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Impact of the industrial rejections on water of Annaba aquifer (Algeria)

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Abstract: The characterization of a hydrologically complex contaminated site bordering the Meboudja wadi (Annaba, Algeria) was undertaken by investigating surface water and groundwater affected by numerous of industrial rejections have been established during 1999-2005 in the El-Hadjar Industrial Development Area. The untreated effluents from the industries are being discharged in Meboudja wadi. Groundwater level and water quality monitoring was carried out during this period in El-Hadjar region. The groundwater shows a high electric conductivity, high chloride and sodium contents and high metallic concentrations are observed for the wells located down gradient and near the industrial rejections. The Meboudja wadi is acting as a diffuse source of contaminations all along its course. Groundwater flow and mass transport models were prepared using visual MODFLOW software. The extent of migration of contaminants from the Meboudja and others streams has been assessed for 6 years.

Keywords: Pollution; Chromium; Industrial rejections; Numerical modelling; Annaba - Algeria

1. INTRODUCTION

Increase of waste production is correlated with economical and demographical development. While life improvement is expected, such development also leads to negative effects on the environment and economy of many countries. The demographical development and the intensification of the economical activities in Algeria are accompanied by an increase of solid waste production (Kherici, 1993; Djabri, 1996; Debièche et al., 2003).

The studied zone, that is expected to be the third industrial node of Algeria, has experienced an intensification of the demography (higher than 800000 inhabitants) and economy (more than 150 industrial units). Environmental problems such as air and water pollution could seriously set back this economical and urban development. Indeed, several hundreds of tones/day of solid and liquid wastes are dumped in the environment without any treatment. This uncontrolled dumping has negative effects that are clearly identified such as nauseous smells, smoke generation, pollution of the water table and the soil (Debièche et al., 2003; Hani, 2003).

The industrial effluents contain appreciable amounts of inorganic and organic chemicals and their bye-products. Most of the industries are in small scale sector and are not having any sewer lines. Even today most of them don't have proper wastewater treatment plants and they discharge industrial effluents in unlined channels and streams and thereby causing enormous contamination of air, water and soil. As a result, the highly coloured and toxic chemical effluents join the Meboudja wadi, which is a left-side tributary of the Seybouse River, polluting the surface water and groundwater.

The degree of contamination has been so intense that in some parts the environment has become unsuitable for human living. Traditionally the people of the lower Seybouse plain are an agricultural community. None of the industrial units took any measures for safe disposal of industrial effluents till 2000. Indiscriminate dumping of wastes all over the place became a routine practice. Soon the levels of toxic elements in soil, water and air drastically exceeded the permissible limits. Crop production fell drastically and aquatic life in the Seybouse stream started perishes. The Seybouse has become highly polluted. In fact, it no longer looks like a water body; what remains is dark, greasy, frothing mass of thick liquid in the summer. Annaba was not like this 20 years ago. Experts attribute the present situation to the lack of planning and mindless sitting of industries (Japanese Agency of International Office of Survey for the Countryside Development, in Zenati, 1999).

In order to determine the impact of these effluents on the quality of the wadi water and groundwater, a monthly monitoring of water chemistry was performed in the wadi and in the water catchment, situated lower Seybouse plain.

In the present study, groundwater monitoring has been taken up for effective assessment through understanding of hydrogeology, geology and water-chemistry of the watershed. The collected basic data is used for the preparation of the groundwater flow and mass transport model for quantitative assessment of impact of contaminant migration in the watershed. To determine the groundwater velocity distribution is used to analyze advective and dispersive transport to determine contaminant migration in the area.

2. STUDY AREA

The studied area is situated in the lower Seybouse plain (NE Algeria) upstream of the Meboudja wadi (Fig. 1). The northern limit of the zone is constituted by the metamorphic basement, whereas the other limits are open limits which are in continuity with the shallow aquifer of the low-lying Seybouse plain.

