

Attenuation capacity of a coastal aquifer under managed recharge by reclaimed wastewater

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ABSTRACT

Managed Aquifer Recharge (MAR) is becoming an increasingly attractive water management option, especially in semi-arid areas. Nevertheless, field studies on the fate and transport of priority substances, heavy metals and pharmaceutical products within the recharged aquifer are rare. Based on the above, the objective of this project is to study the hydrological conditions of the coastal aquifer of Ezousa (Cyprus) and its ability to attenuate pollutants. The Ezousa riverbed is a locally important aquifer used for a MAR project where treated effluent from the Paphos Waste Water Treatment Plant is recharged into the aquifer through a number of artificial ponds along the riverbed. Additionally, groundwater is pumped for irrigation purposes from wells located nearby. The hydrological conditions of the area are unique due to the construction of the Kannaviou dam in 2005 that reduced natural recharge of the Ezousa aquifer significantly, inducing the saltwater intrusion phenomenon. A three-dimensional finite element model of the area was constructed using the FEFLOW software to simulate the groundwater flow conditions and transport of Phosphorous and cooper in the subsurface from the recharge process. The model was calibrated using hydraulic head and chemical data for the time period of 2002-2011. The groundwater model was coupled with a geochemical model PHREEQC attempting to evaluate nitrate and Copper processes. Inverse modeling calculation was used to determine sets of moles transfers of phases that are attributed to the water composition change in groundwater between the mixture of natural groundwater and reclaimed wastewater and the final water composition.

Keywords: groundwater flow and transport, FEFLOW, phosphorus, cooper, PHREEQC, managed aquifer recharge, coastal aquifer

1. INTRODUCTION

Managed Aquifer Recharge (MAR) is becoming one of the most attractive option in semi-arid areas avoiding seawater intrusion and buffering seasonal variation of water supply and demand. For instance, in Israel operates the largest wastewater re-use project, where the coastal aquifer accepts the secondary effluent (95 Mm³/year) of Tel Aviv (1.5 million inhabitants)¹. After approximately two months of transport and processing within the aquifer the recycled effluent is used for irrigation purposes. In Spain, in Sabadell city, 6.9 Mm³/year is the recovered water used in agriculture gained by Ripoll river infiltration of secondary effluent into the aquifer and in Italy, in Nardo city, a water volume of 4.4 Mm³/year is recovered by direct injection via sinkhole in a confined and fractured aquifer. Even though MAR is an attractive option to store water and to improve reclaimed wastewater quality, data from field studies on the fate and transport of priority substances (according to 2008/105EU Directive,) heavy metals and pharmaceutical personal products within the aquifer are very rare^{2,3}. It is well documented that heavy metals such as Cd, Hg, Ni, Pb present in wastewater are attenuated significantly within the aquifer (for instance Ni was decreased from 28.2 mg/L of a tertiary treatment effluent up to 12.1 mg/L through its pass into the aquifer at Depurbaix facility in Barcelona, Spain³. On the other hand, some pesticides are not decreased substantially for instance Diuron (a widely used pesticide) with an influent content of 30.7 ng/L, it is still detected in the groundwater in a range about 24.6 ng/L.

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Despite the relatively high number of MAR schemes and the ordinary monitoring of recovered water, there is no research or methodology developed concerning the potential of the aquifer matrix to retardate, sorb or deactivate the priority substances, heavy metals and nutrients. However many factors, such as soil properties, dilution factors, hydraulic conditions, organic matter content, and microbial load affect the reduction of pollutants from the injected to the recovered water in the aquifer and their role should be determined in the water quality improvement. Thus, the ultimate objective of this project is to study the buffering capacity of aquifer soil matrix to attenuate pollutants. Pollutants of concern are heavy metals and nutrients. The suggested methodology includes laboratory studies to evaluate the effect of aquifer matrix characteristics in heavy metals and nutrients partitioning coefficient. A reactive transport model will be developed based on mass balance equations and new reactions will be introduced to adapt the model. A three-dimensional finite element model of the Ezousa riverbed aquifer was constructed using the FEFLOW software that simulated the groundwater flow conditions and the transport of phosphorus and Copper in the subsurface, emanating from the recharge process. The model was calibrated using hydraulic head and chemical data from the existing monitoring network for the time period of 2007-2011. The groundwater model was coupled with the geochemical model PHREEQC attempting to evaluate nitrate, phosphate and copper transport processes and, by inverse modeling calculation, to determine sets of moles transfers of phases that attribute to the observed change in water chemical composition due to mixing of natural groundwater and reclaimed wastewater.

