correspondence to saline water such as brines; more saline water (Cl^- around 6000 mg/l), shows chemical ratio values of $r\text{Na}/r\text{Cl}$ closely resembling the Mediterranean seawater.

These findings indicate that in stage C of the graph, the influence of seawater or salty water intrusion is predominant, whilst in the previous early-indication-signal stage, groundwater degradation occurs in minor fluctuations along with an increase in other parameters of groundwater chemistry due to various factors.

A factor which can mask this 'ideal' image of the fluctuation stage could be linked to a relatively high Cl^- concentration, originating in part from anthropogenic sources, as indicated by nitrates in Fig. 7. This may explain the presence of the relatively high values of Cl^- in the stage-B portion of the chlorograph of well S6, located in the Gaza strip in proximity to a high-risk polluted area (Melloul and Collin 1994). However, in stage C of this chlorograph, even in polluted areas, the effects of the Cl^- content indicating the key influence of seawater is more pronounced than are the effects of anthropogenic pollution sources.

Degree of heterogeneity of the aquifer and variability in flow velocities

The major factors which influence the appearance and distinctive aspects of early-indicator signals of groundwater contamination may be related to the degree of variation and heterogeneity of the porous media (variation in the physical aquifer parameters like transmissivity and porosity), changes in groundwater gradients and flow velocities which result from management procedures, and pollutant characteristics.

Of major significance are changes in formation porosity, secondary porosity, packing, shape, pore-size and pore-shape, grain, and grain-size distribution (Goldenberg and others 1986, 1993; Goldenberg and Melloul 1994a). Spatio-temporal variations in water-table gradients occur in response to differing rates of recharge and discharge (Rai and others 1994). As a result, variations in matrix cause variations in paths of fluid and particle flow through such porous media with nonuniform velocities, and hydraulic conductivities higher than those of the ambient porous medium. (Moreno and Tsang 1994; Goldenberg and others 1993).

Results of laboratory experiments from Goldenberg and others (1986, 1993) and Goldenberg and Melloul (1994a), show that dynamic variability in flow velocities results also from water-rock interactions (Figs. 8 and 9). Varying flow velocities appear in an aquifer due to the presence of different constant or variable driving forces over time. Stage-B behavior can thus be explained by these forces, which may be expressed at the macro-scale in either the dominant regional hydraulic gradient or in local gradients