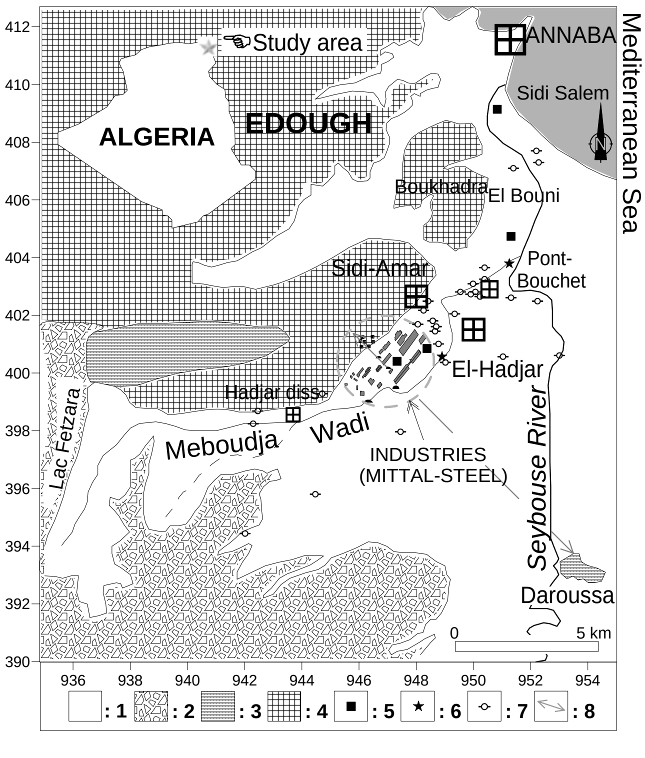

Figure 1. Location map of study area: 1: Undifferentiated Quaternary, 2: Ancient alluvium, 3: Numidian sandstone or clay, 4: Metamorphic formation, 5: Soils sampling locations, 6: Surface water sampling, 7: Superficial aquifer sampling wells, 8: Cross section.

Meboudja wadi is characterized by a permanent flow in winter. Its alimentation comes from rain waters, and drainage of the lake Fetzara (the discharge reaches 16 m^3s^{-1}). During the summer, the water inflows are mainly the outputs of the lake (the flow rate ranges between 1 and 5 m^3s^{-1}). The wadi receives also urban contributions upstream, such as domestic sewage waters. The aquifer reservoir is developed on a clayey substratum. The aquifer formations are represented by 70 loamy sand and 30 clays (Japanese Agency of International Office of Survey for the Countryside Development, in Zenati, 1999). The average permeability ranges between 10^{-3} and 10^{-4} m s^{-1}.

The climate is of Mediterranean type with an annual rainfall of 650 mm, a mean temperature of 18°C and high atmospheric humidity. The dominating wind direction (Northwest-Southeast) blows from the studied area towards the region of Drèan. The effective infiltration is about 15% of the total rainfall that is 100mm per year, which infiltrates through waste, soil and finally to the water table (Hani, 2003, Debieche, 2002).

3. MATERIAL AND METHODS

On the basis of hydrogeological schematization of the site, a sampling strategy was developed in order to obtain the experimental data needed to achieve the characterization objective, i.e. to determine the extent and routes of contamination caused by the Industrial rejections on the surrounding soils and the underlying groundwaters.

Many monthly surveys of the piezometric level and geochemical analysis have been monitored from 1999 to 2005. The analyses are carried out on a network of 30 wells surrounding the Mittal steel industries (Fig. 1). Some sampled wells are used by the neighbouring population for daily drinking, irrigation and animal alimentation. The grounds were taken according to a direction south-north.

The temperature (T), electrical conductivity (EC) and pH were measured in situ using a multiparameter WTW set (Multiline P3 PH/LF SET), an Oxymeter (WTW) with an oxygen probe (CellOx 325) for the measurement of dissolved oxygen. The concentration of chloride (Cl^-), sulfate (SO_4^{2-}), calcium (Ca^{2+}), magnesium (Mg^{2+}), carbonates (HCO_3^-), were determined using the volumetric method (AFNOR, 1987). Nitrate (NO^{3-}) and ammonium ($NH4^+$) were analysed by colorimetry method using spectrophotometer (Spectronic 20 D). The heavy metals (Fe, Zn, Mn, Cu, Cd, Cr) were determined using atomic absorption spectrophotometer (Unicam 929 AA Spectrometer).

In the present study, groundwater monitoring has been taken up for effective assessment through understanding of hydrogeology, geology and water-chemistry of the watershed. The collected basic data is used for the preparation of the groundwater flow and mass transport model for quantitative assessment of impact of contaminant migration in the watershed. To determine the groundwater velocity distribution is used to analyze advective and dispersive transport to determine contaminant migration in the area.

4. RESULTS AND DISCUSSION

The industrial effluents contain appreciable amounts of inorganic and organic chemicals and their bye-products. Most of the industries are in small scale sector and are not having any sewer lines. Even today most of them don't have proper wastewater treatment plants and they discharge industrial effluents in unlined channels and streams and thereby causing enormous contamination of air, water and soil. As a result, the highly coloured and toxic chemical effluents join the Meboudja, a natural water course, polluting soil, surface water and groundwater. In soil, this study highlights a decrease of Chromium concentration from the MITTAL STEEL to the sea (Sidi-Salem) (Fig. 2). The metallic element concentrations exceed significantly the OMS standard for potable water. These high concentrations are linked to the contamination by the industrial rejections, which are high in heavy metals. Other factors favor the metallic groundwater contamination.

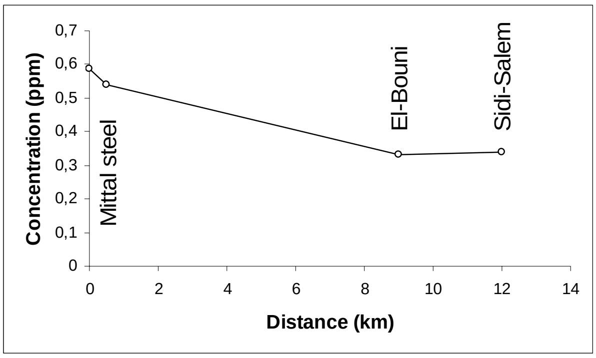

Figure 2. Distribution of Chromium concentration in soil from the MITTAL STEEL to the sea.