2. METHODOLOGY

2.1 Site description

2.1.1 Ezousas Recharge Project

The Ezousas Recharge Project is located in the last section of the Ezousas river, downstream of Kanaviou dam, close to Paphos, in the South-West part of Cyprus (Fig.1). The Ezousa Recharge Project (ERPr.) is designed to supplement the aquifer with 9000-12000 m³/day of treated wastewater effluent from Paphos. The tertiary treated effluent is distributed via a 500 mm pressured main to infiltration ponds about 10 km east of Paphos. The recharge network consists of 23 infiltration ponds organized in groups of two to six⁴. The infiltration area of each pond is approximately 2000m² and 1.5m below ground surface. Controlled abstraction takes place in the lower reaches of the aquifer downstream, thus providing a semi-closed system for the detailed analysis of groundwater quality dynamics. The design and percolation rate (0.6 m/day) is to allow the basins to accept all projected wastewater reuse during the non-agricultural season; about 3 M m³ is abstracted annually. Equivalent annual infiltration or "hydraulic loading rates" typically vary from 30 m/year to 80m/year, depending on soil, climatic conditions prevailed, quality of sewage effluent, and abstraction rates.

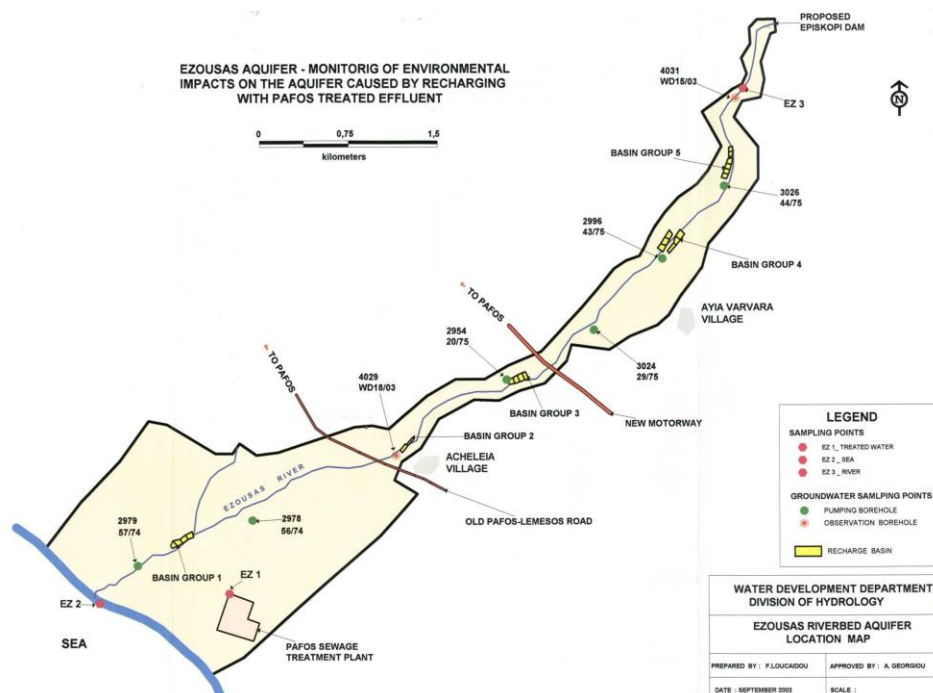


Figure 1. Location of the Ezousas recharge project

2.1.2 Hydrological setting

The Ezousas river flows south from the Troodos mountains to the Mediterranean Sea and covers an area of 239km². The fall is from 1600m to sea level in 30km. Annual precipitation is 300mm along the coast to nearly 1200mm in the Troodos mountains, 80% of the rain falls from November to March. The potential evapotranspiration for the catchment varies from 1060 to 1360mm per year depending on elevation.

The basin can be divided into three distinctive geological zones: an ophiolitic complex in the mountainous north, an overlying sedimentary complex in the south, and Mamonia allochthonous of both igneous and sedimentary rocks juxtaposed elsewhere in the catchment area. The ophiolites include ultramafic rocks, gabbros, sheeted dykes, and pillow lavas, while the sedimentary rocks are mainly chalks, marls, calcarenites, limestones and occasionally gypsums. The latter affects both the surface and groundwater quality which is highly sulphated (>500mg/l) and hard⁴.