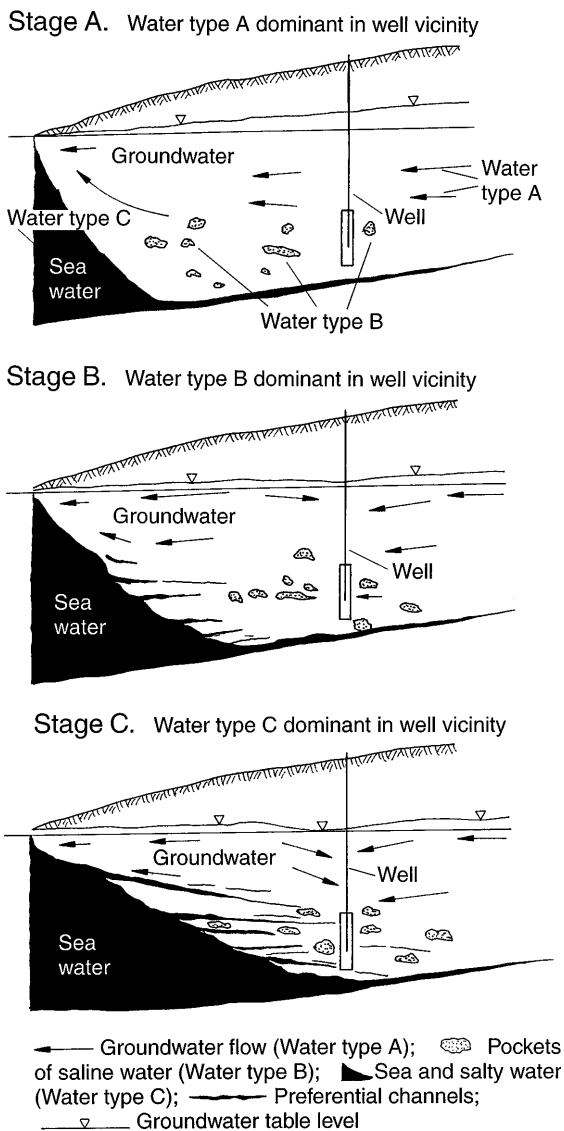


Fig. 10

A schematic presentation of the dynamics of early-indicator signals during the process of seawater intrusion

A proposed explanation of early-indicator signals of groundwater contamination

Explanation for the exchange of freshwater by contaminated water is schematically represented in Fig. 10 by the displacement of three water types (A, B, and C) found within a coastal aquifer where seawater intrusion is expected to be active. Water type A represents a fresh groundwater aquifer type with water arriving from recent rainfalls, or return flow from agriculture and/or other ground surface activities (identified in Table 1 by $rNa/rCl > 1$). Water type B represents "pockets" of water which are slightly to highly saline (brines). Such water, originating from recent or even ancient seawater, may be "trapped" within branches of preferential channels. Such water is identified in Table 1 by $rNa/rCl < 0.7$. Water type B is located between freshwater media (type A) and significantly contaminated saline water of type C such as

seawater (identified in Table 1 by $rNa/rCl = 0.85$), or brines (identified by $rNa/rCl = 0.7$).

When examining chloride concentration changes in wells in the vicinity of A and B water-type media, a scenario such as that schematically depicted in Fig. 10 may be expected involving three stages.

Initially, stage A (Fig. 10a) coincides with the steady-state stage, where freshwater (A) flows directly through the aquifer to the sea. Pockets of saline water that exist in this area remain neutralized by the massive flow of freshwater towards the sea. At this point, water type B is still inactive, and does not move toward the well, so that no altered concentration of aquifer groundwater quality would be observed in the well. This stage is related to the first portion of the chlorograph in Fig. 5.

Stage B (Fig. 10b) is a fluctuation stage, beginning locally, when inadequate management leads to over-pumpage. Local gradients may appear the vicinity of the well in question. In this unbalanced situation, saline water in "pockets" at various depths have become active and start to degrade groundwater quality in the well's vicinity. Degradation of the water occurs when initial flows of saline/contaminated water type B move through permeable portions of the aquifer more rapidly toward water type A than in areas of lower permeability. During this stage, specific dilution and diffusion processes occur. Initial contact of water type A with type B leads to minor increases in salinity. Thus, while water type C remains limited, changes in the matrix of the aquifer can be expected, as shown by Goldenberg and others (1986). These changes in aquifer matrix may result in a decrease in hydraulic conductivity, which can temporarily mitigate intrusion of saline water through certain pockets. Therefore, where the flow of fresh groundwater type A remains predominant, it continues to dilute and 'refresh' the ambient water. This can be noted by the initial fluctuation of chloride concentration in the vicinity of the well (Fig. 6). Subsequent fluctuations appear with minor increases and decreases in salinity, which can continue for some time. These constitute the early-indicator signals of groundwater salinization, characterizing areas of a coastal plain aquifer where groundwater salinization has already affected some of the pumping wells.

The third stage or stage C (Fig. 10c) is a stage of intensive salinization, characterizing areas where the rate of pumpage of the aquifer exceeds its safe yield capacity. In such areas, local gradients begin to reduce recharge of freshwater. At this point, water type B, located in more hydraulically conductive channels and pockets of saline water, is increasingly thrust by water type C toward the vicinity of the well. The influence of water type C subsequently increases dramatically, and groundwater contamination of the aquifer increases sharply and often nonreversibly, as represented in Fig. 6. As indicated by data in Table 1, this stage also appears in the coastal aquifers of Libya, and may be expected in other coastal aquifers of the world.