Table 1 shows that a high electric conductivity (more than 560 cm^{-1} with a maximum exceeding 20000 μScm^{-1}), a high chloride content (with a maximum exceeding 6000 mgl^{-1}), and a high sodium concentration (mean = 420 mgl^{-1}) are observed for the wells located down gradient and near the industrial rejections. Also, high metallic concentrations (0.02-1.25 mgl^{-1} in chromium) are observed in these wells. The statistics of results obtained by chemical analysis of the groundwater samples are summarized in Table 1. The 32 remaining parameters (Table 1) were treated according to the principal component analysis (PCA).

Table 1. Mean values, minimum (min), maximum (max) and standard deviations (Sd) of the parameters measured in superficial aquifer (2005). Mean, minimum (min), maximum (max) and Standard deviation (Sd). Temperature (°C), electric conductivity ($\mu S.cm^{-1}$), Eh (mV) and concentrations ($mg.L^{-1}$)

Variables	Min	Max	Mean	Sd	Variables	Min	Max	Mean	Sd
T	19.8	25.7	22.9	1.64	NO_3	0	296	43.85	64.12
pH	6.88	8.4	7.4	0.31	NO_2	0.07	0.73	0.25	0.21
Eh	238	416	356	32	PO_4	0.11	1.72	0.50	0.45
Si	4.2	12.2	7.16	1.95	NH_4	0	4.5	0.18	0.77
Ca	44	746	209	156	CE	558	20333	3222	3592
Mg	12	495	78.5	94.3	Cr(T)	0.02	1.25	0.07	0.21
Na	35	3670	419.8	642.6	Mn	0	2.48	0.13	0.46
K	1	56	8.7	13.2	Fe(T)	0.01	2.81	0.22	0.61
Cl	48	6274	658	1148	Ni	0.02	0.55	0.1	0.10
SO_4	60	883	204	147	Sn	0	0.24	0.03	0.04
HCO_3	188	610	416	108	Al	0	0.06	0.02	0.01
F	0.05	1.62	0.62	0.40	Sr	0.3	6.8	1.5	1.3

4.1. Application of Principal Component Analysis to groundwater contaminant

PC 1 explains 32% of the variance and is contributed by conductivity, sulphate, calcium, magnesium, chloride, sodium and strontium (Fig. 3). PC 2 explains 15% of the variance and is mainly participated by chromium and tin. We can see that PC 1 contain classical hydrochemical variables originating, at a first glance, from mineralization of geological components of soils, whereas the other PC 2 is contributed by nitrate and heavy metals, i.e. by species whose presence at high concentration levels can be attributed to human action: nitrate may originate from fertilisers (both agricultural and industrial), and heavy metals from leachates of industrial wastes.

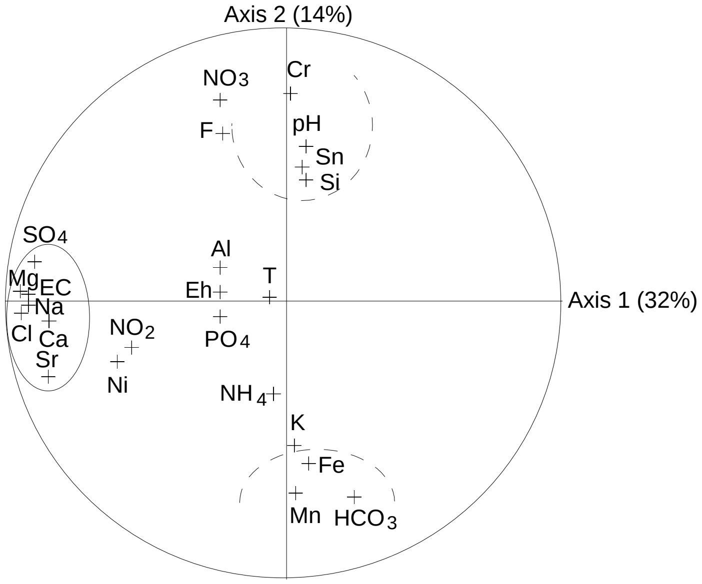

Figure 3. Results of PCA.

4.2. Groundwater Flow and Mass Transport Modelling

Groundwater contamination is limited with the mass transport processes. These processes determine maximum extent of plume spread and geometric character of the concentration distribution. Advection is by far the most dominant mass transport process in contaminant migration. Hydrodynamic dispersion is usually a second order process. The magnitude and direction of advective transport is controlled by the configuration of water table or piezometric surface, presence of sources or sinks, permeability distribution within the flow filed and flow domain.

The hydraulic head is obtained from the solution of a three dimensional groundwater flow equation (Bear, 1972):

$$\frac{\partial}{\partial \chi i}\left[K_{ii}\frac{\partial h}{\partial \chi j}\right]+q_s=S_s\frac{\partial h}{\partial t} \qquad (1)$$

where S_s is the specific storage of the porous material.

