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4 **Identifying the effects of human pressure on groundwater quality to support water**
5 **management strategies in coastal regions: a multi-tracer and statistical approach (Bou-Areg**
6 **region, Morocco)**
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27 **Abstract**

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29 Groundwater pollution from anthropogenic sources is a serious concern affecting several coastal
30 aquifers worldwide. Increasing groundwater exploitation, coupled with point and non-point pollution
31 sources, are the main anthropogenic impacts on coastal environments and are responsible for
32 severe health and food security issues. Adequate management strategies to protect groundwater
33 from contamination and overexploitation are of paramount importance, especially in arid prone
34 regions, where coastal aquifers often represent the main freshwater resource to sustain human
35 needs.
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38 The Bou-Areg Aquifer (Morocco) is a perfect example of a coastal aquifer constantly exposed to all
39 the negative externalities associated with groundwater use for agricultural purposes, which lead to a
40 general increase in aquifer salinization. In this study data on 61 water samples, collected in June
41 and November 2010, were used to: (i) track groundwater composition changes related to the use of
42 irrigation water from different sources, (ii) highlight seasonal variations to assess aquifer
43 vulnerability, and (iii) present a reproducible example of multi-tracer approach for groundwater
44 management in rural coastal areas.
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47 Hydrogeochemical results show that Bou-Areg groundwater is characterized by - high salinity,
48 associated with a remarkable increase in bicarbonate content in the crop growing season, due to
49 more intense biological activity in irrigated soils. The coupled multi-tracer and statistical analysis
50 confirms the strong dependency on irrigation activities as well as a clear identification of the
51 processes governing the aquifer's hydrochemistry in the different seasons. Water Rock Interaction
52 (WRI) dominates the composition of most of groundwater samples in the Low Irrigation season (L-
53 IR) and Agricultural Return Flow (ARF) mainly affects groundwater salinization in the High Irrigation
54 season (H-IR) in the same areas naturally affected by WRI. In the central part of the plain River
55 Recharge (RR) from the Selouane River is responsible for the high groundwater salinity whilst
56 Mixing Processes (MIX) occur in absence of irrigation activities.
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60 **Keywords:** stable isotopes, irrigation, human impacts; PCA, MedPartnership; Morocco
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1. Introduction

The Mediterranean Basin is among the most arid regions in the world, where limited water resources are unevenly distributed in space and time (GWP, 2013). In particular, southern rim countries are either in arid or hyper-arid zones heavily depending on seasonal rainfall (World Bank, 2007). These regions have few permanent rivers (some of which carry runoff from other countries) and therefore often rely on fragile, and sometimes non-renewable, groundwater resources. As a result, more than 180 million people are considered water poor (i.e., their available amount of water is less than 1000 m³ of renewable water per capita per year), and an additional 60 million face water stress (i.e., their available amount of water is less than 500 m³ of renewable water per capita per year; GWP, 2013). The recent demographic growth, with the consequent increasing urbanization, agricultural demand and resource-intensive socioeconomic development, exerts additional pressures on scarce resources and fragile coastal ecosystems (Pereira, 2004). The situation is expected to worsen in the future, due to climate changes strongly impacting on precipitation amount and rainfall type (Navarra and Tubiana, 2013).

Under this pressure, the scientific community recognized the need to better assess the links between water and irrigated agriculture. The emphasis is especially focused on regional and local studies on groundwater suitability for irrigation purposes, resulting in numerous investigations based on different hydrogeochemical tools and indices (e.g. Watanabe et al., 2008; Tadesse et al., 2010; Ravikumar et al., 2011; Romanelli et al., 2012). Nonetheless, studies on the impact of irrigation on groundwater quality are mainly related to the assessment of diffuse pollution impact on groundwater quality, especially associated with fertilizers use and farming activities (e.g. Pionke et al., 1990, El Amrani et al., 2007, Boulabeiz, 2011; Boy-Roura et al., 2013), while relatively fewer report on the impacts of agriculture practices on salinity (e.g. Suarez, 1989; Kass et al., 2005; Qin et al., 2011) and on non-nitrogen contamination (Stigter et al., 2006; Qadir and Oster, 2004; Scanlon et al., 2010; Croon 2013; Oh et al., 2013).

Morocco, as most of the southern rim Mediterranean countries, is a highly water stressed nation with erratic rainfall and frequent droughts (GWP and DBSA, 2012). Under these generalized conditions of physical water scarcity (IWMI, 2007), the current water use patterns and withdrawals are considered unsustainable and water security is becoming a major limiting factor for the socio-economic development of the country (GWP and DBSA, 2012). Among all the productive sectors, agriculture is the largest water user, especially in dry seasons, and at the same time the main source of income for 80% of the rural population and 14-20% of the kingdom's Gross Domestic Product (EMWIS, 2013).

In Morocco, as worldwide, irrigated agriculture has become the predominant groundwater consumer, raising questions about resource sustainability and irreversible degradation, eventually

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4 leading to numerous cross-sector policy and management issues (Foster and Ait-Kad, 2012).
5 Indeed, improving agricultural efficiency would allow better allocating water resources and
6 adequately ensuring the access to safe drinking water and food.
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9 The region of Nador (North of Morocco) is a clear example of the aforementioned needs and
10 problems. Groundwater withdrawn from hand dug wells has been traditionally used to sustain
11 agricultural and domestic needs. Overexploitation and pollution from anthropogenic activities
12 (urban, agricultural and industrial) significantly impaired groundwater quality and soil fertility (El
13 Mandour et al. 2008, El Yaouti et al. 2008, El Yaouti et al. 2009).
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16 A preliminary investigation in the coastal aquifer of Bou-Areg and the adjacent lagoon of Nador was
17 conducted to characterize aquifer recharge and salinization processes (Re et al., 2013a). Results
18 showed that recharge is mainly due to mountain runoff, interacting with local recharge sources
19 (ephemeral rivers and irrigation waters) resulting in a complex system of mixed waters. The high
20 salinity of the aquifer was attributed to the coexistence of dissolution processes of evaporative
21 rocks and carbonates, water-rock interaction, and human impacts. Agricultural return flow was
22 identified as the main anthropogenic contribution to groundwater salinization in the central part of
23 the plain, also causing a general increase in nitrate concentrations. Only locally, nearby the
24 coastline, the high salinization was attributed to the presence of lagoon water intrusion.
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27 This work presents a second step for the complete assessment of recharge and pollution processes
28 in the Bou-Areg aquifer, chosen as one of the pilot study cases within the GEF-funded UNEP/MAP
29 MedPartnership actions (UNEP, 2010) intended to promote the protection of the marine and coastal
30 environment of the Mediterranean Basin.
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33 Within this framework this paper aims at (i) presenting new hydrochemical results confirming the
34 previous findings on aquifer dynamics, (ii) tracking the changes in groundwater composition related
35 to the use of irrigation water from different sources, (iii) highlighting seasonal variations and
36 impacted areas as a tool to assess the aquifer vulnerability, and (iv) fostering the protection of the
37 Mediterranean Basin presenting a replicable example of multi-tracer approach for the support of
38 groundwater management in rural coastal areas.
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41 The understanding of aquifers dynamics and their relation to agricultural practices is an asset for
42 ensuring long term management strategies for the protection of scarce water resources and
43 ensuring food security in arid and semiarid zones. Therefore results are used to evaluate the
44 sustainability of current agricultural practices and to propose guidelines for promoting improved
45 water management in the Mediterranean rim countries.
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47 48 49 50 51 52 53 54 **2. Site description** 55

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57 The Bou-Areg coastal plain is located in the Mediterranean shore of Morocco, in the region of
58 Nador (Fig. 1), and is considered one of the most important irrigated areas in North-Eastern
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4 Morocco. The alluvial plain of Bou-Areg covers a surface area of about 190 km² and it is limited by
5 the Gourougou volcanic massif (NW), the Beni-Bou-Iffrou Massif and the Kibdana range (SE); its
6 northern boundary coincides with the arched shape of the Lagoon of Nador (also known as
7 Marchica or Sebkhah Bou-Areg), while to the south the plain is connected to the adjacent Gareb
8 plain through the Selouane corridor (El Amrani et al., 2005). The Gareb plain is a basin oriented
9 WSW-ENE, separated from the Bou-Areg plain (NE) by the Plio-Villafranchian plateaux located in the
10 Selouane area, which overlays the volcanic formations of Beni Bou Ifrou (Dakki, 2003).