The river alluvial deposits in the lower 8 km of the River basin contain an unconfined shallow aquifer which is a regionally important water recourse. The alluvium consists of coarse to fine feldspathic quartz sand, pebbly sand and gravel with few intervening layers of clayey sands and silty clay. Its thickness ranges from 25m upstream to 40m downstream. Transmissivity is variable between 100 to 2000m²/day according to saturated aquifer thickness. Values from storage coefficients derived from pumping tests range from 4 to 13%. Locally, unsaturated hydraulic conductivity (m/day) estimated from mass flux using a Hantush Derivation was 8.8m/day.

The alluvial aquifer is susceptible to saltwater intrusion for up to 2.4 km upstream. The extensive exploitation of the aquifer in the most coastal region, makes the saltwater-freshwater interface extremely volatile. The project for groundwater recharge with recycled water provides an effective barrier against seawater intrusion as well improving water resources.

2.1.3 Monitoring

To investigate the evolution of groundwater compositions in Ezousa project, groundwater from different parts of the project (8 wells, Fig.1) and natural groundwater (well (B/H4031) was sampled. Groundwater and reclaimed wastewater was collected three times per year during 11/2006 – 10/2011 to analyze the biological parameters (BOD₅, COD, Coliforms, *Escherichia coli*, Intestinal Coliforms, Bacteriophages), physicochemical parameters (pH, Conductivity, Total hardness), major anions (Cl⁻, SO₄⁻², HCO₃⁻², F⁻, NO₃⁻², NO₂⁻) and cations (Na⁺, K⁺, Ca²⁺, Mg²⁺, NH₄), Total Phosphorous (TP) and heavy metals (As, Ni, Cd, Cr, Cu, Zn, Pb, Hg, Co, V, Fe, Se, B, Ba). In addition, toxicity tests were carried out MTX EC20, MTX EC50, Daphnia EC50). Pesticide and insecticides residuals in groundwater and wastewater were also identified. Anion and cation analyses were performed by the Cyprus state Chemical Lab in Nicosia, using ion-exchange chromatography and inductively coupled plasma-mass spectrometry, respectively.

2.2 FEFLOW model setup

For the purpose of modeling the MAR process on the coastal aquifer of Ezousa the finite element subsurface flow and transport simulation system FEFLOW was employed. The model was based on a previously calibrated flow model for the period of 2002-2007 [4]. The model presented here was extended until the end of 2011 and contaminant transport was simulated as well.

The horizontal discretization of the unconfined aquifer was implemented using a triangular finite element mesh consisting of 7738 nodes and 6354 elements. The vertical discretization of the model area was based on vertical cross sections created using information provided by boring logs. The conceptual model comprises an unconfined aquifer that is about 40 m deep on the downstream part and 10 m deep on the upstream part and is discretized in 2 numerical layers: a superficial layer of 1 meter thickness that follows the topography of the area to allow definition of infiltration from rainfall, riverbed and artificial recharge and a lower aquifer layer of variable thickness that follows the underlying bedrock morphology.

The hydraulic conductivity of the model was based on available measurements in the area and was modified during the calibration process in order to match the measurements closely. The final distribution is shown in Figure 2. Figure 2 also shows the monitoring and pumping wells of the area along with the locations of the 5 recharging artificial ponds.

