

Abstract

Two sequential multilevel profiles were obtained in an observation well opened to a 130 m thick, unconfined, contaminated aquifer, in Tel Aviv, Israel. While the general profile characteristics of major ions, trace elements, and Volatile Organic Compounds (VOC) were maintained in the two sampling campaigns conducted 295 days apart, the vertical locations of high concentration gradients were shifted between the two profiles. Principle Component Analysis (PCA) of the chemical variables resulted in a first Principal Component (PC) which was responsible for $\sim 60\%$ of the variability, and was highly correlated with depth. PCA revealed three distinct depth-dependent water bodies in both multilevel profiles, which were found to be shifted vertically between the sampling events. This shift cut across a clayey bed which separated between the top and intermediate water bodies in the first profile, and was located entirely within the intermediate water body in the second profile. Continuous Electrical Conductivity (EC) monitoring in a packed off section of the observation well revealed an event in which a distinct water body flowed through the monitored section ($v \sim 150 \text{ m yr}^{-1}$). Compilation of the aforementioned data and analysis lead to a conclusion that the observed changes in the profiles resulted from dominantly lateral flow of water bodies in the aquifer rather than vertical flow. The significance of this study is twofold: (a) it demonstrates the utility of sequential multilevel observations from deep wells and the efficacy of PCA for evaluating this data. (b) The fact that distinct water bodies of 10–100 m vertical and horizontal dimensions flow under contaminated sites has implications for monitoring and remediation.

1 Introduction

Multilevel sampling (MLS) was found to be a useful method for improving hydrochemical and hydraulic characterization of aquifers relative to other sampling methods (Ronen et al., 1986; Cherry et al., 2007; Müller et al., 2010; Kurtzman et al., 2011).

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Depth-dependent samples from deep aquifers are usually obtained during drilling (Williams and Chou, 2007, Hendry et al., 2011); hence interpretation of aquifer dynamics from these single-time data sets is limited. In the present study we explore deep aquifer dynamics in light of a unique set of observations consisting of two sequential MLS campaigns. Discreet-depth groundwater samples were obtained from a customized monitoring well at a contaminated industrial site located in the Coastal Aquifer of Israel (Graber et al., 2008). Significant vertical variability in the chemical composition of groundwater in this deep well at a single point in time was described by Netzer et al. (2011). The current study focuses on the analysis of the aquifer dynamics that can explain the differences in the observed hydrochemical vertical distributions obtained in consecutive samplings. This analysis includes delineation of discreet water bodies, consideration of vertical and lateral flow, and assessment of a hydro-stratigraphic separation. To do this, Principal Component Analysis (PCA) was employed to examine the depth and time dependent chemical variables from the two multilevel profiles. Groundwater Electrical Conductivity (EC) and head monitoring and measurements provided supplementary information for the investigation of possible dynamics that modified the time-dependent hydrochemical profiles.

PCA is a multivariate statistical procedure used usually for exploratory data analysis. It transforms a large number of possibly correlated variables into a smaller number of uncorrelated (orthogonal) variables called Principal Components (PC). The first PC accounts for as much of the variability in the data as possible, and the succeeding components follow this same principle (StatSoft, Inc. 2011). PCA of major ions and trace element concentrations were used to evaluate recharge in a karst aquifer and groundwater flow in a regional aquifer (Moore et al., 2009; Stetzenbach et al., 1999, respectively). Angelone et al. (2009) used PCA of major ions and trace elements from wells in an area of 900 km² to verify the source of arsenic-rich water. However, to the best of our knowledge, PCA has not been used to analyze time and depth dependent hydrochemical data sets from a single deep well.

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2 Materials and methods

2.1 Site and observation well

5 A 150 m deep observation well that was designed for MLS extends throughout the entire vertical section of the Coastal Aquifer of Israel in Tel Aviv (32°04'21" N, 34°47'58" E). Extensive and intensive industrial contamination of the unsaturated zone and groundwater with chlorinated organic compounds and heavy metals in this area was previously reported (Ronen et al., 2005; Graber et al., 2008). The deep observation well was drilled to explore the vertical distribution of contaminants at the center of the plume. In the study area, a 6 m thick clayey bed between 73–79 m below ground surface (mbgs) divides the aquifer into two main sub-aquifers (Fig. 1). Previous work showed that this type of clayey bed often creates effective hydraulic separations (Nativ and Weisbrod, 1994). At the monitoring location, the full saturated thickness of the aquifer is ~ 130 m from water table to the bottom confining Saqiye Clays (Fig. 1). The monitoring well was drilled by a percussion tool with no drilling mud. It was designed to insure reliable deep multilevel sampling by inserting bentonite and PVC seals into the sand-pack between and within perforated sections (Fig. 1). Two separate pipes, one tapping the deep sub-aquifer between 79 and 149 mbgs (D, Fig. 1), and one tapping the shallow sub-aquifer between 18 to 72 mbgs (S, Fig. 1), were installed in the borehole to avoid cross contamination between the two sub-aquifers (more details in Netzer et al., 2011).