The transport equation is linked to the flow equation through groundwater velocity term given by:

$$v_i=\frac{K_{ii}}{\theta}\frac{\partial h}{\partial \chi i} \qquad (2)$$

where:

K_{ii} a principal component of the hydraulic conductivity tensor;
h hydraulic head;
θ the porosity of the porous medium.

MODFLOW software (McDonald and Harbaugh, 1988) is used for the hydraulic simulation. All these parameters are important in controlling the groundwater velocity, which drives advective transport. Adding dispersion to advective transport can cause important changes in the shape of a plume. Another important process is sorption and irrespective of the model describing sorption, the process was of paramount importance in controlling contaminant transport. The partial differential equation describing three-dimensional transport of contaminants in groundwater (Javandel et al., 1984) can be written as:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial \chi i}\left[D_{ij}\frac{\partial C}{\partial \chi j}\right] - \frac{\partial}{\partial \chi i}(v_i C) + \frac{q_s}{\theta}C_s + \sum_{K=1}^{N} R_k \tag{3}$$

where:

- C the concentration of contaminants dissolved in groundwater;
- t time;
- χi the distance along the respective Cartesian co-ordinate axis;
- v_i the hydrodynamic dispersion coefficient;
- v_i the seepage or linear pore water velocity;
- q_s the volumetric flux of water per unit volume of aquifer representing sources (positive) and sinks (negative);
- C_s the concentration of the sources or sinks;
- θ the porosity of the porous medium;
- R_k chemical reaction term.

Assuming that only equilibrium controlled linear or non-linear sorption and first order irreversible rate reactions are involved in the chemical reactions, the chemical reaction term can be expressed as (Grove and Stollenwerk, 1984):

$$\sum_{k=1}^{N} R_k = -\frac{\rho_b}{\theta}\frac{\partial \overline{C}}{\partial t} - \lambda\left[C + \frac{\rho}{\theta}\overline{C}\right] \tag{4}$$

where

- $\overline{C}$ the concentration of contaminants sorbed on the porous medium;
- λ the rate constant of the first-order rate reactions;
- ρ_b the bulk density of the porous medium.

Rewriting Equation (4) as:

$$\frac{\rho_b}{\theta}\frac{\partial \overline{C}}{\partial t} = \frac{\rho b}{\theta}\frac{\partial C}{\partial t}\frac{\partial \overline{C}}{\partial C} \tag{5}$$

We can rewrite Equation (3) by substituting Equations (4) and (5) as:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial \chi\, i}\left[D_{ij}\frac{\partial C}{\partial \chi\, j}\right] - \frac{\partial}{\partial \chi\, i}(viC) + \frac{q_s}{\theta}C_s - \frac{\rho\, b}{\theta}\frac{\partial C}{\partial t}\frac{\partial \overline{C}}{\partial C} - \lambda\left(C + \frac{\rho\, b}{\theta}\overline{C}\right) \tag{6}$$

Rearranging the terms we get the governing equation of mass transport model:

$$R\frac{\partial C}{\partial t} = \frac{\partial}{\partial \chi\, i}\left[D_{ij}\frac{\partial C}{\partial \chi\, j}\right] - \frac{\partial}{\partial \chi j}(v_i C) + \frac{q_s}{\theta}C_s - \lambda\left(C + \frac{\rho\, b}{\theta}\overline{C}\right) \tag{7}$$

where R is the retardation factor, defined as:

$$R = 1 + \frac{\rho b}{\theta}\frac{\partial \overline{C}}{\partial C} \tag{8}$$

4.3. CONCEPTUALIZATION OF FLOW AND TRANSPORT PROCESSES

Estimation of aquifer parameters is essential for quantifying the groundwater resources and also to determine well characteristics. Pumping tests were carried out on 10 wells dug wells. High transmissivity values were obtained in alluvial formations, in spite of limited aquifer thickness. The permeability values as high as 10^{-4} m s^{-1} are found in the alluvium of the Seybouse around El-Hadjar village and intensive groundwater irrigation has resulted in stream aquifer interaction.

The surface water while seeping through the Meboudja wadi stream-bed carries effluents to groundwater regime, thereby contaminating groundwater up to a distance of 600 m from the wadi (Fig. 4). The most important process contributing to the mass transport in groundwater is advection. Longitudinal dispersion is relatively significant but transverse dispersion could be neglected. The chromium concentration was chosen for a detailed mass transport model study because (a) it has shown consistent concentration of effluent ranging between 1.25-0.06 mg l^{-1} along different reaches of the Meboudja wadi and (b) uniform background level of about 0.02 mg l^{-1} in native groundwater. The following information was used during conceptualization of groundwater flow regime:

- The Meboudja stream behaves as affluent and influent along different passages;
- No flow occurs across the watershed boundaries as the boundaries are coinciding approximately with groundwater divide;
- Groundwater recharge rainfall occurs from the top layer;
- The continuous groundwater pumping in the alluvium has caused lowering of the groundwater levels below the stream bed, thereby resulting in induced seepage from the Meboudja wadi, particularly in lower regions;
- The chromium concentration of groundwater as well as surface water effluent at some places indicated higher levels. The chromium concentration of groundwater was found almost equal to surface water chromium concentration of the Meboudja wadi around Sidi Amar village;
- Contaminants enter the groundwater regime through seepage of effluent from the Meboudja stream acting as diffuse sources all along its traverse.

