Seawater Intrusion and Nitrate Pollution in Coastal Aquifer of Almyros – Nea Anchialos Basin, Central Greece

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Abstract: -The exploitation of groundwater resources is of high importance and has become very crucial the last decades especially in coastal areas of Central Greece, due to urbanization, tourist development and intensive agriculture. The phenomenon of seawater intrusion in coastal aquifers is a significant environmental issue in Greece that nowadays is considered as pollution of groundwater, which could pose a threat to human's health through direct consumption or through irrigation processes. The objectives of this study are to identify and investigate the extent of seawater intrusion in the Nea Anchialos basin in Central Greece. For this purpose, 45 groundwater samples were collected from the study area and analyzed for certain parameters that indicated salinisation of the aquifers. Specifically, water chemical analysis were conducted and statistical analysis regarding spatial distribution were performed. The results demonstrated increased values of the parameters which can be attributed to seawater intrusion. Laboratory analyses indicated that Electrical Conductivity, concentration for the basic cations (Ca^{+2}, Mg^{+2}, Na^+ and K^+), HCO_3^-, Cl^-, and SO_4^{-2} varied greatly. Furthermore, maximum NO_3^- concentration was 138 mg/L and this can be attributed to overexploitation of coastal aquifer for agricultural activities. The results indicated seawater intrusion in the north-eastern part of the study area. Hydrochemical maps were compiled using SURFER version 7 Software, based on models Shepard and Inverse Distance to a Power (IDP), in order to present the results in an integrated way and demonstrate the "hotspots" of pollution.

Key-Words: - Seawater Intrusion, Groundwater, Salinisation, Irrigation, Coastal Aquifer.

1. Introduction

Groundwater has historically supplied domestic water requirements in urban and rural settlements around the world. It is estimated that more than half the world's population relies on groundwater for drinking purposes [6]. However, irrigated agriculture remains the major user of groundwater resources. Due to the fact that water crisis is forecasted in the near future [11], the welfare of the world's population is closely linked to water resources. Sustainable aquifer management has significant importance and it's highly depended on rational management, as well as, preservation of groundwater stock.

Groundwater salinisation derives through a number of mechanisms, mostly is caused by anthropogenic activities. Leaching of saline water in soils towards fresher groundwater occurs due to increased recharge through various processes, such as percolation, infiltration, water table fluctuation. Surface water rich in salt can recharge the aquifer through irrigation. Groundwater can become gradually more saline through plant water use especially in the shallow aquifers, by the recycling of irrigation water which then drains back to aquifer [17]. Additionally, groundwater deterioration could be caused by seawater intrusion, agrochemical inputs for agricultural purposes and geogenetic contamination [21]. Although groundwater salinisation and salt accumulation are often natural processes, they can cause serious environmental problems that threaten human's health and affect biodiversity and agricultural productivity [10].

Groundwater salinisation is considered to cause predominantly from seawater intrusion (ασαφές). This phenomenon is characterized by the movement of seawater into the land which is mixed with the

fresh water of the aquifers due to natural processes or human activities [9]. Seawater intrusion is caused mainly by the decrease of groundwater table or by increasing the seawater levels. When groundwater is pumped from a coastal aquifer, the fresh-water level is lowered and the sea intrudes into the aquifer. Therefore, the amount of freshwater stored in the aquifer is decreased [4].This phenomenon is observed in the majority of coastal aquifers in Centrak Greece and the main cause is the continuous over pumping of groundwater for irrigation or human consumption in the touristic areas during the summer period Many previous case studies indicated that the problem of seawater intrusion is very important worldwide [3, 7, 16, 25, 22, 26].

Deterioration of groundwater quality appears in the semi-arid areas of the Mediterranean countries indicating that this problem is not only a major environmental problem, but also can be characterized as a socioeconomic one [13] which may have a negative impact on the living standards. The use of hydrochemical maps is essential in order to demonstrate in an integrated way the extent of seawater intrusion by modeling spatial variability of environmental variables. These maps constitute a very useful tool for local authorities in order to identify and monitor the existing situation that could degrade fresh water and therefore pose a threat to human health. For this aim, a wise management of water resources could be planned and applied.

Groundwater samples from 45 sites were collected from Almyros and New Anchialos basin in order to describe and assess the groundwater quality in relation with the impact of natural and anthropogenic activities. Ten soil samples were also collected in order to depict the relationship between the quality of soils and both the existing geological and anthropogenic conditions.