2.2 MLS campaigns and chemical analyses

25 Groundwater samples were obtained using a passive multi layer sampler methodology described previously (Ronen et al., 1986; Laor et al., 2003; Kurtzman et al., 2011). Briefly, each sampling unit consists of an individual dialysis cell that can be attached in a modular fashion. Each unit has a stainless steel ring into which a stainless steel cylindrical dialysis cell (150 ml) is inserted. The sampling cells are connected one to

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a 5973 network Mass Selective detector (Agilent Technologies). Dissolved organic carbon (DOC) (MLS2 only) was analyzed, after acidification to pH 3.5 and air bubbling for 2 min, by Formacs TOC analyzer. Chemical analyses were performed at the laboratories of the Agricultural Research Organization, Israel, and the Israel Water Authority.

Two types of errors can affect the determined concentrations: analytical and retrieval errors. Retrieval errors are unique to the specific deep MLS setting reported here. These errors may be significant in deep samples due to the time it takes to retrieve the deep dialysis cells. During retrieval, these cells may be in contact for short times (up to 75 min for the deepest samples, Ronen et al., 2010) with water that is not from the interval they sampled. Examples of total relative errors (sum of analytical and retrieval errors) are 2–12 % for Cl^- and 12–21 % for trichloroethylene (TCE), where the higher errors are for the deeper samples (Ronen et al., 2010). Interpretations herein focus on concentration differences in depth and time that are significantly larger than these errors.

2.3 Principle Component Analysis (PCA)

The groundwater-chemical variables were analyzed by PCA for the 17 sampling depths (in statistical terminology, 17 cases) of MLS1 and the 24 cases of MLS2 (Fig. 2). For MLS1, 23 variables were used for analysis: Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- , SO_4^{2-} , HCO_3^- , NO_3^- , Br^- , As, Al, B, Be, Ba, Co, Cr, Mn^{2+} , Ni, Pb, Se, Sr, U and total dissolved solids (TDS) (if no oxidation state is specified the analysis was of the total element). The same variables were utilized in the PCA of MLS2 except for Pb, Ni and As (concentrations below the limit of quantification in most cases) and Al (not analyzed). One additional variable considered in MLS2 was DOC. The total number of variables for the PCA of MLS2 was 20. VOCs, analyzed at only 12 and 15 depths in MLS1 and MLS2, respectively, were not included in the PCA. PCA was performed with STASTICA software using correlation matrices (StatSoft, 2011).

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2.4 Groundwater head and EC measurements

Head was measured in the deep pipe (D) and the shallow pipe (S) of the observation well (Fig. 1) 25 times between April 2008 and October 2009. A combined water level–temperature–conductivity, logger (LTC, Solinst®), was inserted in between Viton seals (similar to one interval of the MLS apparatus) and deployed at different depths (19, 20 and 31.5 mbgs). The confined vertical length between the two seals was 35 cm. Water level, temperature and EC were recorded every 30 min.

3 Results and discussion

3.1 MLS profiles

Pairs of profiles (MLS1 and MLS2) of 8 representative chemical variables are discussed in this section, whereas a more general analysis obtained with PCA of data of more than 20 analyzed chemicals will be presented in the next section. However, the results and interpretation presented herein involve many links between these two sections which the reader is advised to follow.

The main characteristics of each chemical profile over the 130 m thick aquifer were preserved in both sampling events (Fig. 3). These general traits of the profiles can be classified into 5 characteristic types: (a) concentrations generally increase with depth (e.g. Na^+ and Cl^- , Fig. 3c, d); (b) concentrations generally decrease with depth (e.g. SO_4^{2-} , Fig. 3b); (c) concentrations generally increase with depth but decrease near aquifer bottom (e.g. Mg^{2+} , Fig. 3a); (d) Concentrations generally decrease with depth but increase near aquifer bottom (e.g. Co, Fig. 3f); (e) Extremely high concentrations in the shallow sub-aquifer and orders of magnitude lower concentrations in the deep sub-aquifer (e.g. Cr, TCE and tetrachloroethylene (PCE), Fig. 3e, g, h). Profile types a–d will be referred to in the PCA in the next section.