Preceding the arrival of water type C in the vicinity of a well, early-indicator signals of aquifer pollution may ap-

pear as an ongoing gradual trend, involving slight fluctuations of groundwater chloride concentration. The fluctuation stage can involve a number of years, amounting to around 50%–100% of the background groundwater Cl concentration level, as revealed in Fig. 4.

Conclusions

The stage of fluctuation or early-indicator signals is influenced by the intensity of pumpage, and high groundwater and aquifer media heterogeneity. The findings presented by means of chlorographs and other chemical parameters indicate that only during the last stage of salinization can the main source of contamination be tied to sea or other highly salty water intrusions. During the intermediate stage, the groundwater salinity increase can still not be directly connected with active seawater intrusion.

In the case of contamination of coastal aquifers, small fluctuations and slight increases in Cl^- concentration of groundwater can be used as an indicator, and considered as a 'symptomatic' stage of salinization of coastal aquifers. The increased quantity of Cl^- the ambient native groundwater during the fluctuation stage can reach 50%–100%, relative to the background. The ongoing contamination process of an aquifer involves two major driving forces. On the one hand, the contaminant source (in this case, seawater) attempts to penetrate the native water resource. Over against this are the regional and local gradients of the groundwater body, as affected by aquifer pumpage.

This study focusses upon early-indicator signals as an initial and a particular process of groundwater contamination in areas of sea- or other saltwater intrusion. Such a process may be anthropogenically augmented and largely affected by the intrinsic properties of the aquifer's porous medium.

The relevance of early-indicator signals can be seen in their ability to assess the importance of an initial appearance of a pollutant which may result from point and/or diffuse sources.

For proper management of aquifers, it is critical to predict and maintain a safe level of hydrological yield.

Therefore, monitoring to perceive early-indicator signals should be one of the guidelines protecting water quality in order to maintain the buffering capacity of the aquifer to absorb contaminant input as well as the dynamic hydrological balance between withdrawal and recharge.

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References

- BACHMAT Y, CHETBOUN G (1974) Seawater encroachment in the coastal Plain aquifer of Israel during the period 1958–1971. Hydrol Serv, Rep 2/74, Jerusalem,
- CHOW VT (1964) Handbook of applied hydrology. Mc Graw-Hill, New York
- FINK M (1970) The hydrogeology of the Gaza Strip (in Hebrew). Tahal report no. 3, Tel -Aviv
- GEHRELS JC, VAN GEER FC, DE VRIES JJ (1994) Decomposition of groundwater level fluctuations using transfer modelling in an area with shallow to deep unsaturated zones. J Hydrol, 157: 105–138
- GOLDENBERG LC, MELLOUL AJ (1994a) Some aspects of groundwater rehabilitation. In: Zoller U (ed) Ground cont and control. Marcel Dekker, New York
- GOLDENBERG LC, MELLOUL AJ (1994b) Rehabilitation aspects of groundwater quality management and some relevant laboratory findings. In: Groundwater quality management, Proc GQM 93, Conf, Tallin. IAHS Pub 220, pp 383–392
- GOLDENBERG LC, MELLOUL AJ (1994c) Hydrological and chemical management in the rehabilitation of an aquifer. J Environ Manag 42: 247–260
- GOLDENBERG LC, MANDEL S, and MAGARITZ M (1986) Fluctuating non-homogeneous changes of hydraulic conductivity in porous media. Q J Eng Geology, 19: 183–190
- GOLDENBERG LC, HUTCHEON I, WALDLAW N, MELLOUL A (1993) Rearrangement of fine particles in porous media causing reduction of permeability and formation of preferred pathways of flow: experimental findings and a conceptual model. Transp Por Med 13: 221–237
- GRAHAM WD, NEFF CUR (1994) Optimal estimation of spatially variable recharge and transmissivity fields under steady state groundwater flow. Part 2 case-study. J Hydrol 157: 267–286
- Hydrological Service Situation Report (1994) Development of groundwater resources in Israel up to Autumn 1993 (in Hebrew). Rep 93–1093
- KONIKOW LF, RODRIGUEZ AJ (1993) Advection and diffusion in a variable-salinity confining layer. Water Resour Res 29/ 8: 2747–2761
- LATINOPOULOS P, THEODOSIOU N, MYLOPOULOS J, MYLOPOULOS N (1994) A sensitivity analysis and parametric study for the evaluation of the optimal management of a contaminated aquifer. Water Resour Manage Ser 8: 11–31
- MELLOUL A (1988) Hydrogeological atlas of Israel's coastal plain aquifer geometry and physical properties (in Hebrew). Hydrol Serv Rep 8/88 Jerusalem
- MELLOUL A, BACHMAT Y (1975) Assessment of the hydrological situation in the Gaza Strip as a basis for the management of the coastal Plain aquifer in that region (in Hebrew). Hydrol Serv Rep 3/75, Jerusalem
- MELLOUL A, BIBAS M (1990) Regional and local hydrological situation in the coastal Plain aquifer and water quantities according to quality standards (chlorides and nitrates) from 1987/1988 and anticipated through 1992 (in Hebrew). Hydrol Serv Rep 3/90, Jerusalem
- MELLOUL A, COLLIN M (1992) The "Principal Components" statistical method as a complementary approach to geochemical methods in water quality factor identification. J Hydrol 140: 49–73
- MELLOUL AJ, COLLIN M (1994) The hydrological malaise of the Gaza strip. Israel J Earth Sci 43: 105–116
- MELLOUL A, GILAD D (1993) Movement of seawater intrusion in the coastal aquifer of Israel over the years 1983–1990 (in Hebrew). Water Irrig J 318: 46–48