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13 The Bou-Areg unconfined aquifer is comprised of late-Pliocene to early Quaternary continental
14 sediments of variable thickness (up to about 100 m) and high permeability (up to $7 \times 10^{-4} \text{ m s}^{-1}$),
15 limited to the bottom by a Pliocene substratum of gypsiferous marls (Chaouni Alia et al., 1997)

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18 Its natural recharge is given by groundwater from the adjacent Gareb aquifer, rainwater, stream
19 water from the Selouane River and runoff coming from the massifs bordering the plain. Urban and
20 industrial wastewaters, together with irrigation returns, also contribute to the recharge. Hydraulic
21 head in the aquifer decreases from the bordering ranges towards the Lagoon of Nador (e.g. from 40
22 m a.s.l. close to the Kibdana range to sea level near the shore), and represents the natural outflow
23 from the aquifer system (Dakki, 2003).

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26 The hydrologic network of the area is characterized by the presence of several rivers -locally named
27 *oueds*-, some of which are ephemeral and often serve as sewage outflow for urban areas upstream
28 (Gonzalez et al., 2007). Only few streams are perennial, the most important being the Oued
29 Selouane (or Selouane River).

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32 The region of Nador, as with many other coastal plains along the Mediterranean, is characterized
33 by intense agricultural activities corresponding to more than 62% of the total surface area (El Yaouti
34 et al., 2008; FAO, 2012), with only 20-40% constituting equipped irrigation land (FAO, 2012).

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37 Due to the irregular precipitations and the different kinds of agricultural activities, irrigation water in
38 the past was provided by groundwater withdrawn from hand-dug wells and boreholes. The high
39 natural salinity of this water, its increase in the years, and the emerging soil fertility problems
40 fostered in the 1970's the construction of an irrigation network, whose main channel represents the
41 border of the irrigated area (Fig. 1). Freshwater coming from the Moulouya river is stored in the
42 Mohammed V dam (built in 1967), sent to the Mechraa-Hammadi dam (built in 1956 at the
43 Moulouya gorge, on the western edge of Beni Snassene), diverted towards the cultivated areas
44 through the Zebra channel and distributed via a network of minor superficial channels (Dakki,
45 2003). Diverted waters have been increasingly used to support agricultural activities through flood
46 irrigation, resulting in a slow decrease of groundwater salinity over the years (El Amrani et al.,
47 2005).

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50 The climate of the region is semi-arid with two distinct seasons: the warm period, generally from
51 May to October (average temperature of 23-24°C), and the cold season from November to April,
52 (average temperatures of 15°C; data available from WeatherSpark, 2014). Seasonal climatic
53 variations are usually moderate, with a general high level of humidity (50-80%) due to the proximity
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4 to the sea. Yearly average (1984-2014) precipitations range from 300 to 400 mm/y, and these are
5 generally more abundant in January-March and October-December (Fig. 2). The driest months are
6 May, June and July, with scarce or no rain episodes and higher mean temperatures, inducing a
7 water deficit. Mean rainfall data at the Melilla-Nador station is 487 mm, and annual potential
8 evapotranspiration, estimated using the Thornthwaite equation, rises up to 1030 mm. Total rainfall
9 in 2010, when the field survey was conveyed, was of 482 mm in the area. Therefore, considering
10 the timing of recharge from precipitation and irrigation, it is possible to define two periods (El Yaouti
11 et al., 2008): (i) recharge from precipitation and low use of groundwater/irrigation channel water
12 from October to April (L-IR) and (ii) no recharge from precipitation and high water irrigation from
13 wells and the irrigation network from May to October (H-IR) (Fig. 2).

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15 As previously mentioned, agriculture is the dominant sector in the region of Nador, and in particular
16 in the Plain of Bou-Areg with more than 100 km² (10180 ha) of irrigated land (Agence Urbaine de
17 Nador, 2013), exploited for both local consumption and for agro-industrial production (Khattabi and
18 El Ghazi, 2008). The main crops include cereals, olive, citrus fruit, grapes, sugar beet, vegetables
19 (with and without greenhouses). Another relevant sector is cattle husbandry, with sheep and goats
20 representing the main livestock, although dairy and poultry farming is also practiced in irrigated area
21 (Boelee and Laamrani, 2003).

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31 **Figure 1.**

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33 **Figure 2.**

34 35 36 37 **3. Methods**

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39 Two sampling campaigns performed in June 2010 (Re, 2011) and November 2010 (Fig. 3) allowed
40 for the collection of a total of 61 samples: 56 groundwater samples from private hand-dug wells and
41 boreholes (< 30 m deep, Tab. X1) in the Bou-Areg aquifer and 1 in the adjacent Gareb Plain; 3
42 samples in the Selouane River (Oued Selouane) and 1 sample from the irrigation channel.

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44 Sampling was carried out in the previously defined H-IR and L-IR periods in order to assess
45 seasonal effects and the impacts of irrigation activities. Moreover, H-IR samples have been
46 selected focusing in the agricultural zone whereas in L-IR the sampling network covered the whole
47 plain in order to facilitate the end-member identification.

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53 **Figure 3.**

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57 *In situ* measurements of electrical conductivity, pH and water temperature were performed during
58 both campaigns, using a WTW 340i multimeter. Total alkalinity was also determined in the field by
59 titration using an HACH alkalinity test kit (Tab X1).

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4 Samples for major ion analysis were filtered through 0.45 μm cellulose membrane and stored in
5 high density polyethylene bottles. Samples for cation analyses were preserved by addition of 5N
6 HNO_3 immediately after filtration. Samples for stable isotope analysis were collected and preserved
7 according to the procedures indicated by Clark and Fritz (1997).
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10 Chemical analyses of water samples were performed at the Department of Earth and Environmental
11 Sciences of the University of Pavia (Italy), using a Dionex DX 120 ion chromatograph. The error,
12 based on the charge balance, was calculated to be <5%. Hydrogen isotope compositions ($\delta^2\text{H}$)
13 were measured by water reduction over metallic zinc (Coleman et al., 1982), while oxygen isotopes
14 ($\delta^{18}\text{O}$) were analyzed by water- CO_2 equilibration at 25 $^\circ\text{C}$ (Epstein and Mayeda, 1953). Both results
15 are expressed in ‰ vs Vienna Standard Mean Ocean Water (V-SMOW; Gonfiantini, 1978;
16 Gonfiantini et al., 1995) with uncertainties (2σ) of $\pm 1\text{‰}$ and $\pm 0.1\text{‰}$ respectively. The $\delta^{13}\text{C}$ of DIC
17 was analyzed by direct acidification of the water sample with phosphoric acid (Kroopnick, 1974).
18 Results are expressed in ‰ vs Vienna Pee Dee Belemnite (V-PDB; Gonfiantini, 1978; Gonfiantini et
19 al., 1995), and the uncertainty (2σ) is $\pm 0.3\text{‰}$. $\delta^{15}\text{N}_{\text{NO}_3}$ and $\delta^{18}\text{O}_{\text{NO}_3}$ of dissolved nitrate were
20 analyzed following the procedures described by Silva et al. (2000) and refer to AIR and V-SMOW
21 (Gonfiantini et al., 1995) with uncertainties (2σ) of $\pm 0.5\text{‰}$ and $\pm 1\text{‰}$ respectively. All samples were
22 prepared and analyzed on a Finningan™ MAT 250 Mass Spectrometer at ISO4 private laboratory,
23 Turin (Italy).
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32 Saturation indices for gypsum and carbonates and partial pressure of CO_2 ($p\text{CO}_2$) were calculated
33 using PhreeqC, (Parkurst and Appelo, 1999) while total alkalinity was calculated with the software
34 AquaChem™ (Schlumberger Water Services).
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36 The spatial distribution of conductivity data was compiled using the kriging algorithm included in the
37 GS Surfer® package (Golden Software, Inc., Golden, CO, USA, 2008).
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39 Statistical data treatment, in the form of Principal Component Analysis (PCA; e.g., Chatfield and
40 Collins, 1980) was performed using the statistical package SPSS 15.0 for Windows® (SPSS, Inc.,
41 Chicago, IL, USA, 2004).
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45 **4. Results**

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49 Samples collected in the Bou-Areg aquifer have a general high Electrical Conductivity (EC; Tab X1),
50 ranging from 1.74 mS/cm (well 5b) to 10.52 mS/cm (well 4; average 5.971 mS/cm) in June 2010 and
51 1.78 mS/cm (well 5b) to 10.66 mS/cm (well 32; average; 6.08 mS/cm) in November 2010.
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53 Conductivity maps (Fig. 4), show a similar distribution of this parameter for both sampling
54 campaigns. Higher EC values are found in the south eastern part of the investigated area (near the
55 lagoon shore and close to the city of Kariat Arkmane; wells 3, 4, 15, 32), but also in proximity of the
56 Gareb Plain (wells 10, 27, 34) and in the so-called Selouane Corridor, hosting the river (e.g. wells
57 6b, 7b, 30, 37). These maps indicate that, despite some changes in the location of the sampled
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4 wells, both datasets are comparable and suggest the existence of multiple sources of salinity in the
5 investigated area.
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8 **Figure 4.**
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10 **Figure 5.**