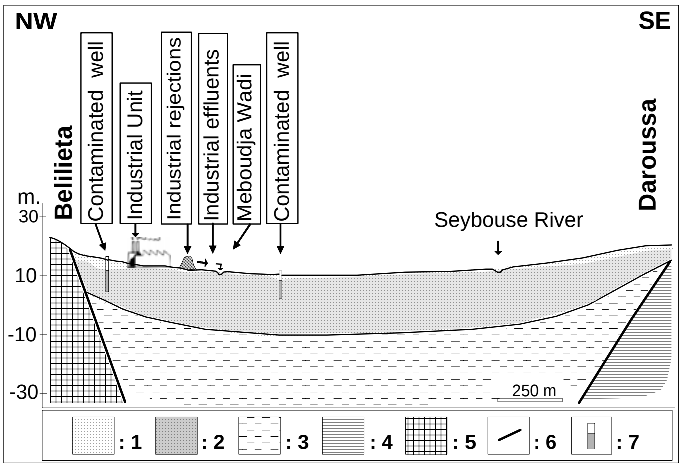

Figure 4. Cross section of low plain of the Seybouse, near Mittal steel industries. 1: Alluvium (unsaturated zone), 2: Alluvium (aquifer), 3: Clays, 4: Numidian sandstone or clay, 5: Metamorphic formation, 6: Faults, 7: Wells.

4.4. Groundwater Flow Model

The numerical approaches for solving mass transport equations are approximate forms of the advection-dispersion Equation (7) as a system of algebraic equations or alternatively simulating transport through the spread of a large number of moving reference particles (particle tracking). The second step is to provide boundary condition and assign values of concentration or loading rates defining various boundaries for all nodes located along boundary of the domain. Initial conditions and transport parameters were specified for all nodes. The seepage from the Meboudja wadi was simulated by giving additional recharge input to the model. Constant concentration was assigned in different parts of the Meboudja wadi based on ambient surface was as well as groundwater concentrations measured during field investigations. The water level data of February 1999 is considered February 2005 for solving groundwater flow equation under steady state conditions and thereby a single velocity field was determined for the mass transport simulation for all times.

The simulated model domain consists of 40 rows and 40 columns and 1 layer covering an area of 8000 x 8000 m. The superficial aquifer mostly consists of a 10-15 m thick alluvium along the Meboudja wadi. The simulated vertical section has a maximum thickness of 15 m. The groundwater recharge at the rate of 100 mm yr^{-1} has been simulated in the top layer. Continuous seepage from the Meboudja stream was simulated as additional recharge in the model. The first stage of modelling is flow simulation for computation of hydraulic head distribution. The distribution of hydraulic head and hence the velocity field is unaffected by migration of plume because density and viscosity of contaminated groundwater is nearly the same as uncontaminated water in the area. The flow equation was therefore, first solved independently of the mass transport equation. Further, water level observations in the area indicate that hydraulic gradients do not change significantly with time. Thus groundwater flow was assumed to be in a steady state and the groundwater heads were computed by visual MODFLOW (Guiger and Frantz, 1996) using Slice Successive Over Relaxation (SSOR) package (McDonald and Harbaugh, 1988). The flow model was calibrated by adjusting several parameters within a narrow range of values until a best fit was obtained between observed data and simulated results. The accuracy of the computed water levels (Fig. 5) was judged by computing mean error, mean absolute error and root mean squared error computed for 15 observation wells. The calculated mean error, mean absolute error and root mean squared error under steady state condition is -0.14, 3.2 and 3.6 m, respectively.