- MELLOUL AJ, GOLDENBERG LC (1994) Groundwater pollution by airborne toxic contaminants: a factor to be considered in groundwater management. In: "Groundwater quality management" Proc GQM 93, Conf Tallin. IAHS Pub 220, pp 95–105
- MORENO L, C-F TSANG (1994) Flow channeling in strongly heterogeneous porous media – a numerical study. *Water Resour Res* 30:1421–1430
- NANCE HS (1988) Interfingering of evaporates and red beds: an example from the Queen Gray burg geological formation: Texas. *Sediment Geol* 56:357–381
- POCIASK-KARTECZKA J (1994) Anthropogenic changes in the water conditions in Cracow (East-Central Europe) since the Second World War. *J Environ Hydrol* 2:14–24
- RAI SN, MANGLIK A, SINGH RN (1994) Water table fluctuation in response to transient recharge from a rectangular basin. *Water Resour Manag* 8:1–10
- SULEIMAN SE (1995) Deterioration of quality of groundwater from Suani Wellfield, Tripoli, Libya 1976–93. *Hydrogeol J* 3:58–64
- TODD DK (1961) *Groundwater Hydrology*. Wiley, New York
- US EPA (1990) *Progress in groundwater protection and restoration: 440/6–001*
- VENGOSH A, STARINSKY A (1993) Relics of evaporated seawater in deep basins of the eastern Mediterranean. *Mar Geol* 115:15–19
- VENGOSH A, STARINSKY A, MELLOUL A, FINK M, ERLICH S (1991) Salinization of the coastal aquifer water by Ca-Chloride solutions at the interface zone, along the coastal plain of Israel (in Hebrew with English abstract). *Hydrol Serv Rep* 91/20
- WHITE KE (1977) Tracer methods for the determination of groundwater residence time distributions. In: *Groundwater quality - measurement, prediction and protection*. Water Research Centre, Medmenham, England, pp 246–273
- YECHIELI Y, MAGARITZ M., LEVY Y, RAHNER S, KAFRI U, (1992) Chemical processes of the interface between the Dead Sea and groundwater. *Internal Rep* 58/91, Dep Environmental Sciences, Weizman Institute of Science, Rehovot and the Geological Survey, Jerusalem