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13 On the Durov diagram (Fig. 5) both campaigns can be clearly distinguished. Samples of the L-IR
14 period display a rather constant sodium-chloride hydrochemical facies whereas the relative
15 proportions of Na^+ and Cl^- decrease in samples of the H-IR period, resulting in a higher variability of
16 hydrochemical facies from sodium-sulphate to sodium-bicarbonate. Such seasonal variability is not
17 associated with dramatic changes in pH (range 7-8) or TDS (range 900-6000 mg/L; Tab. X2).
18 Nevertheless, it is interesting to note that in the TDS part of the Durov diagram, most of the
19 groundwater samples from the H-IR period plot on a line connecting the composition of the irrigation
20 channel and the Selouane River from the L-IR period (red dashed line in Fig. 5) whereas those from
21 the November campaign show also a linear trend, yet with lower Ca+Mg concentrations (in meq %)
22 than those from June.
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26 Finally, another feature of the studied area is the groundwater high nitrate content, with most
27 samples having nitrate concentrations exceeding the drinking water standard of 50 mg NO_3^-/L
28 (WHO, 2011) both in the rural and urban/peri-urban areas (Re et al., 2013b). The origin of this ion
29 has been investigated in detail using nitrate isotopes (Re et al., 2013a) allowing to identify manure
30 and septic effluents, especially in urban areas and in the central part of the plain, and synthetic
31 fertilizers in the agricultural zone as the main drivers for human induced pollution.
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108 **Table 1.**

5. Discussion

The characteristic high salinity of the Bou-Areg groundwater (cfr TDS Tab. X2 and EC values in Fig. 4, using EC as a proxy for salinity) has been pointed out by several authors (Chaouni Alia et al., 1997; El Amrani et al., 2005; El Mandour et al., 2008; El Yaouti et al., 2009; Re et al., 2013a) and generally attributed to the combined action of dissolution processes of evaporites and carbonates from Miocene substratum, water-rock interactions, as well as human inputs, such as agricultural return flows (Re et al., 2013a). As previously mentioned in both H-IR and L-IR periods all groundwater samples (apart from well 5b) have EC values greater than 2500 $\mu\text{S}/\text{cm}$, hence unsuitable for irrigation (Ayers and Westcot, 1994). In addition, in order to better support and promote new integrated irrigation strategies, it is also important to understand how irrigation (and consequently, agricultural return flows) affects groundwater quality.

5.1. Influence of irrigation on groundwater quality

The different sources and processes associated with groundwater recharge are well illustrated in Fig. 6, which compares chloride and sulphate contents in H-IR and L-IR periods. In the figure groundwater compositions of the Gareb Plain (J10,) and of the Selouane River, representing the saline end-members, are clearly distinguished from the irrigation channel. All samples collected in the Bou-Areg aquifer show elevated sulphate concentrations that are attributed to the influence of marly-gypsum outcrops, especially in the upstream zone, as pointed out by El Yaouti et al. (2009). Among the samples collected in H-IR, some have concentrations consistent with the composition of the Selouane River (River Recharge-RR), suggesting a possible occurrence of local lateral recharge from the river or a greater influence of groundwater from the Gareb Plain. Other samples show lower Cl values that might be attributed to a greater impact of waters from the irrigation channel, (Agricultural Return Flow-ARF) as proven by the composition of well J5b, the closest to the irrigation channel (Fig. 3; Cl⁻: 150 mg/L; SO₄²⁻: 385 mg/L; irrigation channel; Cl⁻: 125 mg/L; SO₄²⁻: 365 mg/L). These results indicate the occurrence of a dilution effect of irrigation waters infiltrating in the central part of the aquifer on the general high natural groundwater salinity (El Yaouti et al, 2009; Re et al., 2013a). Only few wells (mainly in L-IR) show intermediate concentrations with respect to the previously described groups. For those wells, salinity content originates from a mixing of irrigation and river recharge or by further dissolution of rock bearing minerals during infiltration of irrigation return, leading to a "natural water-rock interaction" hydrochemical facies (Mixing-MIX).

A different scenario can be deduced from the November 2010 samples (Water-Rock Interaction-WRI; Fig. 6), when the impact of irrigation activities is lower. Here none of the samples showed a composition consistent with that of the irrigation channel, and even well 5b shifts its November

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4 composition towards a decrease of SO_4^{2-} and an increase in Cl^- , tending to the natural high salinity
5 of the aquifer.
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7 Therefore, the groundwater hydrochemical composition in the two periods reflects the impact of the
8 irrigation practices. In fact, recharge from high precipitation and low water irrigation from the
9 channel occurs from November to April (El Yaouti et al., 2008) while a period of low precipitation
10 recharge and high water irrigation from both the canal and groundwater takes place from May to
11 October. Hence we can assume that in L-IR period the system is less affected by human induced
12 recharge and, therefore, it shows a more natural composition, due to dominant water-rock
13 interaction processes.
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19 **Figure 6.**
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23 Based on the features evidenced by the SO_4^{2-} versus Cl^- plot, groundwater samples have been
24 attributed to the previously described groups. For such groups, means and standard deviations of
25 each parameter in the different seasons are presented in Tab. X3.
26

27 While sulphates and chloride are indicative of water-rock interaction processes, carbonate equilibria
28 are highly reactive and can be used in conjunction with $\delta^{13}\text{C}$ of DIC to highlight biological processes
29 influencing groundwater chemistry. As already shown by the Durov diagram (Fig. 5), a major
30 difference in groundwater chemistry between H-IR and L-IR periods is related to the bicarbonate
31 content, which is sensibly higher in the former (Fig. 7A). This higher content is not strictly related to
32 an increase in groundwater pH, but rather to a higher pCO_2 (Fig. 7B), likely attributable to a more
33 intense biological activity in irrigated soils during the May-October period. Indeed, most of the
34 samples from the November campaign, and those from the Oued Selouane, lay on the pH-pCO_2
35 line corresponding to calcite saturation under open system conditions, whereas samples from the
36 June survey show a slight displacement towards higher pH values for similar initial pCO_2 conditions,
37 which indicates a tendency towards closed system conditions in the system $\text{H}_2\text{O-CO}_2\text{-CaCO}_3$
38 (Drever, 1982).
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45 Saturation indices for carbonates and gypsum are plotted with respect to TDS in Fig. 7C and 7D
46 respectively. It appears that, while groundwater samples from the L-IR period are generally
47 undersaturated with respect to carbonates, they become oversaturated during the H-IR period. The
48 same observation does not apply to gypsum saturation, which instead only depends on the
49 groundwater TDS.
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53 Additional information can also be obtained from $\delta^{13}\text{C}$ measurements in DIC (Fig. 7E), especially
54 when the behavior the carbonate system between the two periods is compared (Fig. 7F). Several
55 processes, all associated with agricultural practices, could potentially be responsible for changes in
56 Ca^{2+} content, as indicator of calcite weathering, and $\delta^{13}\text{C}$:
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- 4 1) an increase in the soil biological activity, increasing the soil pCO_2 which is buffered in the
- 5 subsurface by carbonate dissolution from the aquifer matrix. In this case, we should expect
- 6 high Ca^{2+} concentrations in the H-IR period, as well as high $\delta^{13}C$ values (assuming soil
- 7 carbonates have a dominant marine signature);
- 8
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- 10 2) an increase in the soil biological activity influencing the soil pCO_2 whereas irrigation water
- 11 dissolves gypsum precipitated in the unsaturated zone. In this case, Ca contents should
- 12 increase with no appreciable change in $\delta^{13}C$ values;
- 13
- 14
- 15 3) a direct influence of the isotopic composition of rechargewater

16 In Fig. 7E, a remarkable feature of groundwater samples from the L-IR period is that the previously
17 defined groups are characterized by distinctive Ca concentrations at varying pCO_2 , yet in
18 agreement with their water origin and hydrogeological evolution. However, in the H-IR period, WRI,
19 MIX and ARF groups display an increase in Ca^{2+} , at higher pCO_2 , likely resulting from the intense
20 root activity during the crop growing season. Regarding $\delta^{13}C$ a large range of values was observed
21 in both sampling periods, from $\delta^{13}C \approx -14.0\text{‰}$ to -5‰ . However, the main recharge sources (i. e., the
22 Oued Selouane and the irrigation channel), have enriched $\delta^{13}C$ contents (-6‰ and -9‰ ,
23 respectively) which are in agreement with those groundwater samples attributed to river recharge
24 (RR). Nevertheless, those samples whose origin is related to water-rock interactions (WRI) and
25 agricultural return flow (ARF) generally show lower $\delta^{13}C$ contents which are consistent with a higher
26 contribution of soil carbon dioxide of organic origin that dissolves calcite in its path towards the
27 water table, even during the L-IR, when pCO_2 and infiltration rate are small. In summary, $\delta^{13}C$
28 values confirm the different sources and processes occurring at the Bou-Areg aquifer that were
29 previously identified using hydrochemical data.