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Despite the general similarities in chemical trends in MLS1 and MLS2 profiles, a detailed examination of the profiles reveals distinctive differences between them. The most pronounced difference is the upward vertical shift of the boundary between concentrations typical to the upper part of the aquifer and concentrations typical to the lower part of the aquifer for many chemical variables (e.g. Mg^{2+} , SO_4^{2-} , Cr, Co, TCE, and PCE, Fig. 3a, b, e–h). In MLS1, the boundary is consistent with stratigraphy, falling between the sampling depths of 70.3 and 84.2 mbgs, the interval that contains the 6 m thick clayey layer (Fig. 1), with Cr being the only exception. In MLS2, this hydrochemical boundary lies apparently between 59.5 and 67.2 m depth, i.e. within the upper sandstone sub-aquifer (Figs. 2 and 3a, b, e–h).

Whereas the shift in the hydrochemical boundary between the shallower and deeper waters was not depicted in the profiles of Na^+ and Cl^- (Fig. 3c, d), a change in the depth of the highest concentration-gradient within the deep sub-aquifer is distinct. In MLS1 it occurred in the 84–96 m interval whereas in MLS2 it occurred between 102–114 m (Fig. 3c, d). These large concentration gradients in the 102–114 m interval that were observed only in MLS2 were pronounced also in other variables (NO_3^- , Br^- , B, Ronen et al., 2010), and will show clear in the PCA hereafter.

3.2 PCA of the sequential MLS profiles

In the following, results and interpretations concerning the main sources of variability, and water-body classification will be discussed using the projections of variables (chemicals) and cases (depths) on the plane of the first and second PCs (Fig. 4). The correlation of a variable with a PC is the value of that PC-component in the variable projection on the PCs plane (e.g. in Fig. 4a we learn that in the PCA of MLS1 the correlation of Ca^{2+} with PC1 (horizontal axis) is 0.84 and its correlation with PC2 (vertical axis) is 0.03).

In both the PCA of MLS1 and MLS2, Cl^- , Na^+ , K^+ have a high correlation with PC1 (negative correlation in MLS1 and positive in MLS2, Fig. 4a, b, respectively). These variables show a type a profile (concentrations increase with depth) and will be referred

form a non-monotonous profile (e.g. compare profiles and PC2-correlations of Mg^{2+} and SO_4^{2-} (Figs. 3a, b and 4a, b)). In light of this analysis of PC2 (19% of variability in both MLS1 and MLS2), it can be concluded that the second important control over chemical variability in the profile is the different hydrochemical conditions that prevail at the bottom of the aquifer.

In both MLS1 and MLS2 case-projections, three groups of cases are distinct (Fig. 4c,d). These three groups are consistent with depth, i.e. all cases within a group are continuous sampling depths with no exceptions (Figs. 2 and 4c,d). The clear distinction of vertically-separated three water bodies in the PCA of MLS1 and MLS2 chemical data is the most significant outcome of the PCA concerning the aquifer dynamics characterization. Hereafter the three water bodies are denoted as top, intermediate and bottom water bodies (Fig. 4c, d).

A significant difference between the PCA of MLS1 and MLS2 is that some sampling depths are in different water bodies: Depths 67 and 70.3 mbgs were in the top water body in MLS1 and moved to the intermediate one in MLS2. Depths 114, 130 and 140 mbgs were in the intermediate water body in MLS1 and are in the bottom water body (or at least much closer to it) in MLS2 (Fig. 4c, d). The boundary between the top and the intermediate water bodies was between 70 and 84 m in MLS1 (consistent with stratigraphy, Fig. 1), whereas in MLS2 this boundary lays between 59 and 67 mbgs. It also appears that the boundary between the intermediate and bottom water bodies shifted from between 140–146 mbgs in MLS1 to between 102–114 mbgs in MLS2 (Fig. 4c, d). These changes in the position of the boundaries between the chemically-different water bodies that can be obtained, to some extent, by exploring the MLS1 and MLS2 profiles (Fig. 3) becomes sharper with the PCA (Fig. 4). A discussion on aquifer dynamics that may have caused the aforementioned vertical shifts of water-bodies boundaries will follow after a section on supplemental observations that are significant for this discussion.

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3.3 Supplemental information: groundwater head and EC measurements

During the period between MLS1 and MLS2, the groundwater head measured in the deep pipe (D, Fig. 1) was higher than that measured in the shallow pipe (S, Fig. 1) in 11 out of 12 measurements (~97% of the time, Fig. 5). The difference between the overall head in the deep sub-aquifer and the head in the shallow sub-aquifer during the 295 days period ranged between -2 to 27 cm (Fig. 5). This data is suggestive of an upward flow from the deep to the shallow sub-aquifer during the time-interval between the two sampling dates. However, this is not a definitive interpretation, seeing as these head measurements reflect the overall head in the tapped part of the aquifer rather than the specific head at a given point in the screened interval. Therefore we can only conclude that, looking over the entire width of the aquifer during this period, the vertical component of flow was more upward than downward, but cannot conclude anything about the prevailing flow at any specific depth.