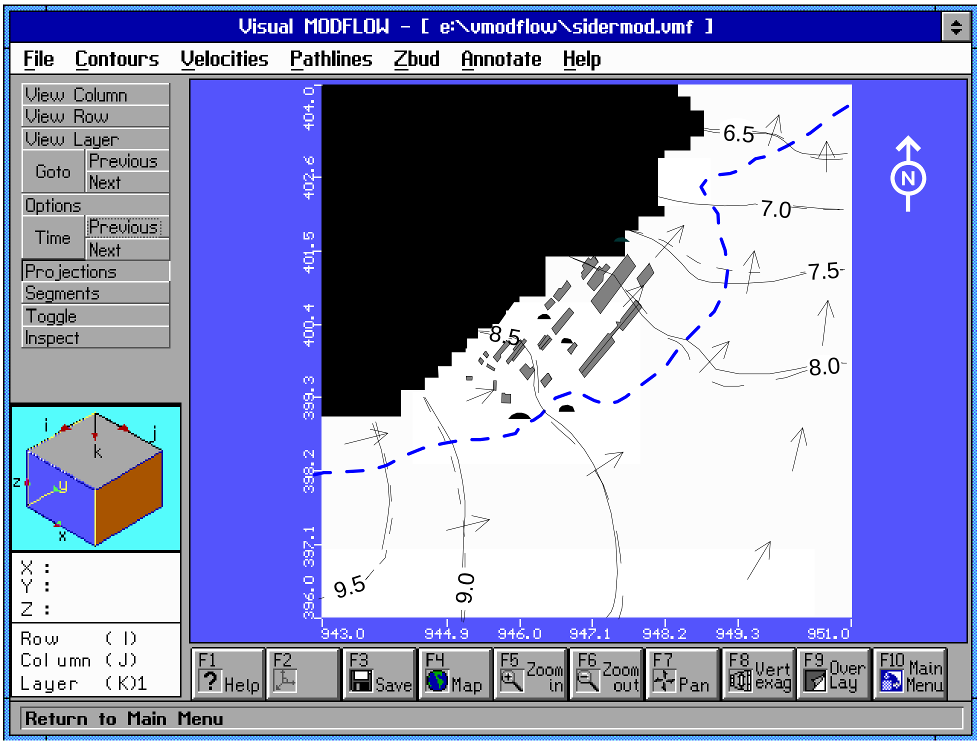

Figure 5. Distribution of hydraulic heads for February 2005. Dashed lines, simulated; continuous lines, measured. contour interval, 0.5 m.

4.5. Mass Transport Model

Mass transport in three dimensions (MT3D) is a computer model for simulation of advection, dispersion and chemical reactions of contaminants in three-dimensional groundwater flow systems

(Zheng, 1990). The model is used in conjunction with a block-centred finite difference flow model MODFLOW. Dispersion was accounted for the particle in motion by adding to the deterministic motion a random component, which is a function of dispersivities. The mean concentration for each grid block was calculated as the sum of the mass carried by all the particles located in a given block divided by the total volume of water in the block. The values of dispersivity in longitudinal and two transverse directions (Y and Z) were assumed to be 10, 1 and 0.1 m, respectively, and the values were taken from the literature (Kimbrough et al., 1999; Domenico and Schwartz, 1990; Tevissen, 1993). The tendency for α_L to be about 10 times larger than α_{TH} and for α_{TZ} to be much smaller than either of them is in line with the concentrations observed in the area. The initial chromium concentration assigned in the rest of the area is about 0.02 mg l^{-1}. The relatively smooth decline of chromium concentration away from the Meboudja wadi suggests a relatively constant rate of loading. Thus a constant chromium concentration at different nodes on the Meboudja wadi was assigned varying from 1.25 mg l^{-1} at source (Mittal steel) near the Sidi Amar and 0.06 mg l^{-1} away from the Mittal steel at about 7 km downstream of the Meboudja wadi. The computed iso-concentration contours for 2005 indicate that the plume is expanding and follows the hydraulic gradient implying that advection is the dominant mechanism of spreading.

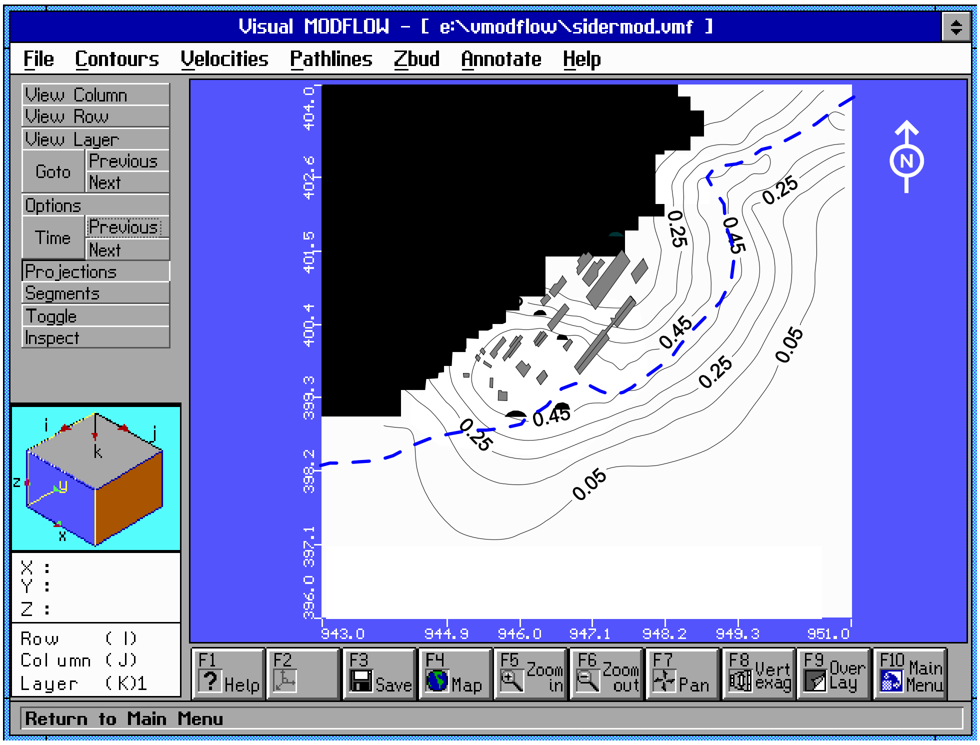

Figure 6. Computed Chromium concentration (mg L^{-1}) of groundwater for 2005 in the lower Seybouse plain.