30 Furthermore, specific L-IR samples as those with $\delta^{13}C < -11\text{‰}$ that show a decreasing trend on the
31 plot of pCO_2 (Fig. 7F) and low SI_{cal} (Fig. 7C), belong to wells influenced by urban impacts. Indeed,
32 although the majority of the households ($\sim 47\%$) are connected to the sewage network, septic tanks
33 are still widely used ($\sim 29\%$ of the households) for wastewater evacuation in both the rural and peri-
34 urban area (Khattabi and El Ghazi, 2008), still representing one of the main drivers for groundwater
35 contamination.

36 It must also be considered that cation exchange in the aquifer may be responsible for a decrease in
37 Ca^{2+} and an enrichment in Na^+ content in groundwater (El Yaouti, 2009). This additional process
38 may explain some low SI_{cal} values during the November campaign (when the infiltration rate of
39 rainfall is assumed smaller than that of the irrigation period), and some general gypsum
40 undersaturation.

41 **Figure 7.**

42 **Figure 8.**

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4 Isotopic data ($\delta^2\text{H}$ and $\delta^{18}\text{O}$) also show different behaviors between the June and November
5 campaigns. In fact, considering the isotopic composition of the water molecule (Fig. 8) with respect
6 to the Global Meteoric Water Line (GMWL, Rozanski et al., 1993) all the samples appear to be
7 enriched in oxygen-18, suggesting the occurrence of evaporation processes, as already indicated
8 by Re et al. (2013a). Indeed, samples from both campaigns may be distributed along two
9 evaporation trend lines (namely, EVAP-1 and EVAP-2, in Fig. 8), with slopes around 5.2, which
10 correspond to evaporation with an environmental humidity of 75% (Gonfiantini, 1986); this value is
11 in agreement with the mean annual atmospheric humidity in the Nador Airport weather station
12 (WeatherSpark, 2014).

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17 In the L-IR period, when, based on hydrochemical data, groundwater compositions are mostly
18 determined by natural water-rock interaction processes, the regression line ($\delta^2\text{H} = 5.34 \delta^{18}\text{O} - 6.60$;
19 $R^2 = 0.77$; $n = 26$, i.e. excluding N31 which is out of the Bou-Areg basin) crosses the GMWL at a
20 value of about -5.9‰ in $\delta^{18}\text{O}$ and -38‰ in $\delta^2\text{H}$. This value could be attributed to the mean rainfall
21 isotopic composition in the Nador area, although no measured data are available, as it is roughly in
22 agreement with the interpolated precipitation compositions for the region (Bowen and Revenaugh,
23 2003). This finding suggests that during the L-IR period groundwater is mostly recharged by local
24 precipitation affected by evaporation prior to infiltration (line EVAP-1 in Fig. 8). According to
25 Gonfiantini's equations, the largest evaporation percentage would be less than 5%, indicating a
26 short ponding time in the field surface or evaporation in the vadose zone (Gonfiantini, 1986). In the
27 H-IR period, the more depleted values correspond to the irrigation channel and well J5b, the closest
28 to the irrigation channel. Assuming the isotopic composition of irrigation water as an end-member, a
29 similar evaporation process could account for the isotopic composition of most groundwater
30 samples (EVAP-2) influenced by irrigation water. The lines EVAP-2 and GMWL cross at an isotopic
31 value of about -7‰ in $\delta^{18}\text{O}$ and -47‰ in $\delta^2\text{H}$. Such depleted values for meteoric waters may
32 correspond to precipitation at higher altitudes in the Atlas chain, and they are seldom recorded in
33 the south-western Mediterranean coast (Saighi, 2005). Indeed, such hypothetical values are in
34 good agreement with the isotopic composition of the Moulouya river feeding the Mohammed V
35 basin (-6.92‰ in $\delta^{18}\text{O}$ and -47.7‰ in $\delta^2\text{H}$; inverted triangle figure Fig. 8; IAEA, 2010.), where
36 water is stored and likely evaporated prior to being diverted to the Zebra irrigation channel. The
37 irrigation channel sample shows an evaporation percentage of $\approx 5\%$, based on isotopic data, and
38 subsequent evaporation after irrigation increases it up to a 7.5% (Gonfiantini, 1986). As already
39 evidenced by hydrochemical parameters, and also supported by isotopic data, mixing with seawater
40 is not occurring in the Bou-Areg aquifer, except in a few wells nearby the Nador Lagoon. Therefore,
41 the prevailing mechanism for groundwater salinization is the dissolution of salt minerals and
42 agricultural as well as urban inputs to the subsurface.

5.2 Statistical data treatment

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4 PCA was performed in this study to better support geochemical evidences, identify end-members,
5 and understand the seasonal variability and its relation with agricultural activities (Menció et al.
6 2012). Three varifactors were extracted (Table 1) with a total cumulative variance explained of
7 about 75%. In particular, each varifactor provides the following hydrogeological associations:
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- 11 • Varifactor 1 (VF1) explains 41.3% of the total variance and is participated by EC, Ca²⁺, Cl⁻,
12 Si_{Gypsum}, Na⁺, Mg²⁺, SO₄²⁻, and δ¹³C. This varifactor is thus representative of the total
13 aquifer salinization, including both mixing processes and lateral river recharge (Fig. 9A). For
14 both the campaigns, the samples with positive scores (VF1>0) belong to RR or MIX groups,
15 while lower scores (VF1<0) characterize the samples where agricultural return flow and
16 water rock interaction processes dominate.
17

18 Hence, high values of V1 imply high aquifer salinization, but do not allow for a clear
19 distinction between L-IR and H-IR (as VF1 also includes K⁺, Ca²⁺ and SO₄²⁻).
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- 21 • Varifactor 2 (VF2), with 21.9% of the variance, is highly correlated with HCO₃⁻ total alkalinity
22 and pCO₂, and refers to carbonate equilibria. HCO₃⁻ increase (and positive relation with
23 VF2) for H-IR samples (Fig. 9B) was previously explained by their high bicarbonate content
24 being determined by the increased biological activity (i.e., higher pCO₂) during the crop
25 growing season. In addition, the negative correlation between VF2 and δ²H (Table 1)
26 suggests that high irrigation activities correspond to samples with lower δ²H, with the lowest
27 values associated with the irrigation channel. VF2, related to δ²H also distinguished those
28 samples along the evaporation lines EVAP1 and EVAP2, which indeed relates to the water
29 origin in the area. This reinforces the role of the irrigation channel water for aquifer
30 recharge, especially during H-IR.
31