EC was monitored in 35-cm packed sections at 3 depths (19, 20 and 31.5 mbgs) for 2 months prior to the deployment of MLS1. Although the EC monitoring did not take place during the MLS campaign, some time-dependent EC results at 31.5 m depth shed light on aquifer dynamics. An abrupt and significant decrease in EC (~0.6 mS cm⁻¹, Fig. 6) was observed once during the 2 months of observations in which the LTC probe was positioned at the three depths (~3 weeks per depth). Head and temperature measured simultaneously did not show any such contemporaneous discontinuities. Hence, the change can only be explained by lateral flow of a fresher water body through the monitored section. Change in concentrations in the packed-of interval during this passage is caused by water from the interval with concentration C leaving the interval and new water with lower concentration C_2 entering the interval (Eq. 1, Fig. 6).

$$\frac{dC}{dt} = -\frac{Q}{V}(C - C_2) \quad (1)$$

where t is time (T), Q is flow-rate through the interval (L³T⁻¹) and V is the volume

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of the packed of interval (minus the probe volume) (L^3). The solution of Eq. (1) is $\ln((C - C_2)/(C_1 - C_2)) = -Q/V(t - t_1)$ where C_1 and t_1 are the time and concentration at the beginning of the concentration change (Fig. 6). Knowing V enables determination of Q . Groundwater specific discharge $-q(LT^{-1})$ is derived by Eq. (2).

$$q = \frac{Q}{LD\alpha} \quad (2)$$

where L and D are the section's length and diameter, respectively (L) and $\alpha(-)$ is a factor correcting for the convergence of the natural aquifer flow towards the well (= 2 for homogenous media). This analysis resulted in $q \sim 40 \text{ m yr}^{-1}$. If we use an effective porosity of 0.25 for this aquifer (e.g. Assouline and Shavit, 2004) we have a groundwater velocity in the neighborhood of $v \sim 150 \text{ m yr}^{-1}$. This lateral velocity estimate suggests that some tens to a hundred m may divide laterally between the water bodies sampled in MLS1 and MLS2. EC and chloride concentration from the two MLS show a significant linear relation ($R^2 = 0.76$, $P < 1 \times 10^{-11}$), hence the EC observations can be approximated as concentrations for the above calculations.

3.4 Aquifer dynamics that can explain the change between MLS1 and MLS2

In the following discussion, two "end members" of possible flow regimes in the vicinity of the observation well will be examined in light of the previous observations and analyses. Assuming a 1-D vertical system and neglecting change in concentrations of major and trace elements due to reactions during the 295 days between MLS1 and MLS2, we can hypothesize a vertical flow regime that may explain the changes in the vertical distribution of the three water bodies obtained by the PCA (Figs. 4c, d and 7a) as follows. The water from the bottom of the aquifer flowed upward to form a larger water body which is a mix of previous bottom water and the deeper part of the previous intermediate water (Figs. 4d and 7a). The upper part of the intermediate water body was pushed up over the intermediate clay. Relatively low heads at $\sim 65 \text{ m}$ depth may have been the trigger for the upward flow in the column which moved the high chemical-gradients from over the clay into the shallow sub-aquifer (Figs. 7a, and 3a, b,

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3f–h). Within the upper section it looks as though there was also an upward movement raising the upper sub-aquifer's peaks in MLS2 (Figs. 7a and 3b–e, g).

The other “end member” flow regime would be lateral (horizontal) flow of different water bodies that pass through the observation well (Fig. 7b). No assumptions are needed to explain the differences between MLS1 and MLS2 with this flow regime except for assuming the existence of chemically-distinctive water bodies throughout the entire depth of the aquifer. Magnitude and direction of lateral velocities at different depths may differ.

The vertical velocity component is usually much smaller than the horizontal components in natural groundwater flow, and our observation well was not in close vicinity to a pumping well. However, we observed high levels of contamination as deep as 50 m below the water table (Fig. 3e, g, h). These contaminants originated at the surface probably after the 1940s (Graber et al., 2008), hence a vertical-velocity component on the order of 1 m yr^{-1} existed at least at the top sub-aquifer.

Evidence of lateral flow of distinct water bodies through the monitoring well at 31.5 mbgs ($\sim 13 \text{ m}$ below the water table) that was observed by EC monitoring (Fig. 6) supports the lateral flow hypothesis. Water parcels of different chemical characteristics at smaller scale (0–2 m below the water table) in a polluted site in the same aquifer were reported by Ronen et al. (1987). A lateral flow regime suggests that the intermediate water body in MLS2 was formed in a place in the aquifer where the 73–79 mbgs clayey layer is absent and after flowing laterally into the clay-divided area of the aquifer it exists both on top and below the clay layer (Fig. 7b). Clayey layers (lenses) at these depths in this part of the aquifer are of continental origin and known to be discontinuous over short distances in many cases (Ecker, 1999). Therefore, the lateral flow explanation makes more sense than fast vertical flow across the clayey layer.