2. Study Area
The study area is situated in Almyros - Nea Anchialos basin (Fig. 1), and belongs to the Prefecture of Magnesia, south-east Thessaly, Central Greece.
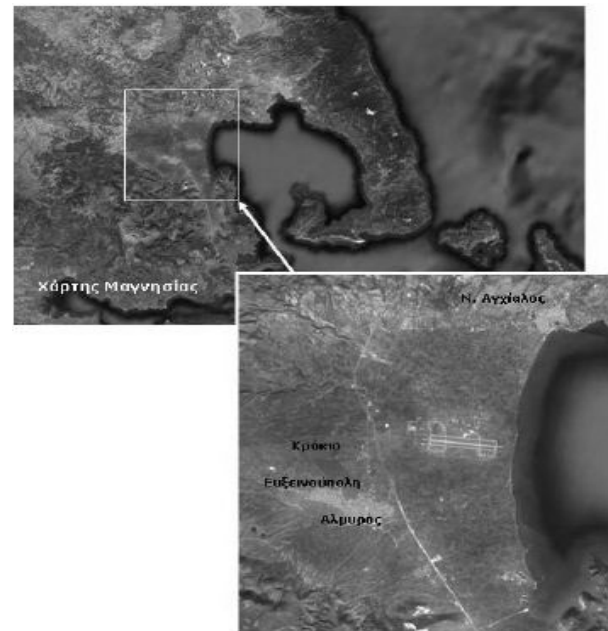

Figure 1: Map of the study area.

The Almyros basin is quiet flat and its extend has a total area of 865 km^2 while the altitude ranges from zero up to 93 m (Fig. 2). The main part of the area is used for agricultural activities and grazing.
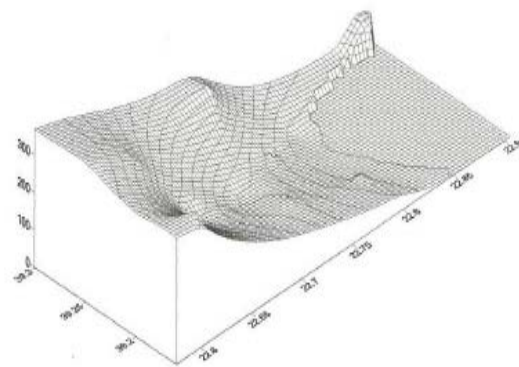

Figure 2: Study area elevation.

According to the received data from the Hellenic National Meteorological Service, the climate of the study area is typical of the Mediterranean climate, with a mean annual rainfall which ranges from 400 to 600 mm per year (Fig. 3).
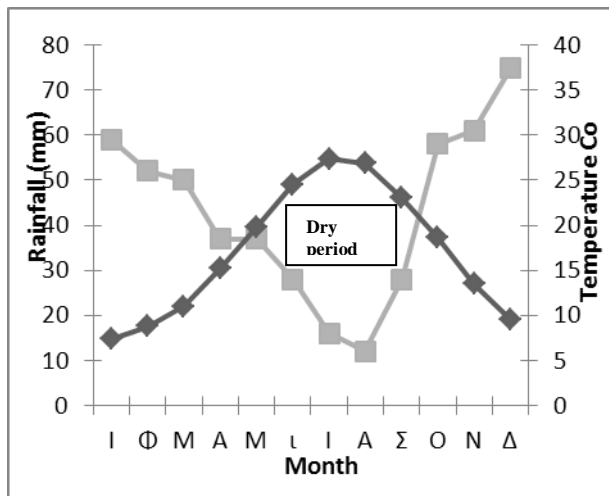
Figure 3: Ombrothermic diagram of the study area (source: meteorological station of Nea Anchialos).

The geology of the study area is characterized by the presence of limestones (Triassic and Cretaceous), dolomites, schists, shales, sandstones and sedimentary formations. These rocks are covered by Neogene and Quaternary sediments. The Neogene sediments are composed of marly limestones, conglomerates and marls. The Quaternary sediments are consisted by brown coloured clayey-sandy materials with dispersed cobblesrubbles. The geological formations of the study area that cover the highland in the border area are: limestones (Jurassic or upper Cretaceous age), flysch, volcanic rocks and ophiolitic rocks. These rocks have primary or secondary porosity, due to the intense tectonic activity. Limestones occur in the northern (Nea Anchialos) and the southern part (Sourpi) of the plain dominates by karstified formations [24].