- 32 • Varifactor 3 (VF3), explaining 12.2% of the variance, includes δ¹⁸O (highest weight), δ²H,
33 NO₃⁻, and it is negatively correlated with δ¹³C. This varifactor corresponds to the nitrate
34 pollution trend (Fig.9C). In fact, an increase in VF3 is associated with an increase in [NO₃],
35 in most of the cases due to agricultural pollution in H-IR. By comparison, most of the
36 samples of the L-IR period, with lower NO₃⁻ contents, generally display negative scores for
37 VF3. The positive correlation of VF3 with δ¹⁸O and δ²H indicate that nitrate content is larger
38 in isotopically enriched samples corresponding to low altitude recharge (Menció et al.,
39 2011). L-IR samples with a larger VF3 score are located in the urban/peri-urban area of
40 Nador (N8b, N36, N38 and N39) and reflect the strong contribution of domestic pollution, as
41 pointed out by Re et al. (2013b). Only for sample N33, located in the rural zone and having
42 the highest δ¹⁸O value for the L-IR campaign but low nitrate contents, the large VF3 score
43 is likely related to the higher contribution of δ¹⁸O in VF3. On the other hand samples J1, N7
44 and N28 (Fig.9C) are also likely to be affected by local (point source) contamination (i.e.
45 manure and/or septic effluents) in the agricultural part of the Bou-Areg Plain.
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4 **Figure 9.**
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7 Fig. 10 shows the correlation between the main varifactor scores for each sample. In the plot of VF1
8 (salinization) versus VF2 (soil biological activity; Fig. 10A), samples are consistently grouped with
9 the groups identified by the geochemical interpretation. In particular, VF2 clearly distinguishes L-IR
10 from H-IR samples. The dominant driver for groundwater composition in H-IR is the increasing soil
11 biological activity during the crop growing season, as also detected by comparing VF2 and VF3
12 (Fig. 10C). Concerning those samples under the influence of the Selouane River recharge (RR),
13 they appear to be dominated by different factors, according to the season: in L-IR, groundwater
14 composition is mainly influenced by a recharge from the Selouane River (plotting in the second
15 quadrant, Fig.10A), while in H-IR groundwater samples are also affected by biological activity
16 (orange samples in the first quadrant, Fig. 10A), as indicated by a low, yet positive, score of VF2.
17 On the other hand no relationship between VF1 and VF2 is visible for WRI samples and the
18 irrigation channel (third quadrant Fig 10A), highlighting the relatively low salinization due to the
19 absence of irrigation activities.
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21 In the VF1 (salinization) versus VF3 (nitrate pollution) plot, different situations can be described.
22 Some samples are characterized by high values of VF3 (fourth quadrant, Fig.10B), highlighting the
23 occurrence of agricultural pollution (ARF) and contamination from manure and septic tanks in the
24 urban area (see also Fig. 10C). In the second quadrant (high scores in VF1 and low scores in VF3)
25 plot the RR and some of the MIX samples, which are characterized by high salinization rather than
26 nitrate pollution, whereas neither salinization nor nitrate pollution affects the composition of the
27 irrigation channel waters and the L-IR samples in the agricultural zone (Fig. 10B). In other words,
28 samples in quadrants I and IV are those related to urban pollution or groundwater use for irrigation
29 (local recharge - given the high contribution of $\delta^2\text{H}$, $\delta^{18}\text{O}$ in VF3 - and high nitrate pollution).
30 Samples with the lowest NO_3^- pollution and river influence lay in quadrant II, while those samples
31 with highest channel influence, lower $[\text{NO}_3^-]$ (in H-IR), and the aquifer signature characteristics (L-
32 IR) are located in quadrant III.
33

34 PCA analysis synthesizes the fact that samples can be distinguished by soil biological activity
35 magnitude, as HCO_3^- , pCO_2 and $\delta^{13}\text{C}$ contents, while they may be affected by a wide range of
36 salinization and nitrate pollution levels, depending on land use and on anthropogenic pressure on
37 water resources. The two main groundwater recharge sources, the irrigation channel and the
38 Selouane River, consistently relate to the samples scores, supporting the hydrogeochemical
39 conclusions reached in the previous discussion.
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55 **Figure 10.**
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59 **5.3 Water management issues**
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6 When groundwater resources, such as those found in the Bou-Areg aquifer, are used for irrigation,
7 their high salinity content can eventually accumulate in the crop root zone and cause yield reduction
8 (Ayers and Westcot, 1994). As a result, the crop is no longer able to extract sufficient water from the
9 salty soil solution, resulting in a water stress for a significant period of time. Good quality irrigation
10 water is therefore essential for ensuring crop productivity, and water salinity is thus a serious
11 limiting factor, especially for crops with limited salt tolerance (fruits, citrus, beans). Although since
12 the construction of the irrigation channel groundwater withdrawal for this purpose significantly
13 decreased, farmers with equipped wells still resort to –costless- groundwater, neglecting its
14 negative effect on crop productivity.
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17 Besides the ascertained salinity of the aquifer and its impact on agricultural activities, it is also
18 important to define how irrigation (and agricultural return flows) affects the natural groundwater
19 quality and quantity, in order to better support and promote new integrated irrigation strategies.
20 Moreover, excluding groundwater, the Moulouya River resources, which feed the irrigation channel
21 in the study area, represent almost 90% of the water used for irrigation in all the North-Eastern
22 coastal plains of Morocco (Sebra, Bou-Areg and Gareb plains). However, its runoff is severely
23 threatened by the combined impact of climate change and silting of the Mohammed V dam, where
24 water is stored prior to distribution (Dakki, 2003). In addition, the rapid demographic increase and
25 growing tourist sector (for instance, the new MarchicaMed project, along the sand bar that limits the
26 Nador lagoon with the open sea) will also compete for water resources allocation, and will
27 contribute to the decrease in water available for irrigation. A lack of adequate planning taking into
28 account the hydrodynamics of the hydrogeological system may represent a further, irreversible
29 impact to the area, as already observed in many locations, especially along the Mediterranean
30 coast.
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33 In summary, some specific issues arise from the Bou-Areg aquifer study that may lead to further
34 improvements in water resources management in the region, as well as being replicated in similar
35 contexts along the Mediterranean shore:
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- 37 • Despite a high vulnerability to agricultural (and urban) pollution the Bou-Areg coastal
38 aquifer has shown a good resilience to intense agricultural activities as proven by the
39 seasonal hydrochemical variations.
- 40 • Channel water from the Moulouya River has the best quality with the lowest EC values and
41 the lowest [NO₃]. Therefore this resource must be protected from contamination and
42 excessive exploitation in its headwaters, in order to allow irrigation in the lowlands. Further
43 agricultural and tourist development should face water scarcity (present and future) to
44 ensure smart water allocation that fosters economical as well as social development and
45 environmental preservation.
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- Selouane River presents the highest EC values and medium [NO₃], hence groundwater withdrawal close to this river, possibly causing head level decrease, can lead to stream leakage to the aquifer and thus, aquifer salinization. Extended irrigation of the area using channel water may lead to a decrease the infiltrating rate of stream water, thus enhancing groundwater quality and allowing a larger discharge for surface dilution processes and ecosystem services.
 - The highest [NO₃], concentrations have been mainly attributed either to urban or to agricultural sources. Previous studies (Re et al., 2013a; Re et al., 2013b) pointed out that septic effluent (especially in urban areas), agricultural return flow, or a mixing of these processes, are responsible for such high concentrations. This implies that improved sanitation facilities and a better control on groundwater withdrawal and fertilizer use are of paramount importance for both water quality protection and food security issues.
 - Finally, the increasing climatic uncertainty coupled with the potential emerging conflicts in water use (agriculture, tourist and industrial sector) will probably put water scarcity issues at the highest level of planning and management discussions. Alternative irrigation sources, such as reclaimed wastewater reuse, may be of capital relevance. Moreover, the available hydrochemical information relates only to the upper levels of the Bou-Areg aquifer system. Research on groundwater quality in deeper layers may evidence alternative water resources (e.g. large scale flows system, paleowaters) whose sustainable exploitation should be carefully evaluated.

36 **Figure 11.**

37 38 39 **6. Conclusions**

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43 In the framework of a common strategy for the safeguarding and long-term protection of the Mediterranean coastal zones, this study presents a hydrogeochemical characterization of aquifer dynamics and pollution of the Bou-Areg aquifer, identified as one of the pollution hot-spots under investigation in the MedPartnership actions.

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49 Results of the hydrogeochemical and multivariate analysis of the Bou-Areg aquifer on samples collected in 2010 show a strong seasonal variation of groundwater quality, mainly associated with the impact of agricultural practices. In absence of irrigation activities (L-IR), natural water-rock interaction processes and pollution inputs from civil sources control the salinity of the system, together with a possible contribution of groundwater from the nearby Gareb Plain through the Selouane corridor. On the other hand, during the irrigation season (H-IR) the impact of irrigation practices is associated with an increase of the soil biological activity and of agricultural pollution, mainly consisting of high nitrate concentrations in groundwater.