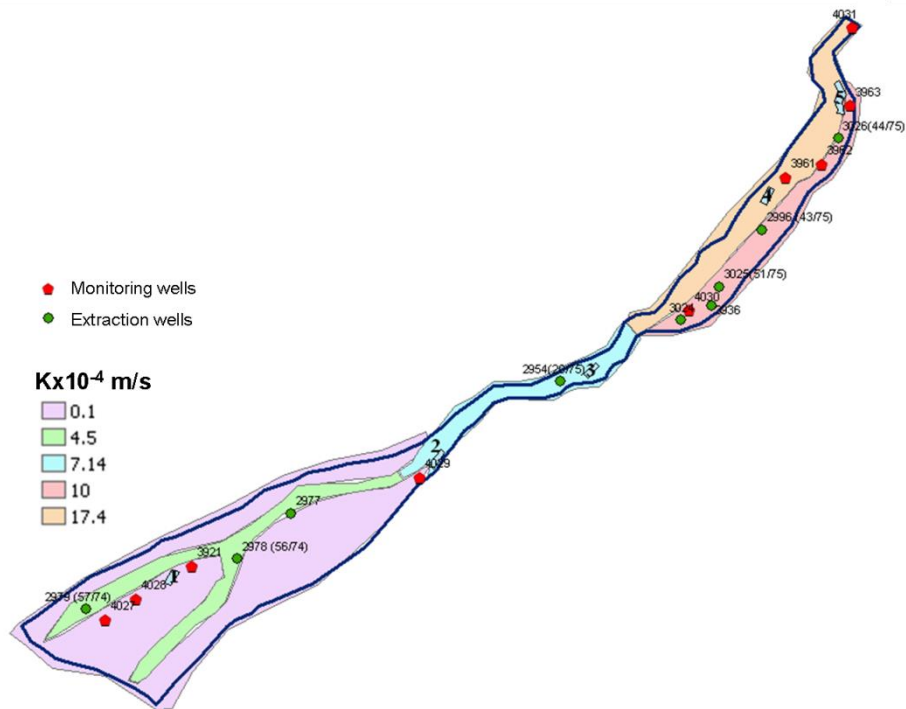


Figure 2. Calibrated hydraulic conductivity and location of extraction and monitoring wells

2.2.1 Boundary conditions for flow

The main types of boundary conditions applied in the model are the following: 1) A first type BC (constant head of 0 masl) on the coastline boundary 2) A first type BC (constant head of 77.1 masl) on the upstream boundary to simulate ambient flow (estimated at approximately at $5 \times 10^5 \text{ m}^3/\text{yr}$) 3) A first type BC (decreasing constant head over a length of 500 meters, varying between 77.1 and 72.7 masl) to simulate river infiltration (estimated at approximately at $2 \times 10^6 \text{ m}^3/\text{yr}$ before and $5 \times 10^5 \text{ m}^3/\text{yr}$ after the construction of the Kannaviou dam) 4) A first type BC (time varying constant head) on the 5 artificial ponds to simulate artificial recharge, as observed on nearby wells. This type of boundary condition was chosen because the amount of effluent recharged in the ponds is not available. 5) A well BCs (flux in m^3/d) to simulate the pumping occurring at the extraction wells. Rain infiltration was assigned as a uniform value on the top layer according to meteorological data from a nearby station. Cases 2 and 3 were linked to a constraint that water may only infiltrated into the system.

The model was divided in three time periods: 1) October 2002 – December 2003 (0-456 days). This period corresponds to the steady state simulation of the model during which no artificial recharge occurs. The coastline, ambient flow and river infiltration BCs are active at this time. 2) January 2004 – July 2005 (456-1002 days). This is the period when the artificial recharge starts and the ambient flow and river infiltration BCs are still active. 3) July 2005 – December 2011 (1002-3375 days). At the beginning of this period the Kanaviou dam was built thus the ambient flow and river infiltration BCs are turned off while artificial recharge and pumping continues.

The information regarding the daily extraction rates for the 9 extraction wells that was introduced in the model was available from January 2004 – December 2011 and is shown in Figure 3.