According to Mountrakis (1985) [23], the study area is part of the Pelagonian zone, as far as the geological framework is concerned. The geological setting is mainly consisted of Mesozoic limestones and dolomites schists and gneisses, which pass upward to sedimentary formations (sandstone, shale conglomerate ect.). Those are covered by Neogene and Quartenary sediments. The Neogene sediments are composed mainly of marl and coglomerates. The Quaternary sediments consist of clayey-sandy materials with dispersed cobbles- rubbles [5].

As far as the hydrogeological framework is concerned, the study area is characterized by the presence of a rich surface hydrographic net, mainly due to impermeable bedrock that impedes the percolation of water to groundwater aquifers [5, 12].

3. Materials and Methods

A bailer sampler was used in order to collect 45 groundwater samples from different wells of the study area (Fig. 4).
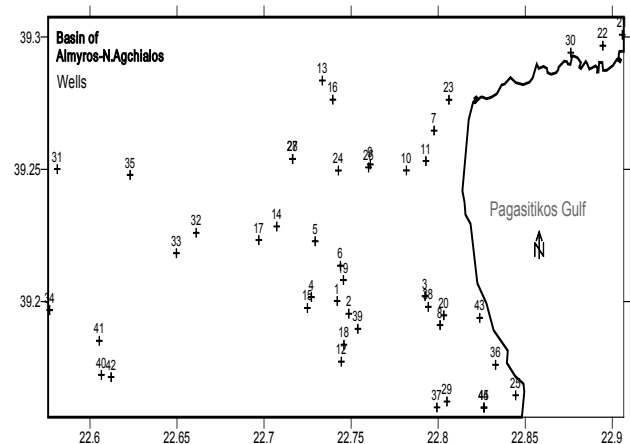

Figure 4: Groundwater wells network used for sampling.

Physico chemical parameters, such as pH, electrical conductivity (EC), temperature, and total dissolved solids (TDS) were measured immediately in the field, using a pH-meter, conductometer and thermometer and TDS meter, respectively. Afterwards the samples were transferred to the University of Thessaly, School of Agricultural Sciences, Department of Ichthyology & Aquatic Environment, Geochemistry lab for storage and analysis. The collection, transfer and storage of the samples were performed according to the protocol ISO 5667-3. The samples were filtered in order the suspended particulates ($>0.45\mu$m) to be removed and prevent the dissolved elements absorbed into particles. The samples were then analyzed by Ion Chromatography (IC). Anions were determined by the use of Ion Chromatography with Conductivity Detector (IC-CD) according to the protocol ASTM D4327, while cations were determined by using Metrhom's Ion Chromatography with autosampler 728, Micrometrics according to the protocol ISO 14911:1998. Nitrate values that were found below detection limits were estimated by the use of UV/Vis spectrophotometer, Analytic Jena. Hydrochemical maps were compiled using SURFER 7 software based on Shepard and Inverse Distance to a Power (IDP) models. These maps were based on the calculated values of Revelle Coefficient, SAR (Sodium Adsorption Ratio), chloride anion concentrations, SEC (Specific Electrical Conductivity) and nitrate concentrations, parameters that indicate seawater intrusion in freshwater aquifers. The curves represent areas with

similar concentration of each variable. The coordinates of the sampling wells were recorded by using Garmin Global Positioning System (GPS) and were also used for the preparation of the hydrochemical maps.

4. Results and Discussion

The monitoring network of the study area contains 45 groundwater wells, used mostly for agricultural purposes in order to cover the irrigation requirements of crops. The pH values varied from 7.13 to 8.12, while Total Dissolved Solids (TDS) were estimated between 267 and 4736 ppm. The results of the chemical analyses concerning the main and major ions of the investigated aquifer system are demonstrated in Table 1.

In Table 2 we can see the minimum, maximum and average values of Standard Deviation (SD) and Coefficient of Variance (%) (CV), of the main ions.

The piper diagram (Fig. 5) shows the general chemical composition of the groundwater samples of the study area. Two areas (I, II) are highlighted in this figure that is in the salination zone with one sample characterized of intense values of total hardness.
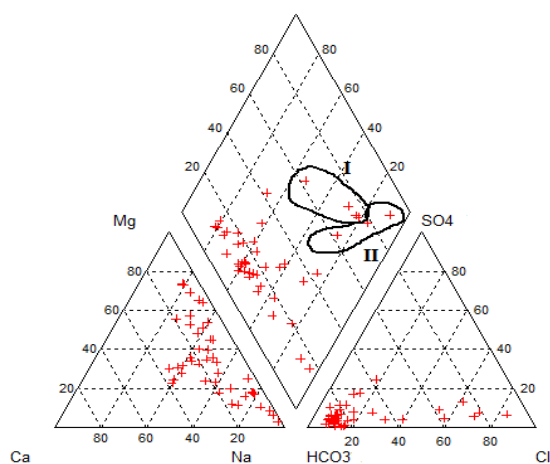

Figure 5: Piper diagram.