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4 The coupled hydrogeochemical and multivariate analysis proved to be a useful tool for the clear
5 identification of following dominant processes governing the aquifer's hydrochemistry in the different
6 seasons: (Fig. 11):
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- 8 • Water Rock Interaction processes (WRI) dominate the composition of most of groundwater
9 samples in L-IR. In some cases, especially in the peri-urban area of Nador (left bank of the
10 Selouane River) human activities also affect the natural composition of the aquifer, resulting
11 in severe nitrate contamination (Re et al., 2013a);
12
- 13 • Agricultural Return Flow (ARF) mainly affects groundwater salinization in H-IR in the same
14 areas naturally affected by WRI. However, a strong seasonality effect governs groundwater
15 quality in the aquifer,
16
- 17 • River Recharge (RR) is responsible for the high salinity of the samples in the proximity of
18 the Selouane River. This can be associated with natural lateral discharge or to stream
19 capture driven by groundwater abstraction in the irrigation season. Groundwater flow from
20 the Gareb Plain may contribute to the background hydrochemical composition of the
21 aquifer.
22
- 23 • Mixing Processes (MIX) occur along the Selouane corridor, in absence of irrigation
24 activities. Here, groundwater chemical composition is influenced both by the Selouane
25 River, or groundwater coming from the Gareb Plain, and WRI processes.
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28 As discussed, these findings have sound management implications and must be taken into account
29 for future agricultural development plans of the region. In fact, the high aquifer vulnerability implies
30 the need to better control both the quantity and the quality of irrigation waters. Managing the
31 impacts of agricultural return flow and urban inputs will enhance groundwater quality with relevant
32 positive effects on crop efficiency, soil salinization, and environmental issues. Conversely, results
33 highlight the need for a more efficient use of available water resources coupled with the
34 identification of alternative irrigation sources, and the implementation of more efficient agricultural
35 practices.
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45 **Acknowledgements**

46 This study was partially supported by the Italian Ministry for Environment, Land and Sea as a
47 contribution to the GEF UNEP/MAP Strategic Partnership for Mediterranean Sea Large Marine
48 Ecosystem (MedPartnership) under the sub-component executed by UNESCO-IHP on the
49 "Management of Coastal Aquifer and Groundwater". The authors would like to thank Prof. N. El
50 Hamouti and Mr. R. Bouchnan for the help during the June 2010 sampling campaign, and Dr. T.
51 Lovato for his support during the November 2010 one. We are grateful to UNESCO-IHP and
52 UNESCO office in Rabat (Morocco) for the technical support during the execution of the project. We
53 thank Dr. Enrico Allais and ISO4, for their help in the chemical and isotope analysis.
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56 In loving memory of Prof. G.M. Zuppi, coordinator of the project.
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Table 1[Click here to download Table: Table 1.docx](#)

Variable	VF1	VF2	VF3
EC	<u>0.959</u>	-0.136	0.013
Ca ²⁺	<u>0.943</u>	0.087	-0.026
Cl ⁻	<u>0.936</u>	-0.070	-0.106
SI _{Gypsum}	<u>0.902</u>	0.142	0.094
Na ⁺	<u>0.852</u>	-0.298	-0.063
Mg ²⁺	<u>0.812</u>	0.331	0.006
SO ₄ ²⁻	<u>0.777</u>	0.023	0.123
K ⁺	<u>0.611</u>	0.081	0.227
HCO ₃ ⁻	-0.074	<u>0.952</u>	0.154
Total Alkalinity	-0.066	<u>0.947</u>	0.151
pCO ₂	0.092	<u>0.882</u>	0.146
δ ² H	0.184	<u>-0.645</u>	<u>0.588</u>
δ ¹⁸ O	0.122	0.031	<u>0.860</u>
NO ₃ ⁻	-0.236	-0.150	<u>0.601</u>
δ ¹³ C	<u>0.467</u>	0.074	<u>-0.474</u>
Eigenvalues	6.201	3.283	1.831
% Variance explained	41.339	21.884	12.206
% Cumulative variance	41.339	63.223	75.430

Table 1. Loadings of 15 variables on 4 significant varifactors (VFs). Underlined values represent relevant loadings.

Figure 1

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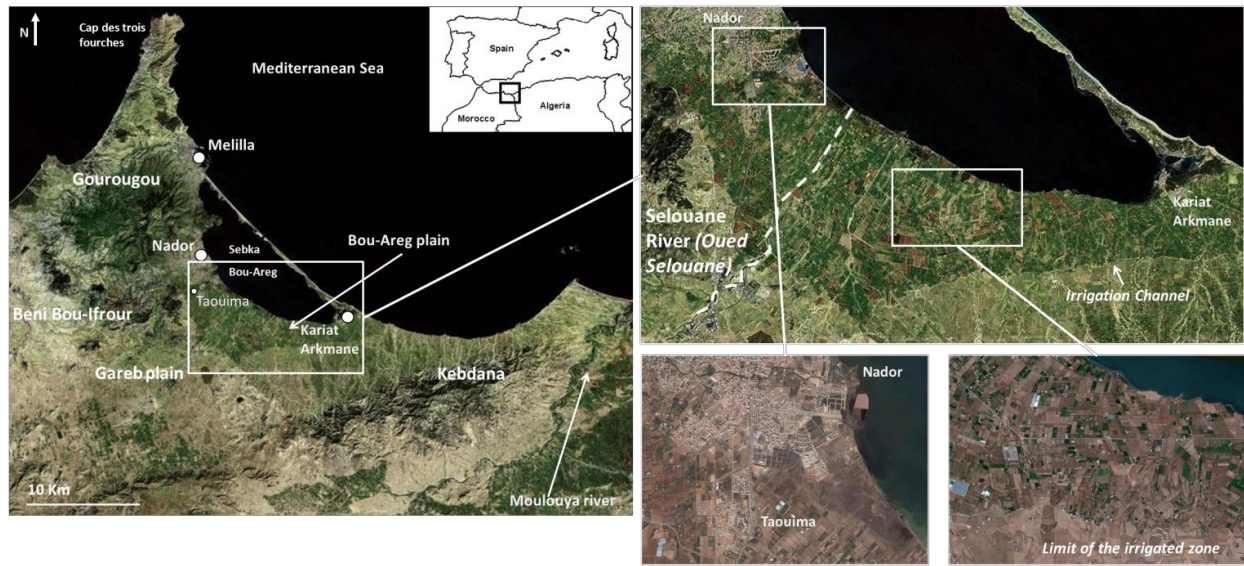


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Figure 2

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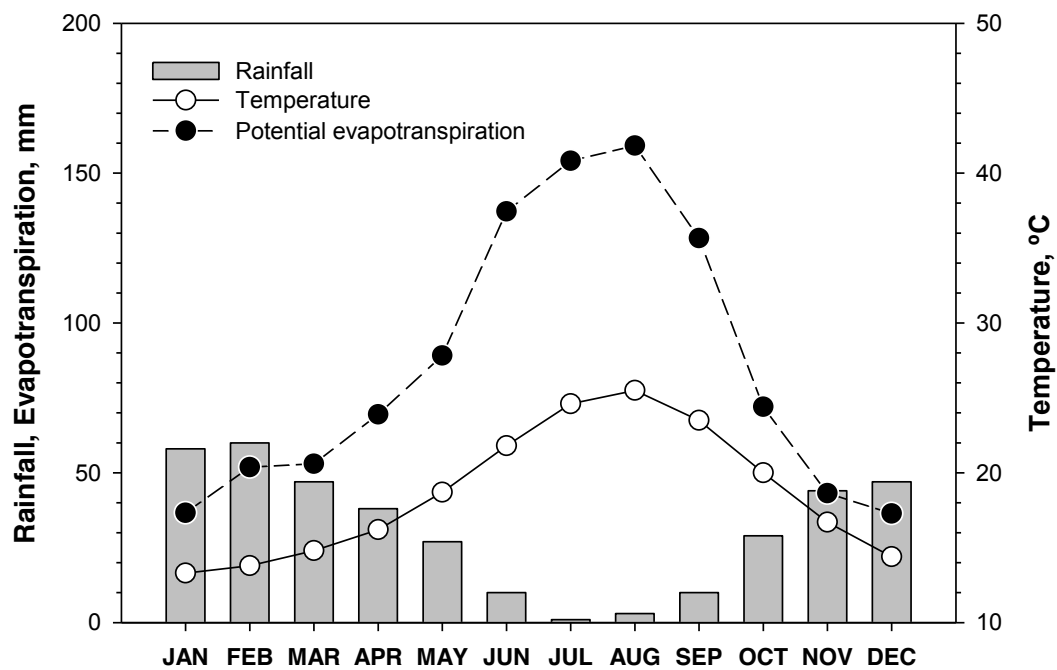