If we assume that the differences between the profiles occurred due to upward flow we should expect a better conservation of mass within the water bodies. Looking at the profiles of the industrial contaminants it is clear that whereas the mass of TCE and Cr in the shallow sub-aquifer increased significantly from MLS1 to MLS2, the mass

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of PCE decreased (Fig. 3e, g, h). High TCE concentrations increased from $\sim 50\,000$ to $\sim 120\,000\ \mu\text{g l}^{-1}$, while those of PCE decreased from ~ 1800 to $\sim 700\ \mu\text{g l}^{-1}$, such that these changes cannot be explained by degradation of PCE to TCE. It is more likely that MLS1 and MLS2 sampled different water bodies of different contamination characteristics due to their different lateral origin and history.

4 Summary and conclusions

Two sequential MLS campaigns were conducted in a 150 m deep observation well penetrating the entire section of the Israeli Coastal Aquifer in Tel-Aviv. Major ions, trace elements and VOCs were analyzed. The general characteristics of the profiles of the chemical substances were maintained in the two sampling campaigns 295 days apart, yet significant differences in details were found (e.g. change in the depth of major vertical concentration gradients). PCA of the chemical variables resulted with a first PC that is highly correlated with depth (PC1 $\sim 60\%$ of the variability). PC2 ($\sim 20\%$ of the variability) was found to be associated with the variability caused by the different hydrochemical characteristics at the bottom of the aquifer, which causes sharp changes in concentration of some chemical variables. Three distinct depth-dependent water bodies were found in the PCA of both MLS profiles. The two vertical boundaries separating the three water bodies shifted upwards between MLS1 and MLS2. A clay bed which divided the top and intermediate water bodies in MLS1 was entirely within the intermediate water body in MLS2. An evaluation of whether vertical or lateral flows were the most probable cause for this shift concluded that most likely lateral flow was responsible for the observed changes. This conclusion is based on: (1) lateral flow ($v \sim 150\ \text{m yr}^{-1}$) of distinct water bodies was observed in this well by continuous EC monitoring in a packed off section. (2) Mass of contaminants was not preserved within the water bodies and the contamination characteristics were different in the two MLS profiles. (3) It is easier to explain the inconsistency of the water-bodies boundaries with stratigraphy in MLS2 with horizontal flow and limited lateral extent of the clayey bed, than rapid vertical flow across this bed. The aforementioned methods and findings

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are probably applicable to other contaminated sites in deep unconfined aquifers, especially, the utility of sequential multilevel observations from deep wells and the efficacy of PCA of this type of data. The fact that distinct water bodies of 10–100 m vertical and horizontal dimensions flow under such sites has implications for monitoring and remediation.

Acknowledgements. This study was financed by the Israel Water Authority. The conclusions presented represent those of the authors of this study.

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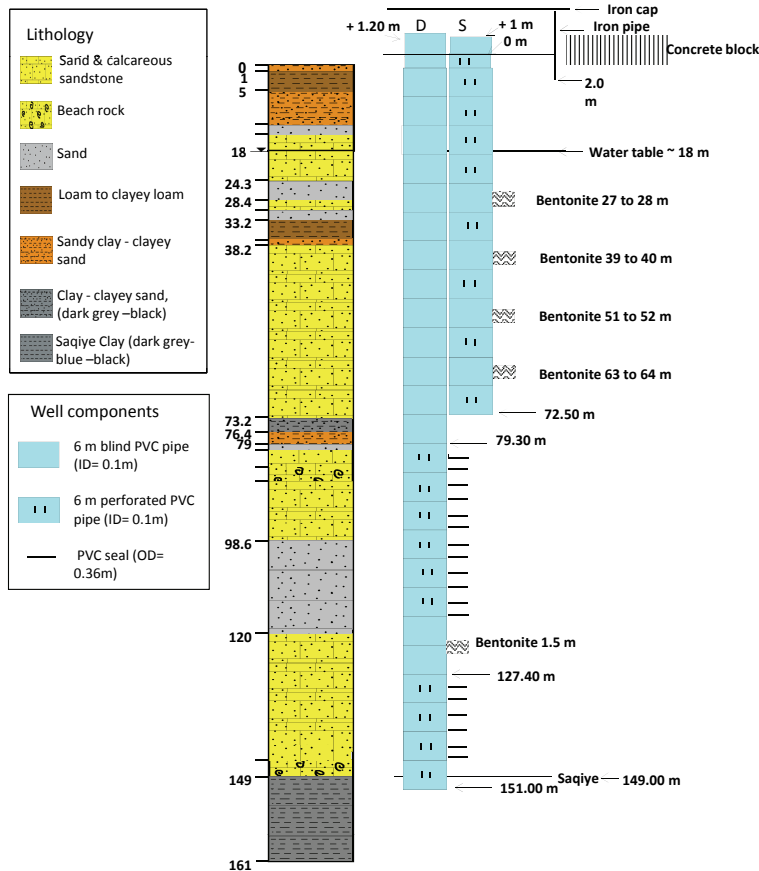


Fig. 1. Lithological log and perforation details of the observation well that was used for sequential MLS. D – pipe perforated against the deep sub-aquifer (79–149 m); S – pipe perforated against the shallow sub-aquifer (18–72 m), all depths are relative to ground surface.