The minimum, maximum and average values of Standard Deviation (SD) and Coefficient of Variance (%) (CoF) are shown in Table 3. Specifically, Sodium Adsorption Rate (SAR), Sodium content (as a percentage), Specific Electrical Conductivity (SEC), Na^+/Cl^- ratio and Revelle coefficient were estimated (calculated?). These parameters are valuable indicators for the assessment and classification of fresh water salinisation.

4.1 Chloride Ions (Cl^-)

An increased Cl^- ion concentration gives a good indication of seawater encroachment [28]. Although chloride is essential to plants in low quantities, it can cause toxicity to sensitive crops at high concentrations [19]. The classification of water's quality according to Soulios (2004) [27] is demonstrated in Table 4.

The chemical analysis of water samples denotes that 56% of the samples contain low amounts of chlorides, with a minimum value of 19 mg/l. However, 18% of the samples exceed the value of 100 mg/l, with a maximum value of 2185 mg/l, while the trigger value for drinking water according to national regulatory law (Joint Ministerial Decision, Ministry of Development/2600/2001) is 250 mg/l. Fass et al., (2007) [8] suggested that elevated concentrations of chloride ions can be attributed to seawater intrusion due to frequent over pumping. High values of chloride ions are shown in the northeast part of the study area and specifically in close proximity to the sea (Fig. 6). The distribution of chlorides shows a general increase of the concentration while the hydraulic gradient decreases towards the coastal line of the study area.
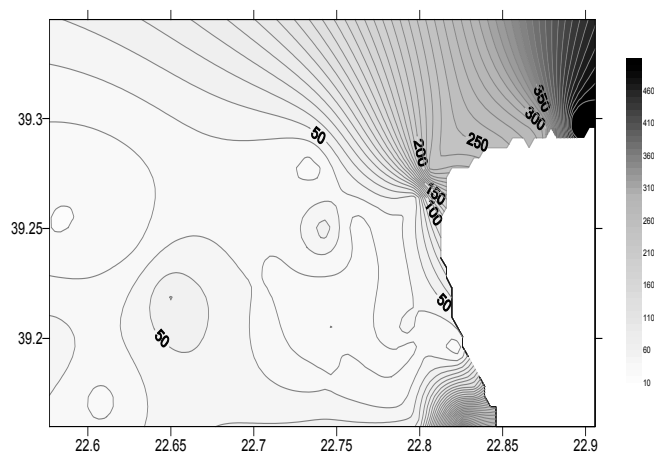
Figure 6: Hydrochemical map based on the distribution of chloride anions (Cl^-) concentration in mg/l.

4.2 Specific Electrical Conductivity (SEC)

The value of Specific Electrical Conductivity is a qualitative parameter that indicates increased levels of salt content both in water and soil. Elevated value of Electrical Conductivity is also an indication for possible lack of rainfall or limited available water volume due to over pumping of groundwater resources [2], and suggest high salt content [18]. Chemical analysis indicated that high values of EC (>900 μS/cm) appeared in 22% of the studied

samples and specifically in those that stem, from the wells located near the sea. The obtained results were used to compile the hydro chemical map (Fig. 7) which explains the problem in the northeast part of the study area.

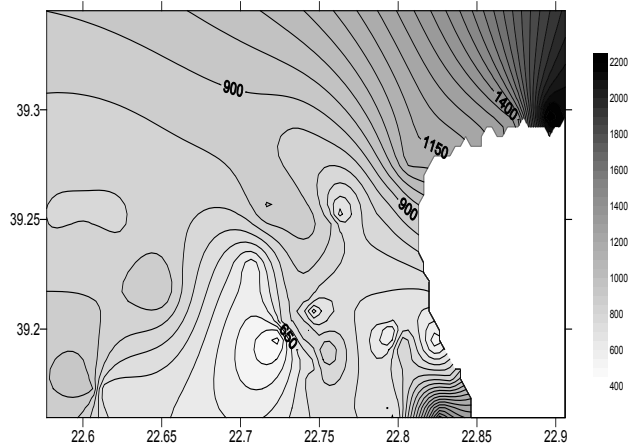**Figure 7:** Hydro chemical map based on the distribution of Specific Electrical Conductivity (SEC), in μS/cm.