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Figure 3

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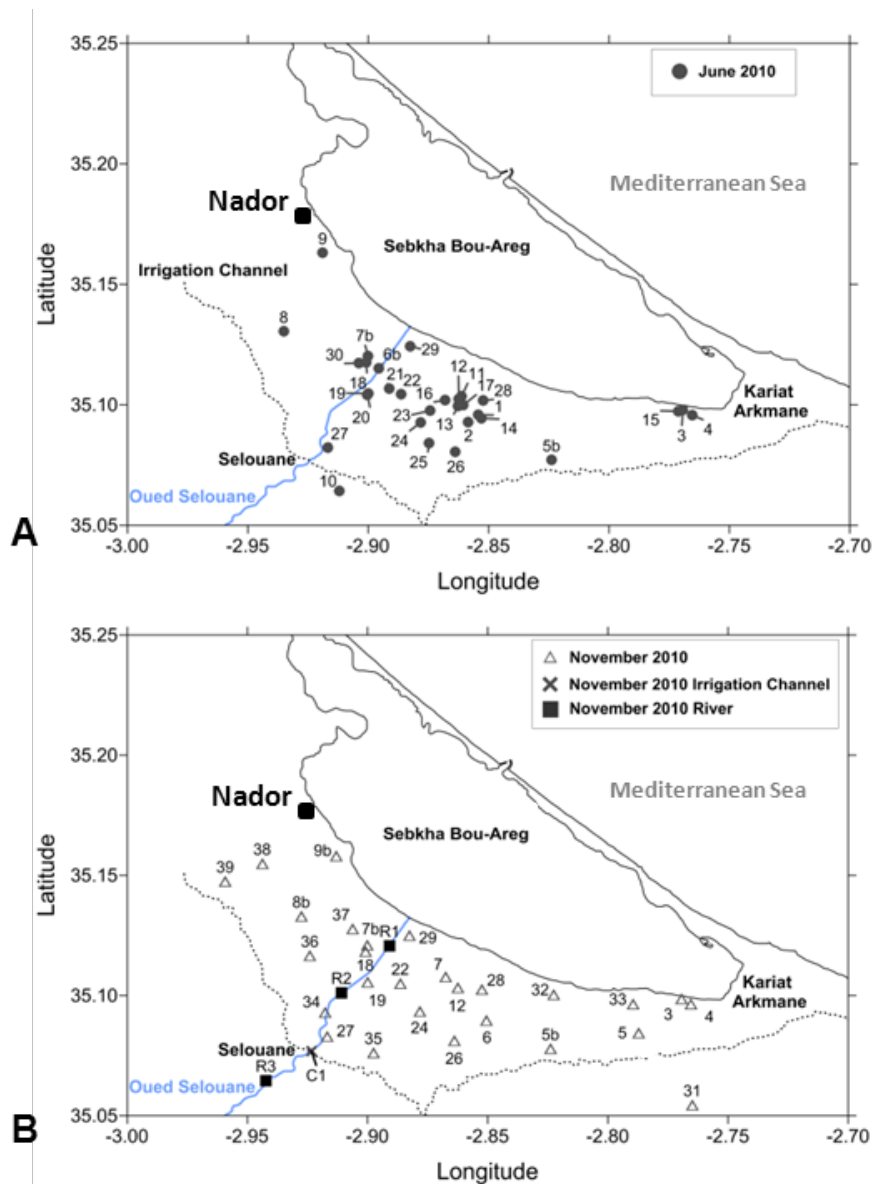


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Figure 4

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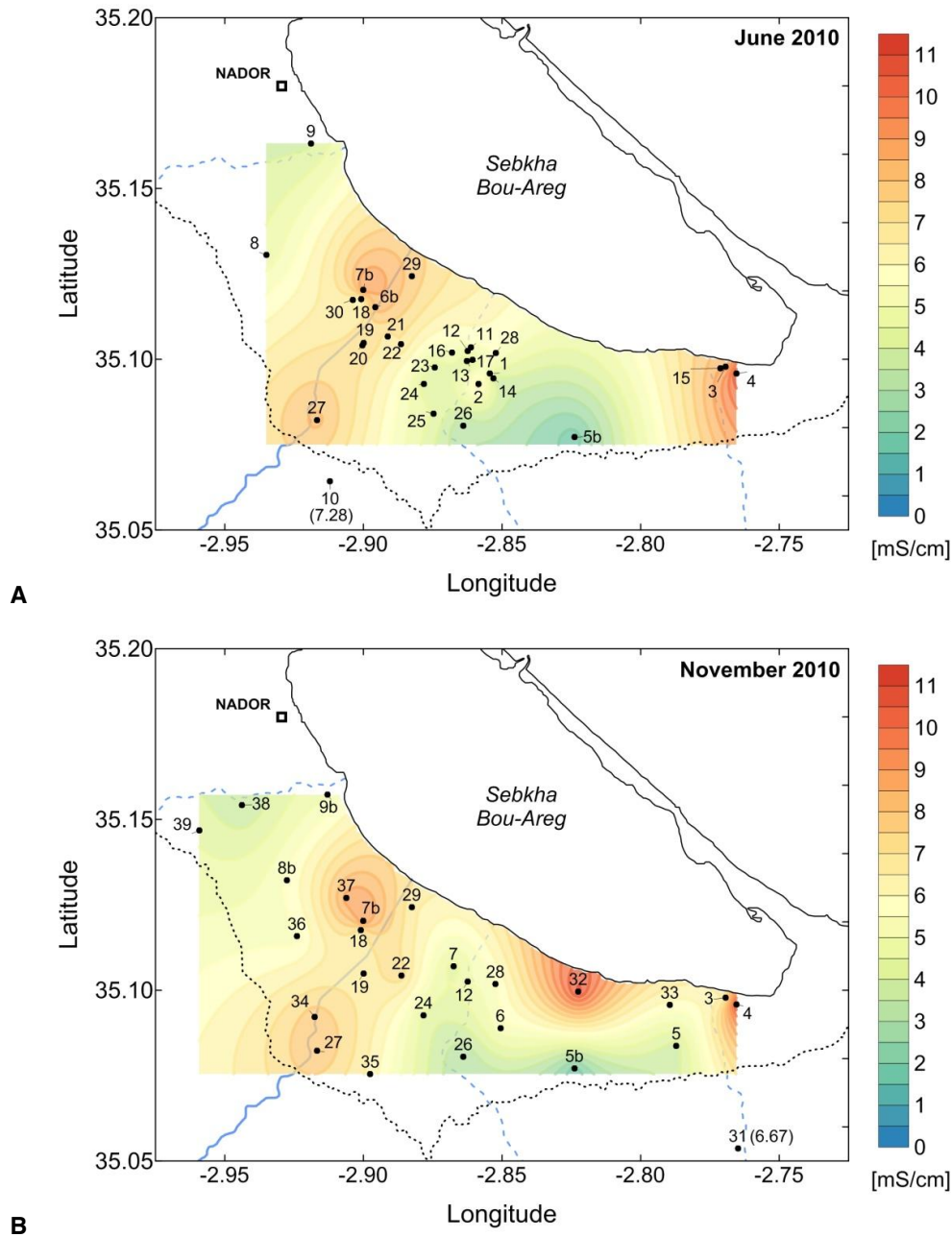


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Figure 5

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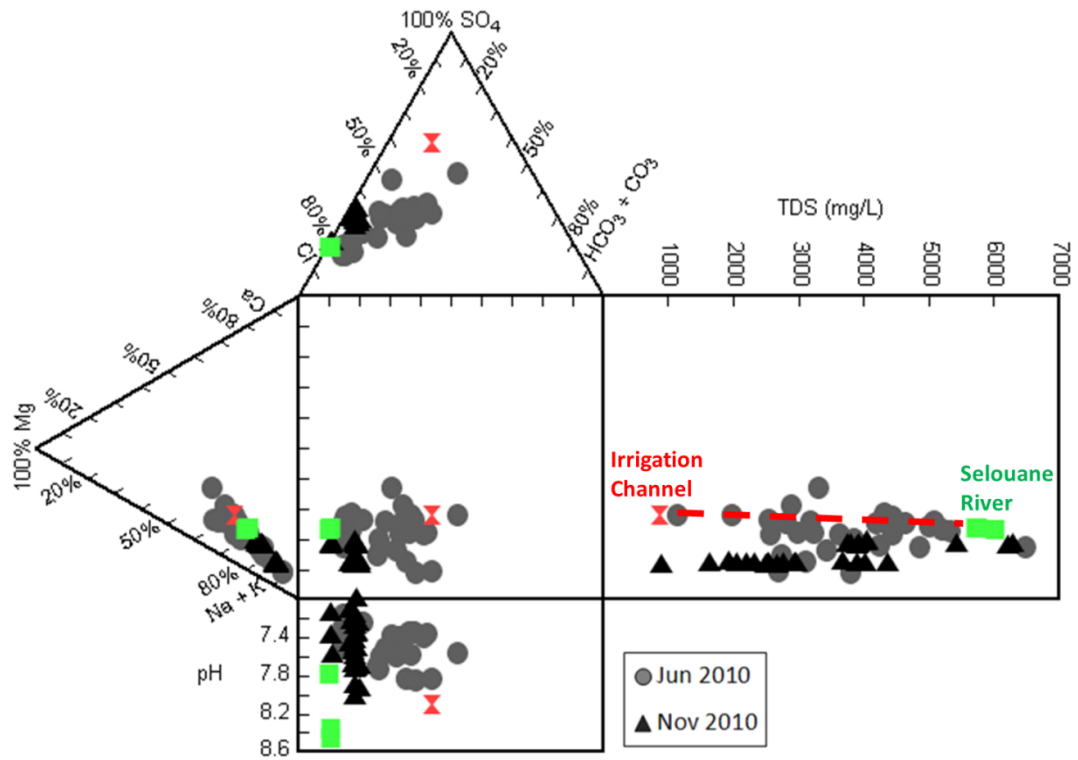