The extent of contaminant migration from the Meboudja stream could be seen by computed iso-concentration of chromium contours of transport model for the period of 1997-2005. The contaminant migration was found expending up to 600 m from the Meboudja wadi during the last 5 yr (Fig. 6). Inaccuracies in the simulated flow field could have existed, which produced somewhat more divergent flow pattern from what actually exists. Because we have given uniform pumping rates for the wells and diffuse source concentration at all nodes of the Meboudja wadi, this problem could be related to the complex interaction between groundwater and surface water.

4.6. Development of the contaminated site conceptual model and exposure diagram

The available overall information gained on soil and groundwater contamination was used to develop the site conceptual model and to identify the exposure scenarios, which are the main outcomes of the site characterization, according to the terminology of risk assessment procedures (US-EPA, 1989; ASTM-RBCA, 1998; CARACAS, 1998).

The combination of the hydrogeological conceptual model and the results of soil and groundwater contamination allowed to obtain the conceptual model showed in Fig. 7 which summarizes the site information on hydrogeology and type and extension of the chemical contamination.

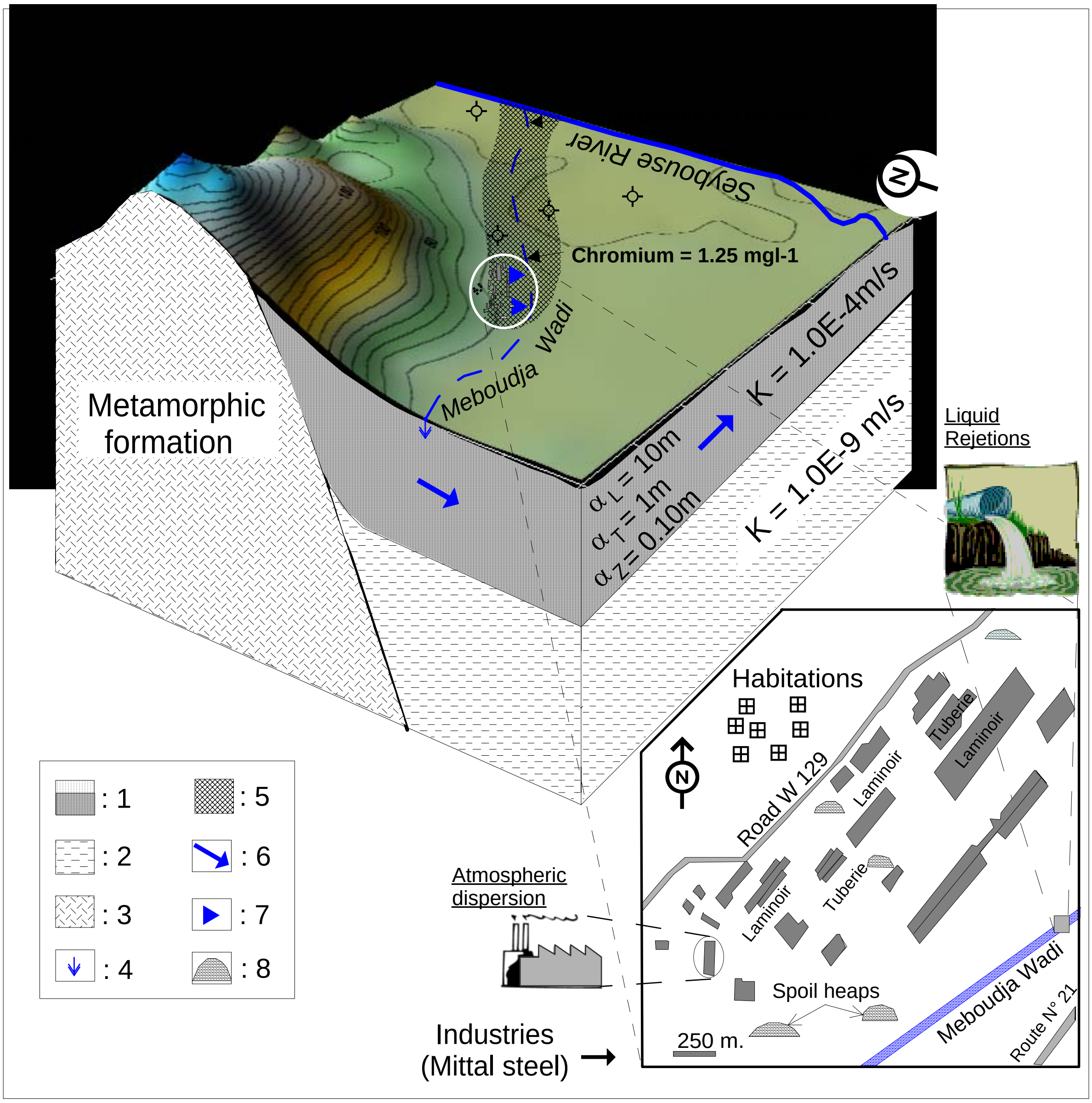

Figure 7. Conceptual model of lower Seybouse plain, near Mittal steel industries.