4.3 Sodium Percentage Content

Sodium ions can reduce the aeration and the permeability of soils by means of the process of exchange with calcium ions, causing soil deflocculation (Kallioras, 2006). Sodium content is usually expressed as a percentage and can be determined by the following equation: Na (%) = (Na + K) * 100 / (Ca + Mg + Na + K), concentrations expressed in meq/l.

According to Kaur & Singh (2011) [15], water can be classified to hazard classes depending on the sodium content (Tab. 5). The calculation of sodium content in the water samples showed that 11% of samples are classified as "permissible", while 7% are considered "doubtful", with values over 60 Na%.

4.4 Sodium Adsorption Rate (SAR)

The most common measure to assess sodicity in water is the Sodium Adsorption Rate (SAR). The SAR assesses the potential for infiltration problems due to a sodium imbalance in irrigation water. The sodium content is usually expressed as a percentage of exchangeable sodium and is calculated according to the following equation in meq/l.

$$S.A.R. = rNa / \sqrt{\frac{r(Ca^{+2} + Mg^{+2})}{2}}$$

Calculation of SAR demonstrated that 98% of the samples revealed values less than 10. These values combined with a mean value for electrical conductivity of 750 μS/cm indicate small to medium sodium hazard, while the 2% of the samples with elevated values (15.65 μS/cm) indicate high sodium hazard [2]. Figure 8 demonstrates the distribution of SAR values in the study area.
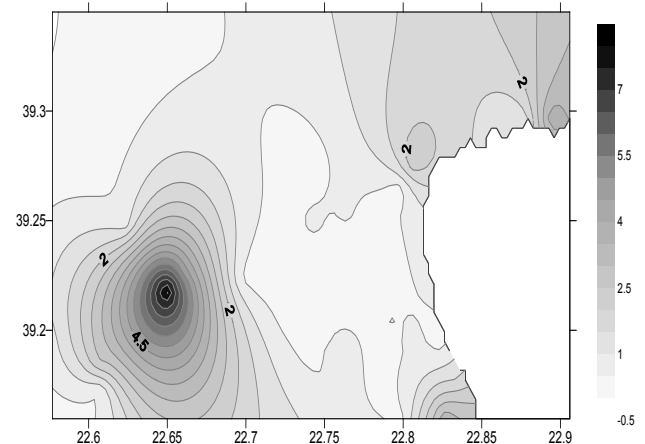
Figure 8: Hydro chemical map based on the distribution of Sodium Absorption Rate (SAR).

4.5 Nitrate Concentration

The nitrates concentration which is originated from the application of nitrogenous fertilizers to crops is a serious problem. It was recorded that 25-30% of the samples were polluted by nitrates and the content was higher than 45 mg/l. Nitrate in elevated concentrations can cause adverse effects on human health and livestock, as well. Nitrate concentrations are affected seriously by the amount of applied nitrogen by means of fertilization, soil drainage, run off in slopes (Fig. 9). Specifically, high levels of nitrates are associated with intensively cultivated areas in the plains, while decreased values appear in mountainous areas. Mihaiescu et al. (2008) [20] came to the same conclusion, when they observed an increase of nitrate concentrations from highlands towards to lowlands.
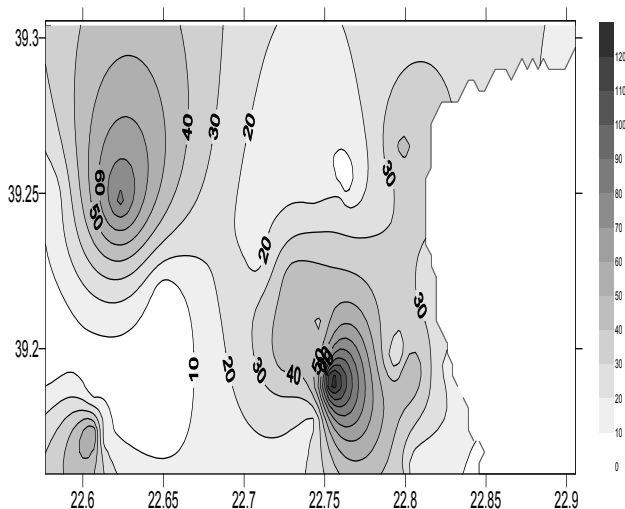
Figure 9: Hydro chemical map based on the distribution of nitrate concentration (mg/l).