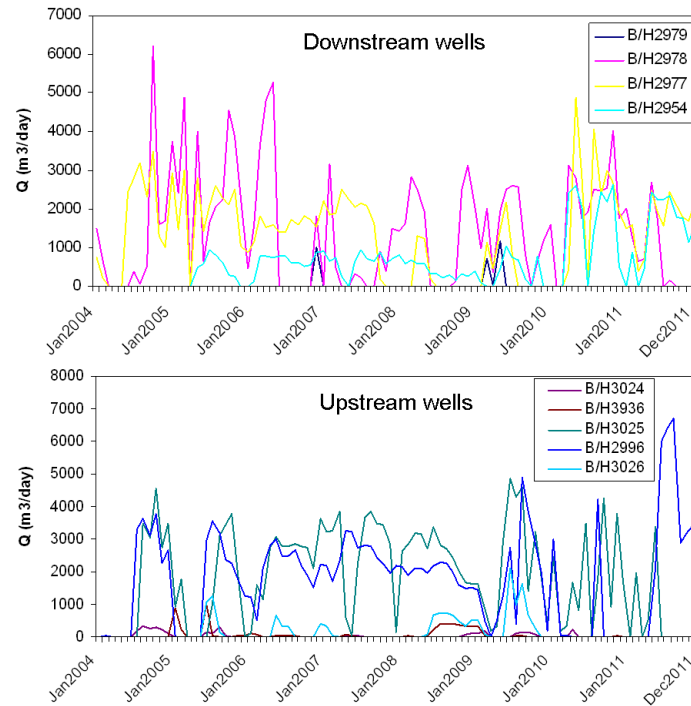


Figure 3. Extraction rates (m³/d) for the 9 pumping wells

2.2.3 Boundary conditions for mass

The subsurface transport of Phosphorous and Copper emanating from the recharge process was also studied using the FEFLOW model. To this end, the available measurements for the native groundwater (at well 4031) and the recharged effluent (at the five ponds) for the period of 25th January 2007 – 19th October 2011 were used as time dependent constant concentration boundary conditions (Figure 4). A high load of Phosphorus in the reclaimed wastewater was observed in May 2007 that was reduced over time. The native groundwater Phosphorus concentrations remain relatively low for the whole time period examined here. A high concentration of Copper was observed in 2007-2008 for both native groundwater and reclaimed wastewater that was reduced onwards and concentrations remained relatively low for the rest of the monitoring period.

The model was calibrated in order to match the observed Phosphorous and Copper concentrations at the upstream monitoring wells H3026 and H2996. These wells exhibit the most interesting results, since most of the wastewater (about 75%) is recharged in ponds 4 and 5.

The sorption isotherms used in the model for the Phosphorous and Copper were determined by sorption experiments and are described in the next section. The porosity was estimated at 0.3 and the longitudinal and transverse dispersivities were determined (after the process of calibration) at 20 m and 2m, respectively.

In order to account for the natural (background) groundwater concentrations of phosphorus and Copper, the native groundwater measurements that were taken at well 4031 were set as boundary conditions at various locations over the entire study area due to lack of additional measurements and assuming that the background concentrations do not vary significantly over the aquifer.

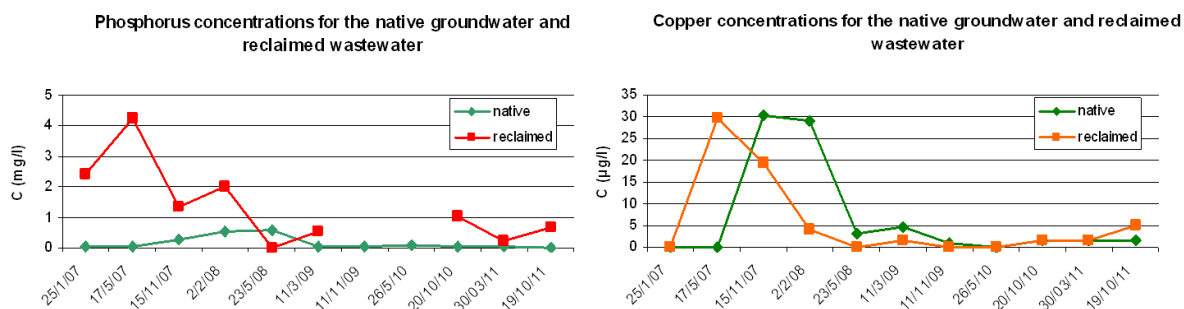


Figure 4. Measured phosphorus and Copper concentrations for the native groundwater and the recharged effluent for the period of 25th January 2007 – 19th October 2011

2.3 PHREEQC model setup

Inverse modelling was carried out using the geochemical modelling program ^{5, 6}, which simulates combinations of end-members proportions and amounts (i.e. meq) of minerals and gas mole transfers that are responsible for the differences in composition between water. An uncertainty limit is specified by the user for each component. Inverse modelling was performed for Ezousa Recharge project to understand the contribution of reclaimed water to groundwater and especially the vulnerability of groundwater during the transition from dry to wet condition. Possible end members (initial solutions) considered in the model were natural groundwater, groundwater from the well B/H3026 (downstream of the basin 5, Figure 1) and reclaimed wastewater. The evolution of groundwater in the wet period was studied using samples taken in 25/01/2007 and for the dry period in 20/10/2010.

2.4 Laboratory experiments

Groundwater and reclaimed water chemical analysis and its comparison to natural groundwater indicated strong phosphate sorption, nitrate and copper dilution. In order to understand the role of sediments processes in groundwater final synthesis after the mixing of natural groundwater to reclaimed wastewater specific leaching and sorption experiments of Ezousa sediment were carried out. Sediment samples were taken from different depths (0.3m, 1.0, m and 3.0 m) in the infiltration pond 5 (Fig.1) to identify the sediments capacity in pollutants attenuation. Natural sediment was sampled upstream to act as reference conditions. Sediment samples were analysed for basic metals (XRF), trace metals (ICP) and minerals (XRD) to identify the phases that contribute in precipitation and dissolution processes. A release kinetic experiment took place to assess the capacity of sediments to release phosphorous and heavy metals. A sorption batch experiment was also conducted to estimate sediments capacity to attenuate phosphate and copper.