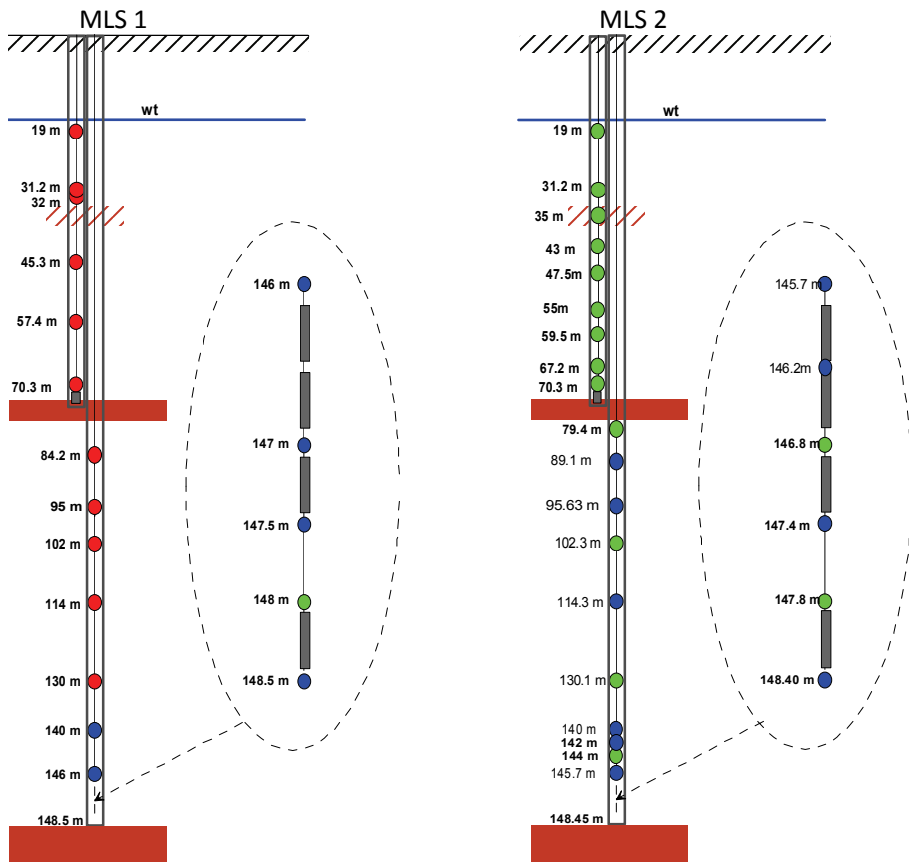


Fig. 2. Depth in m below surface of MLS units in the two campaigns. Blue – 1 cell per depth (major ions and trace elements analysis), green – 2 and red 3 cells per depth (major ions, trace elements and volatile organic compound analysis).

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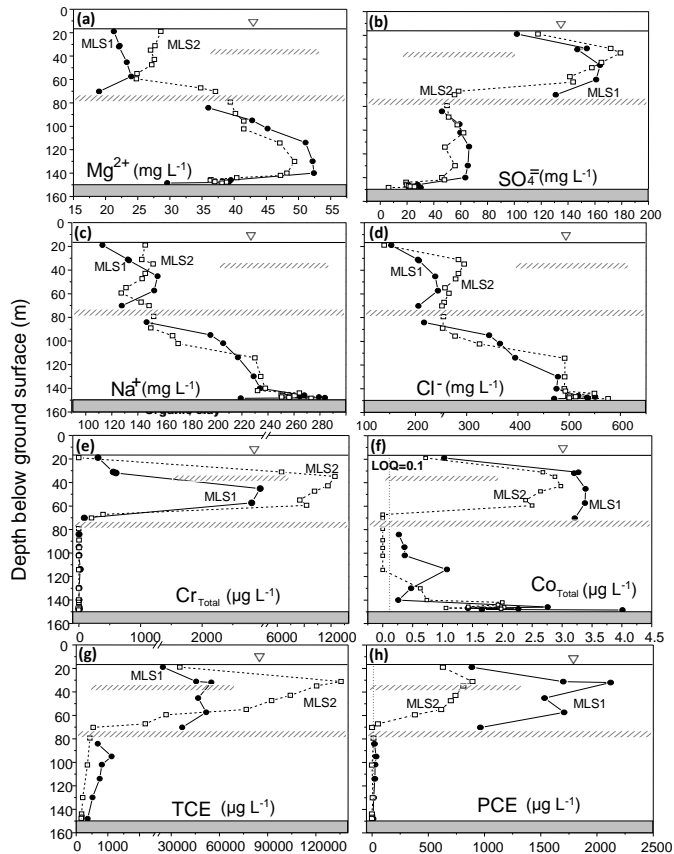