4.6 Revelle Coefficient

Revelle coefficient is considered an important criterion to determine phenomena's of seawater intrusion. It is expressed as the ratio of $R = Cl/(CO_3 + HCO_3)$ in meq/l. Generally, values of $R \geq 1$ are thought to be a valid indication of seawater intrusion (Kallergis, 2000). Table 6 illustrates two different classifications of water's quality, using the Revelle coefficient.

Results indicated that 8.8% of the samples presented low level of contamination, while 2.2% of them indicated medium level of pollution. Spatial distribution (Fig. 10) showed elevated concentrations in the south-east part of the study area, in the wells located near the sea and this may be attributed to water over exploitation for irrigation.

Variation of Revelle index can be attributed mainly to seawater intrusion, while the respective variation of SAR is strongly influenced by the concentration of Na^+
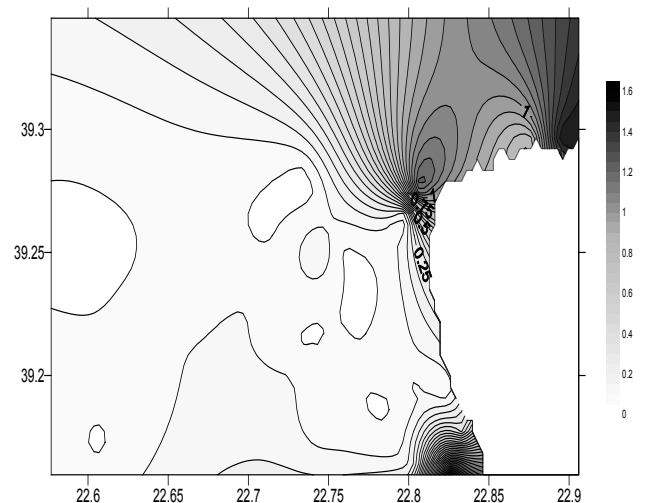
Figure 10: Hydrochemical map based on the distribution of Revelle coefficient.

4.7 Comparison of results with previous studies conducted in the Almyros Basin

In this chapter discussion is focused on comparison of average values of the current works with previous works carried out (Tab. 7) in the same area and attention is paid in international criteria which have been set up by internationa regulations (Tab. 8).

According to Table 7 it is obvious that during the last 9 years (2005-2013) the ground waters in the Almyros basin are in a better state in terms of quality due to the decreased values in the studied hydro chemical parametrs. This decline may be attributed to the decreased ground water exploitation, while strict rules and requirements are in force, in order the competent authorities to provide license for the construction of new boreholes. In the following Table 8 we can see the comparison of the current work with international criteria for the water quality or so.

Table 8 also shows the relationship between our study area and the regulations 98/83/EEC, 80/778/EEC. The analyzed ground water samples of Almyros basin for the physicochemical parameters Cl, SO_4, Na, K, Mg, Ca and pH doesn't exceed the admissible limits of the regulations 98/83/EEC, 80/778/EEC. The only exception concerns the mean value of electrical conductivity.

5. Conclusions

Chemical analyses indicated a great variation for most of parameters for the studied water samples. results. Coefficient of variance was higher for Revelle index following by SAR.

Piper diagram classifies the surface water chemical data set into two types: First type: where sovereign ions are HCO_3^-, Mg^+, Ca^+ and second type where sovereign ions are Na^+ and Cl^-.

Regarding to suitability of the surface water for usage in the hinterland of the study area, it can be argued that quality can be characterized as suitable (C2 - S1 field in Richards's diagram) with some restrictions in certain locations. Water can be used in soils with good permeability and in soils with high degree of degradation. In the antipode the samples near coastal zone in the estuaries, are not suitable for usage due to salinisation, as shown in Richards and Wilcox diagrams.

The most serious problem in the study area is the increased level of nitrate concentrations. It has been recorded thatin 25 - 30% of samples nitrates content ranged between 45 and 50 mg/l. The increased content originatesfrom leaching of nitrates which the applied nitrogenous fertilizers contain.

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TABLES

Table 1: Results of the hydrochemical analyses of the water samples.