Results - Discussion

2.5 Water quality

Table 1 shows the average heavy metal and nutrients content of reclaimed water of native groundwater and the groundwater. In general a weak dilution of nitrate in the coastal aquifer is observed since the infiltrated reclaimed water has an average nitrate concentration of 13.9 mg/L, the native groundwater 4.9 mg/L and the abstracted groundwater 12.1 mg/L. In contrast total Phosphorous is strongly absorbed and attenuated into the aquifer matrix since the content of the reclaimed water is 1.289 mg/L and the content of the abstracted groundwater is 0.102 mg/L. Concerning the heavy metals the groundwater shows higher values in comparison to reclaimed water but similar to that of native groundwater, that means that heavy metals origin is mostly geogenic than artificial.

Table 1. Average heavy metals content and nutrients in Ezousa recharge project for the period 11/2006 up to 10/2011 (n=14 samples)

	groundwater		native groundwater		reclaimed water	
Heavy metal Concentration in µg/L						
	average	standard deviation	average	standard deviation	average	standard deviation
Cadmium, Cd	0.1	0.2	0.1	0.2	0.1	0.1
Copper, Cu	12.8	20.2	6.3	11.1	7.1	10.3
Chromium, Cr	4.9	5.8	11.6	15.0	2.5	2.0
Zinc, Zn	61.0	36.3	103.1	102.5	27.8	20.5
Lead, Pb	6.6	7.4	7.1	8.1	4.8	5.7
Cobalt, Co	1.6	1.0	1.6	2.0	0.4	0.3
Vanadium, V	10.0	3.8	14.5	12.3	2.7	0.2
Arsenic, As	1.3	0.8	0.8	0.8	1.0	0.9
Nickel, Ni	8.4	3.4	13.7	14.8	7.8	8.0
Beryllium, Be	N.D.	-	0.1	0.1	0.03	0.0
Selenium, Se	0.8	1.3	2.3	2.3	4.6	2.3
Mercury, Hg	0.3	0.5	0.2	0.0	0.2	0.0
Boron, B	219.0	189.9	347.5	322.6	315.8	138.9
Manganese, Mn	111.2	75.8	85.9	90.5	58.7	28.4
Iron, Fe	526.5	652.3	1242.0	2412.0	176.3	130.5
Barium, Ba	31.2	17.4	48.6	26.7	18.3	14.7
Nutrients Concentration in mg/L						
NO3	12.1	2.9	4.9	5.6	13.9	7.6
TP	0.102	0.1	0.135	0.2	1.289	1.3

2.6 FEFLOW results

3.2.1 Flow results

The flow model calibration was based on the calibrated model by Milnes (2007)⁷. Slight modifications were made to obtain a better match between the results of the two models. In accordance to the previous model the percentage of treated wastewater recharged in each pond was 54.1% for the Pond 5 and 22.4 % for Pond 4. Pond 3 received almost no wastewater, while the two infiltration ponds in the downstream aquifer portion were artificially recharged with 8.5% (Pond 1) and 14.5% (Pond 2), respectively.

The calibration results for the steady state are shown in Figure 4. The results for the transient period are similar (not shown here for brevity) with the difference that on the downstream area a more prominent drawdown occurs. This is due to the increased extraction rates that, combined with the construction of the Kannaviou dam, have resulted in lowering the hydraulic head levels of the Ezousa aquifer, to a certain degree; a fact that could induce the saltwater intrusion phenomenon in the costal area. In general, it is observed that, artificial recharge can compensate the lack of upstream recharge only to a point. Depending on the distribution of artificial recharge in space and time a water deficit is possible.