Fig. 3. Representative deep aquifer profiles obtained in sequential sampling campaigns: **(a–d)** – major ions; **(e, f)** – trace elements and **(g, h)** VOC's. MLS1 (circles) obtained 10 August 2008; MLS2 (squares) – obtained 1 June 2009. Saqiye Clay in gray is the bottom aquifer boundary and clayey layers around 35 and 75 m depth are illustrated in dash lines (Fig. 1). LOQ – limit of quantification.

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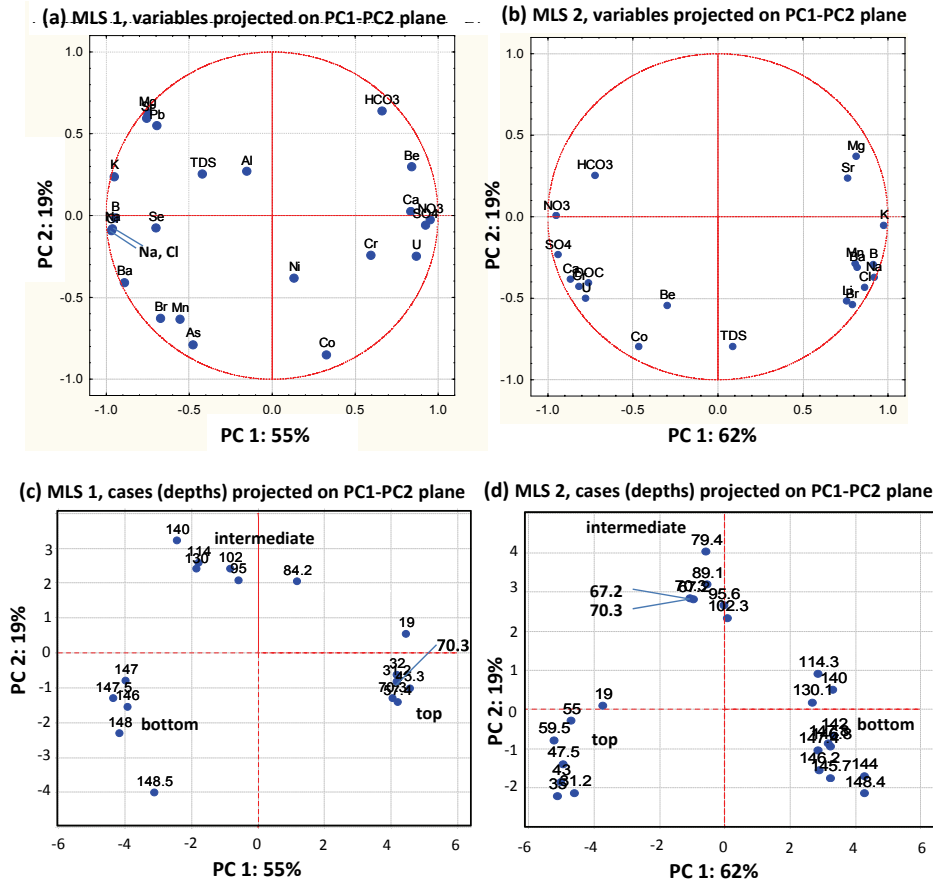


Fig. 4. PCA of the MLS campaigns. **(a, b)** Variables and **(c, d)** cases projected on the plane of the largest two principle components. Cases are labeled by the sampling depth in m below ground surface. Top, intermediate and bottom groups of cases are distinct. Oxidation states of ions were omitted for graphical simplicity.