	Ca^{+2}	Mg^+	Na^+	K^+	Cl^-	NO_3^-	NO_2^-	SO_4^{2-}	PO_4^{3-}	TDS	TH $CaCO_3$ (ppm)	HCO_3^- (ppm)	pH	EC ($\mu S/cm$)
1	133.2	15.1	13.6	1.0	26	48	<0.02	12	<0.1	529	396	390	7.13	826
2	132	12.8	10.9	1.0	28	49	<0.02	34	<0.1	482	383	350	7.20	753
3	110.3	13.7	21.8	1.2	46	29	<0.02	8	<0.1	428	333	340	7.41	669
4	128.0	13.2	14.4	1.2	27	48	<0.02	23	<0.1	470	375	360	7.41	734
5	126.6	16.1	14.9	1.3	27	46	<0.02	21	<0.1	492	384	370	7.42	769
6	134	10.5	12.3	1.1	21	49	<0.02	17	<0.1	493	379	380	7.40	771
7	70.5	76.6	14.6	1.2	43	43	<0.02	17	<0.1	592	495	480	7.68	925

8	118.5	31.7	24.8	1.9	39	49	<0.02	48	<0.1	506	428		390	7.54	790
9	47.9	47.1	25.5	1.7	25	2	<0.02	6	<0.1	389	316		390	7.80	608
10	58.3	68.5	13.5	1.1	39	26	<0.02	3	<0.1	500	431		440	7.79	782
11	35.7	56.1	19.0	1.0	43	30	<0.02	13	<0.1	525	323		325	7.71	820
12	77.7	40.0	29.3	2.2	41	19	<0.02	7	<0.1	470	361		410	7.73	735
13	57.7	85.0	17.4	1.3	30	13	<0.02	20	<0.1	526	498		540	7.90	822
14	86.7	28.5	9.4	1.1	27	15	<0.02	18	<0.1	351	335		350	7.63	548
15	51.5	15.6	16.2	2.0	28	46	<0.02	8	<0.1	267	194		175	7.88	417
16	64.9	75.3	14.0	0.9	28	16	<0.02	20	<0.1	525	476		510	7.90	820
17	64.2	37.2	20.9	1.2	43	26	<0.02	27	<0.1	402	316		300	7.80	628
18	104.3	18.6	15.7	1.2	28	46	<0.02	14	<0.1	428	338		320	7.67	668
19	74.0	18.0	18.4	1.4	32	35	<0.02	3	<0.1	356	260		250	7.74	556
20	106.2	15.2	28.0	0.7	39	50	<0.02	60	<0.1	499	329		275	7.64	780
21	145.0	60.0	192	5.3	503	18	<0.02	50	<0.1	357	613		260	7.46	2120
22	139.0	67.0	223	5.5	549	19	<0.02	55	<0.1	1453	627		280	7.77	2270
23	54.0	48.0	101	4.2	237	34	<0.02	63	<0.1	717	335		145	7.56	1120
24	34.0	70.0	26.0	1.2	57	17	<0.02	96	<0.1	563	377		275	7.40	880
25	234.0	115	1174	38.2	2185	7	<0.02	214	<0.1	4736	1064		370	7.62	7400
26	94.4	50.2	19.6	1.9	27	20	<0.02	8	<0.1	506	445		490	7.35	790
27	63.0	78.0	20.2	1.1	34	15	<0.02	18	<0.1	581	483		495	7.73	908
28	108.0	7.5	9.5	0.9	23	40	<0.02	9	<0.1	390	301		290	7.34	610
29	110.0	9.0	39.0	1.0	92	12	<0.02	14	<0.1	515	313		275	7.40	805
30	122.0	70.0	53.0	1.5	287	21	<0.02	29	<0.1	861	597		280	8.12	1345
31	109.0	37.0	32.0	2.2	19	12	<0.02	51	<0.1	508	427		490	7.94	794
32	37.0	19.1	146	1.3	51	16	<0.02	2	<0.1	518	172		450	7.49	810
33	27.0	8.0	190	0.7	61	3	<0.02	3	<0.1	582	101		490	7.83	910
34	130.0	28.1	42.0	1.8	48	0	<0.02	85	<0.1	563	442		425	7.16	880
35	98.0	29.0	34.0	0.4	22	84	<0.02	20	<0.1	506	366		375	7.47	790
36	94.0	19.0	64.8	2.4	128	18	<0.02	16	<0.1	538	314		270	7.38	840
37	74.0	29.5	17.0	0.9	61	30	<0.02	24	<0.1	410	308		250	7.42	640
38	64.0	26.2	12.0	0.7	22	17	<0.02	18	<0.1	307	269		275	7.78	480