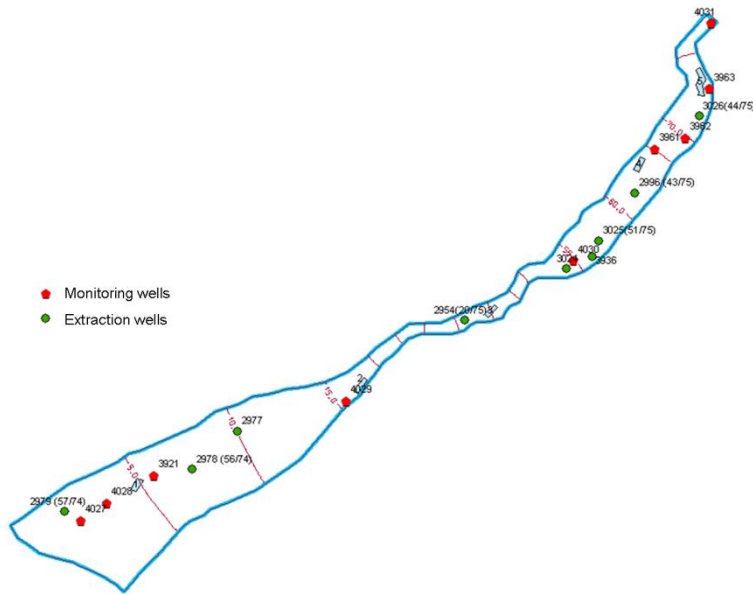


Figure 4. Steady state model calibration results (December 2003)

3.2.2 Mass transport results

The model results for the Phosphorous and Copper transport in the subsurface are presented in Figure 4. In general, the results for are in relatively good agreement with the observed values for both wells. For phosphorus, the simulation did not capture the behavior of phosphorus concentration for the period 23/5/08-26/5/10 very well. This is due to the fact that there was no available data for the reclaimed wastewater for that period and in addition the concentrations of the native groundwater were very low in contrast to the observed values at the wells. An unusually high Copper concentration is observed in 2007 that is attributed to high concentrations in both the native and reclaimed water and is captured relatively well by the model. The results for Copper also show a deviation from the measured values for the same time period 23/5/08-26/5/10. This could be attributed to the fact that during this period Copper concentrations for both the reclaimed wastewater and native groundwater become very low fast but it takes more time for the model to reduce these concentrations. Note that for the last observations the measured values are not 5 but $<5 \mu\text{g/l}$, thus the modeled values could be much closer to the real value than shown in the graph.