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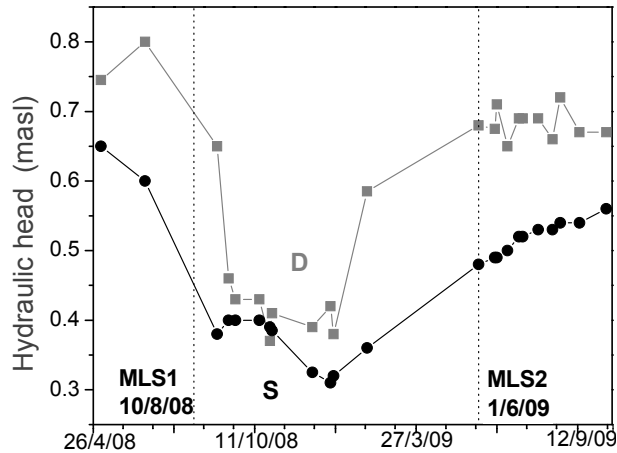


Fig. 5. Elevation of the hydraulic heads observed in the deep sub-aquifer in gray (measured in pipe D, Fig. 1) and the shallow sub-aquifer (measured in pipe S, Fig. 1). The dotted vertical lines denote the retrieval dates of MLS1 and MLS2. masl – meters above sea level.

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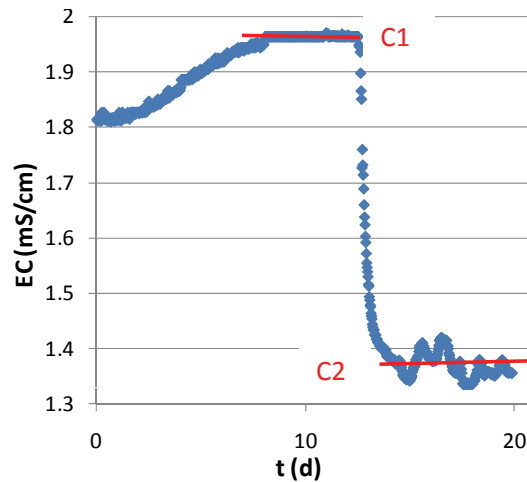


Fig. 6. EC monitored in a 35 cm packed-off section of the observation well at 31.5 mbgs. Note the sharp change in EC at day 12, suggesting a relatively fresh water body flowed through the monitored section.

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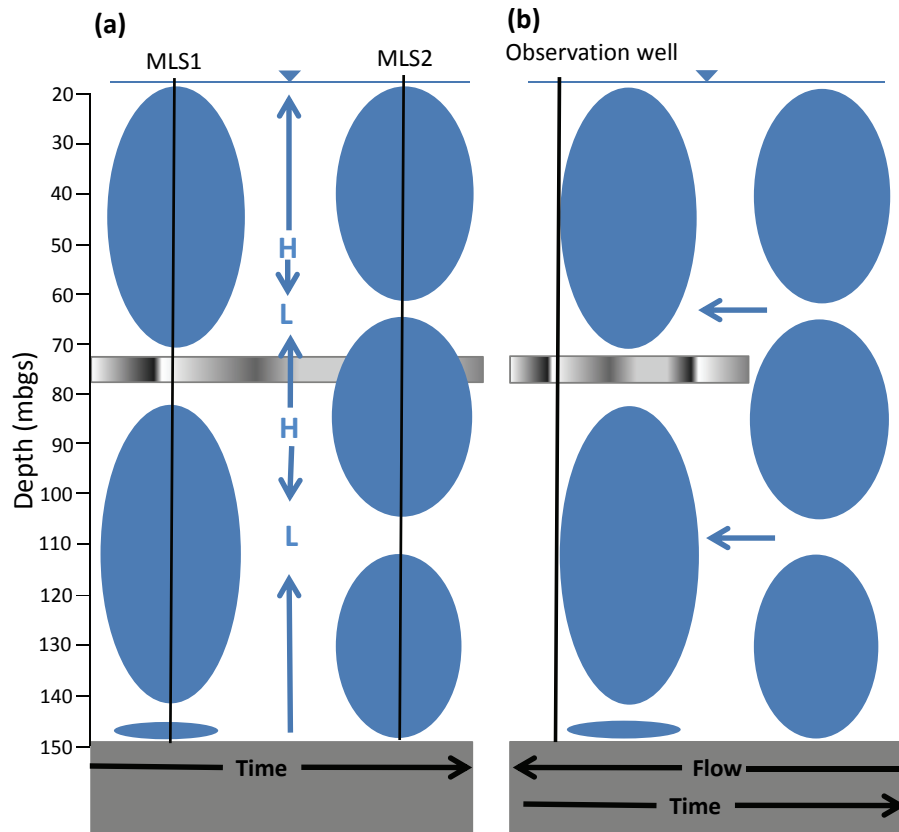


Fig. 7. Considering possible flow regimes in the vicinity of the observation well. Blue ellipses resemble the water bodies inferred in the PCA of MLS1 (left) and MLS2 (right). **(a)** Vertical-dominant flow regime at the time between samplings that may support the differences in the vertical distribution of the water bodies observed in the two campaigns. L and H are relative low and high hydraulic heads. **(b)** Lateral flow of water bodies passing through the observation well as an alternative explanation of the differences.

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