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39	135.0	14.1	35.9	1.0	31	138	<0.02	24	<0.1	557	396		330	7.71	870
40	153.0	27.0	28.0	1.4	26	67	<0.02	31	<0.1	585	495		485	7.18	914
41	73.0	31.1	97.0	`1.4	34	7	<0.02	56	<0.1	564	312		470	7.34	882
42	100.0	17.0	41.0	0.6	28	15	<0.03	16	<0.2	416	321		390	7.53	650
43	85.0	14.0	12.8	2.8	22	18	<0.04	6	<0.3	320	271		280	7.55	500
44	106.0	25.7	149	5.2	352	13	<0.05	48	<0.4	947	372		165	7.24	1480
45	99.0	9.2	43.0	1.9	133	32	<0.06	28	<0.5	499	286		140	7.64	780

Table 2: Minimum, maximum, average and standard deviation values of other physicochemical parameters of the study area.

	Cl^-	Ca^{+2}	Na^+	Mg^{+2}	K^+	NO_3^-	SO_4^{-3}	NO^{-2} (mg/l)	PO_4^{-3} (mg/l)	pH	EC	TDS	TH	HCO_3^- (mg/l)
Min	19	27	9.4	7.5	0.4	0	2	0	0	7.13	417	267	194	140
Max	2185	153	1174	115	38.2	138	214	0.05	0	8.12	7400	4736	1064	540
Mean	126	95	70	36	2.5	30.17	30.28	0.02	0	7.57	997.5	638.4	385.7	350.8
SD	336	40	176	26	5.6	24.37	35.47	0.008	0	0.23	1036.9	663.6	147.6	102.02
CoF (%)	266.7	42.1	251.4	72.2	224	24.1	35.07	-	-	0.05	1025.3	656.2	145.9	100.88

Table 3: Statistics of hydrochemical parameters of the groundwater of the study area regarding their influence on agricultural yield.

VALUES	SAR	%Na	SEC	Na^+/Cl^-	Revelle
Minimum	0.224	6	417	0.28	0.03
Maximum	15,703	80	7,400	4.8	4.5
Average	1,433	20.0	998	1.19	0.34
Std Deviation	2,652	17.9	1,037	1.03	0.76
Coefficient of Variance (%)	185.1	89.5	103.9	86.6	223.5

Table 4: Classification of groundwater's quality based on the chloride (Cl^-) concentration.

Cl⁻ (mg/l)	Waterquality
<300	Groundwater of good quality
300-5000	Moderate salinity
>5000	seawater

Table 5. Sodium hazard classes.

Range of sodicity	Samples	Waterclass
<20%	73%	excellent
20 - 40%	9%	good
40 - 60%	11%	permissible
>60%	7%	doubtful

Table 6: Values of Revelle coefficient that indicate deterioration due to seawater intrusion according to Simpson 1946.

Suggested limits (Simpson)	Suggested limits Kallergis (2000)	Water quality
0.5	<1	Groundwater ofgood quality
1.3	1-2	Low level of contamination
2.8	2-6	Medium level of contamination
6.6	6-10	Serious level of contamination
15.5	10-150	Dangerouslevelofcontamination
200	>150	Seawater

Table 7. Correlation with other similar works.

Parameters / Works	Ca^{+2} (mg/l)	Mg^{+2} (mg/l)	Na^+ (mg/l)	K^+ (mg/l)	Cl^- (mg/l)	NO_3^- (mg/l)	SO_4^{-2} (mg/l)	HCO_3^- (mg/l)	pH	EC (μs/cm)
Current work	95	36	70	2.5	126	30.17	30.28	350.8	7.57	997.5
Myriounis et al., 2006, (May	118.6	44.8	45.3	2.0	174.1	49.0	30.6	475.0	7.4	819.0

2005)										
Myriounis et al., 2006, (Sept. 2005)	104.7	40.0	74.5	1.4	149.9	48.9	34.8	318.0	7.3	846.8

Table 8. Comparison of the current work with international criteria (98/83/EEC, 80/778/EEC).

Parameter	Parametric Value	Current Work
pH	6.5 – 9.5	7.57
E.C. (μS/cm)	400	997.5
Ca^{+2} (mg/l)	100	95
Mg^{+2} (mg/l)	50	36
K^+ (mg/l)	12	2.5
Na^+ (mg/l)	200	70
SO_4^- (mg/l)	250	30.28
Cl^- (mg/l)	250	126