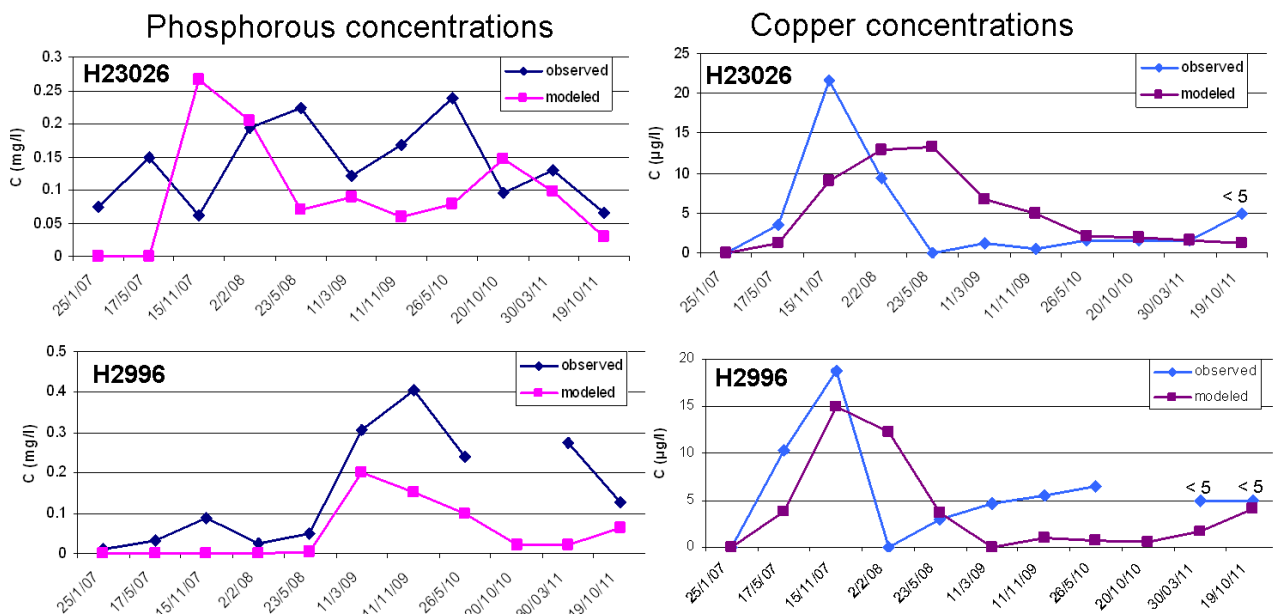


Figure 4. Mass transport results for Phosphorous and Copper

2.7 PHREEQC results

The mineralogical analysis of Ezousa project sediments indicated slightly differences of the sediments concerning minerals and heavy metal content in the various depths. The main minerals are of siliceous and carbonaceous synthesis (Calcite (30-44%), Quartz (19-36%), Montmorillonite (0-3%), Labradorite (22-26%) and Clinocllore (8-9%). XRF analysis indicated that sediments contain high silica minerals SiO₂ (51%), calcite (27.5%), ferrous oxide (5.5%) and Mg (3.6%) and Na (2.7%) oxides.

The synthesis of the water used for the study of dry and wet period is shown in Table 2. PHREEQC modelling indicated that aquifer in the upper part of Ezousas project is recharged during the wet period by groundwater (97-100%) and reclaimed water (0-3%). In the dry period the groundwater synthesis is attributed mainly to groundwater (mainly reservoir losses) (95%) and reclaimed water (5%). In Table 2 is presented the mole transfer between the mineral phases and the water. The highest mole transfers occur for hematite and goethite phase.

Table 2. Mole exchange between the water and the minerals in Ezousa aquifer

	Ca	Mg	Na	C(4)	Cl	S(6)	N(5)	pH	P
	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l		mg/l
wet period - 25/01/2007									
natural gw recharge	242	59	106	336	133	594	2.03	7.7	0.023
water reclaimed	198	49	126	342	151	459	2	7.4	0.074
wastewater	64	30	190	320	204	171	1.58	7.9	2.4
Phase	Ca-Montmori-llon	Quartz	Calcite	Goethite	Gibbsite	Gypsum	Hematite	Halite	
mole transfer	-2.87E-03	1.05E-02	6.76E-04	1.15E+01	6.68E-03	-5.76E-04	-5.75E+00	8.93E-04	
dry period - 20/10/2010									
natural gw recharge	241	59.2	105	347.7	116	661	0.7	7.2	0.024
water reclaimed	215	49.7	94.5	347.7	121	576	1.8	7	0.096
wastewater	63.5	31.6	188	375.2	185	156	2	7.4	1.007
Phase	Ca-Montmori-llon	Quartz	Calcite	Goethite	Gibbsite	Gypsum	Hematite	Halite	
mole transfer	-	-	0.000203	-68.83	-	-0.00827	28.33	-0.00388	

Sorption isotherm was conducted in 20°C for Ezousa sediments (3m depth) to evaluate the partitioning coefficient for Phosphorous. At low P concentration (4-5 mg/L P-PO₄) the isotherm is linear and the K_d estimated 2.8168 ml/gr at pH 7.8. In lower than 4 mg/L concentration of P the adverse effect of P leaching added high uncertainty in the sorption experiment results.

3. CONCLUSIONS

Weak leaching capacity of heavy metals and nutrients was observed during the leaching experiments but strong phosphate and Copper sorption. The linear partitioning coefficient for Phosphorus was estimated at 2.8168 ml/gr and for Copper at 16.9-18.8 ml/gr at pH 7.8.

From the flow model it is suggested that due to the increased extraction rates that, combined with the construction of the Kannaviou dam, a lowering of the water table levels of the Ezousa aquifer has occurred, to a certain degree. This

suggests that artificial recharge can compensate the lack of upstream recharge only to a point. Thus, a water deficit could potentially be created that will induce the saltwater intrusion phenomenon in the costal area.

Regarding the contaminant transport model, the simulation captured the general behavior of phosphorus and Copper relatively well with some exceptions. Regarding the contaminant transport model, the simulation captured the general behavior of Phosphorus and Copper relatively well with some exceptions. An unusually high Copper concentration is observed in 2007 that is attributed to high concentrations in both the native and reclaimed water and is captured well by the model. The results for both Phosphorus and Copper show a deviation from the measured values for the same time period 23/5/08-26/5/10. For phosphorus, this could be attributed to the fact that there are no available data for the reclaimed wastewater for that period and in addition the concentrations of the native groundwater were very low in contrast to the observed values at the wells. For Copper, the concentrations for both the reclaimed wastewater and native groundwater become very low fast but it takes more time for the model to reduce these concentrations.

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