Examination of Fig. 7 triggers a special attention on: (a) the heavy contamination by heavy metals in the surface and sub-surface soil surrounding the Mittal steel industries; (b) the contaminants percolation from the Mittal steel industries (primary exposure source) to reach the surrounding soil (secondary exposure source) under the complex hydrogeological condition of the surface aquifer; (c) the contamination of the surface and superficial aquifer.

Finally, the obtained results were used to construct the exposure diagram showed in Fig. 8, where the receptor (humans, wildlife, ecosystem) were linked to primary and secondary transport mechanisms.

The considered exposure pathways were the inhalation of volatile substances and the direct contact with soil (ingestion and dermal contact) that could threaten either humans or wildlife, on-site or off-site. In addition, the groundwater was considered to be a potential risk pathway, especially for the human, wildlife and ecosystem communicating through the Meboudja water and superficial aquifer.

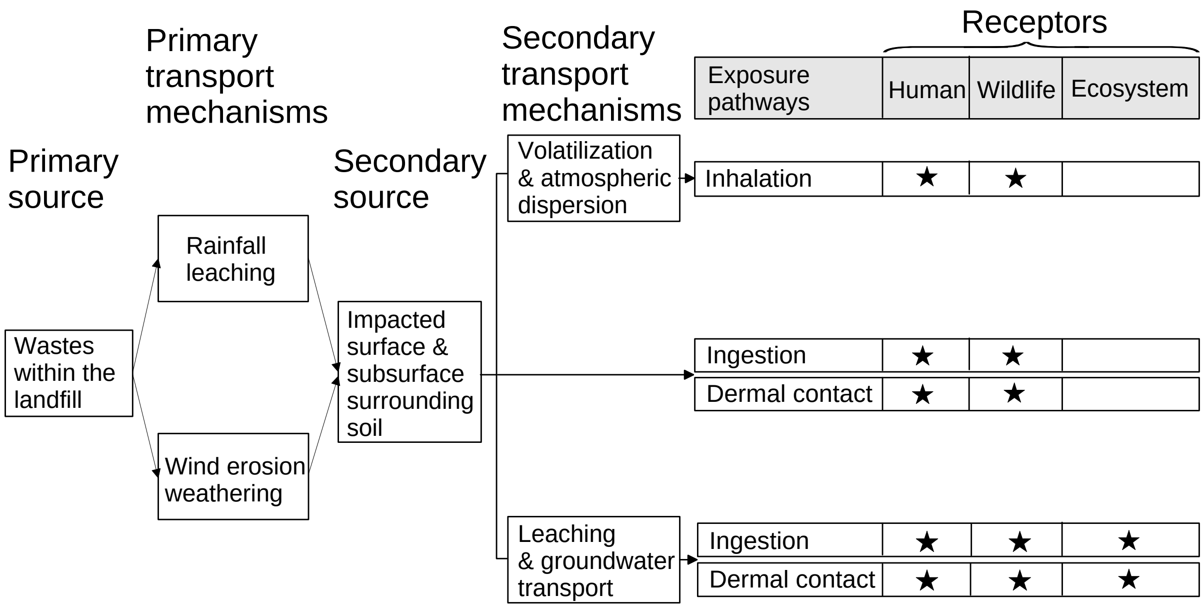

Figure 8. Exposure diagram for the Meboudja contaminated site.

5. CONCLUSIONS

Here a case study of groundwater/surface water pollution due to uncontrolled industrial effluent discharges and its environmental impact on groundwater regime is presented. Groundwater pollution extends laterally 500-600 m from the Meboudja wadi, in which initial pollutants load, in the alluvial areas covering Sidi-Amar and El-Hadjar cities. The extension of pollution is due to heavy pumping for irrigation, resulting in induced seepage from Meboudja wadi due to stream aquifer interaction, which in turn carries surface water effluent to the groundwater regime. The contaminated groundwater is being exploited for agriculture and industrial purposes in the absence of major surface water sources in the area. The modeling study has helped to gain a better insight of the hydrogeologic set up and assessment of contaminant migration. The overall obtained information was sufficient to develop exposure scenarios for risk analysis.

The untreated effluents emerging from the industries must be monitored for maintaining the standards prescribed for Chromium concentration by the environment inspection for various industries in the region. The present study provided a base line data for assessment of contamination in the El-Hadjar area. For reduction of the stream aquifer interaction, the pumping around the Meboudja wadi should be reduced. Periodical monitoring of the water quality has to continued to check the rise in chromium concentrations of groundwater.

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