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Figure 6

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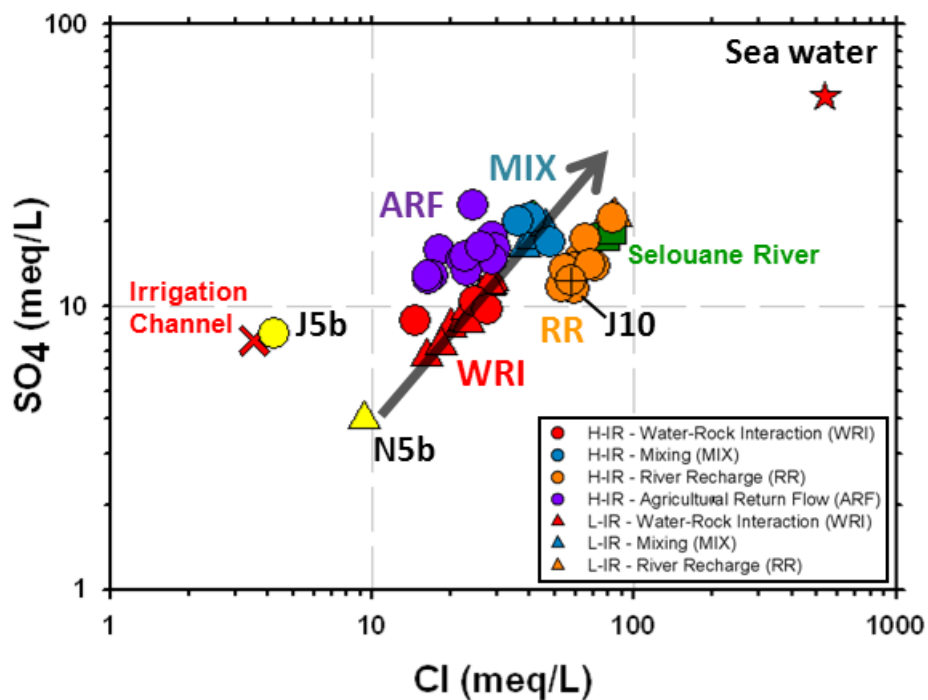


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Figure 7
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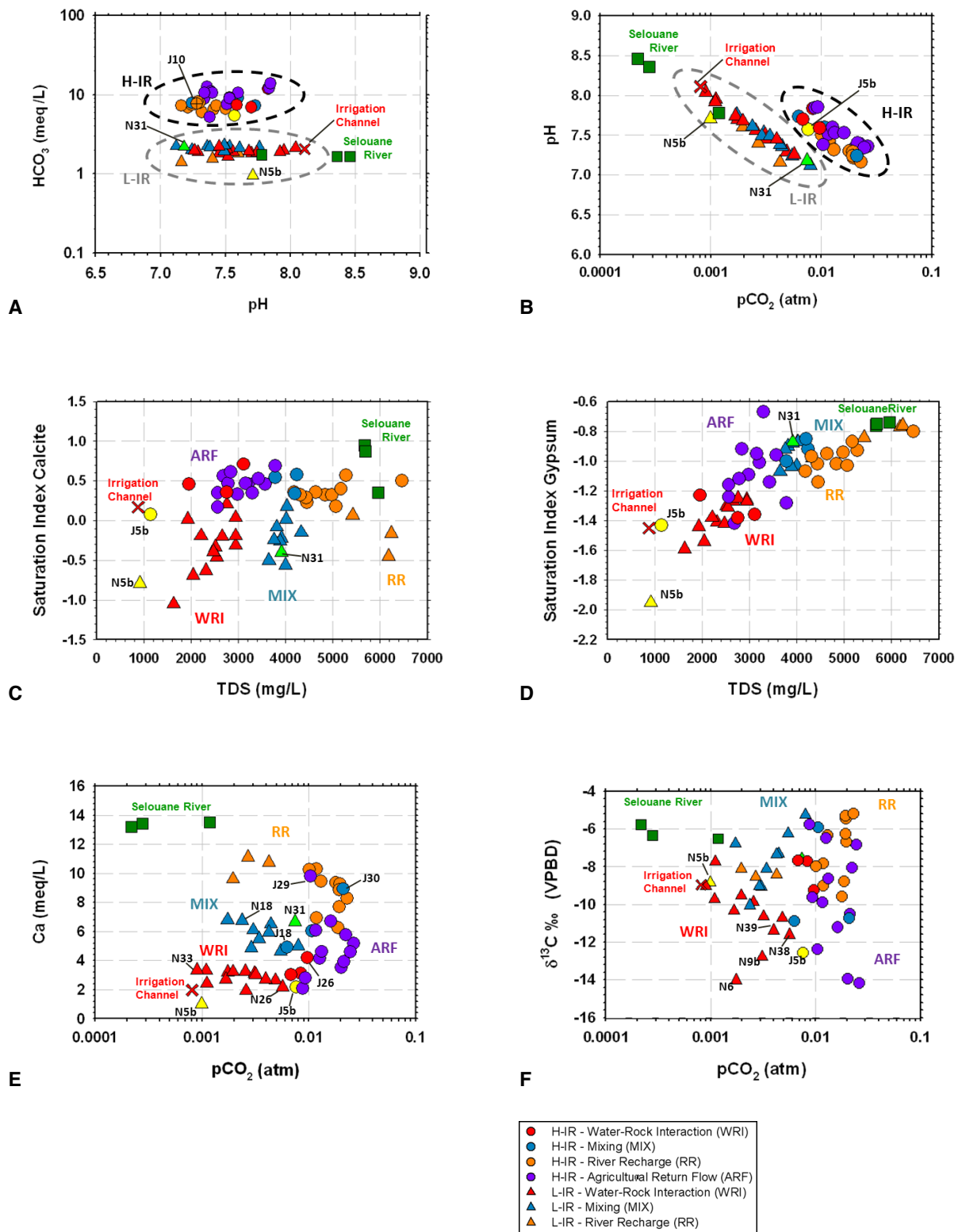


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Figure 8

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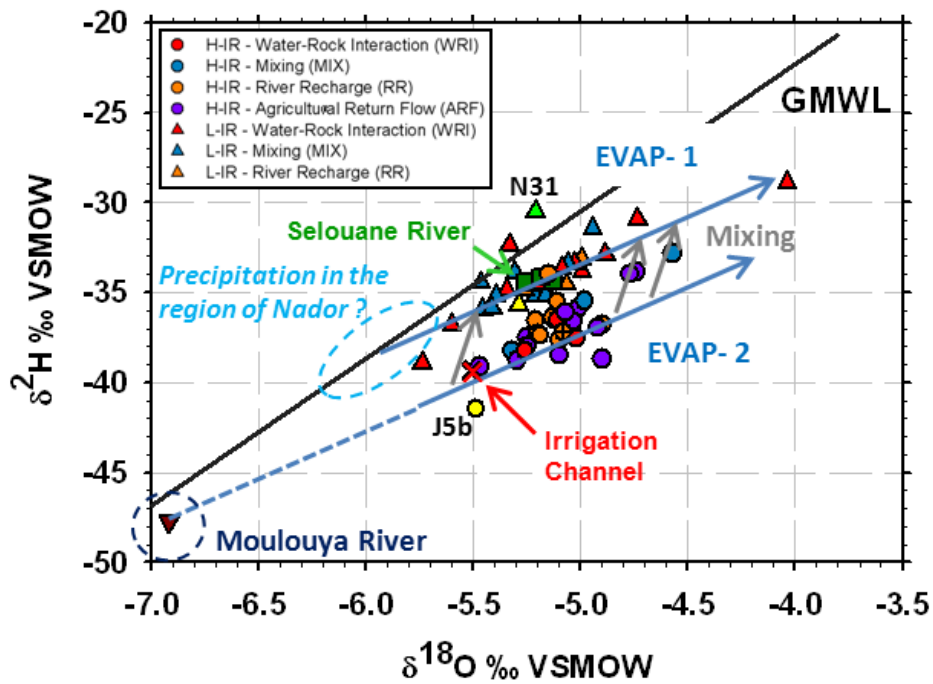


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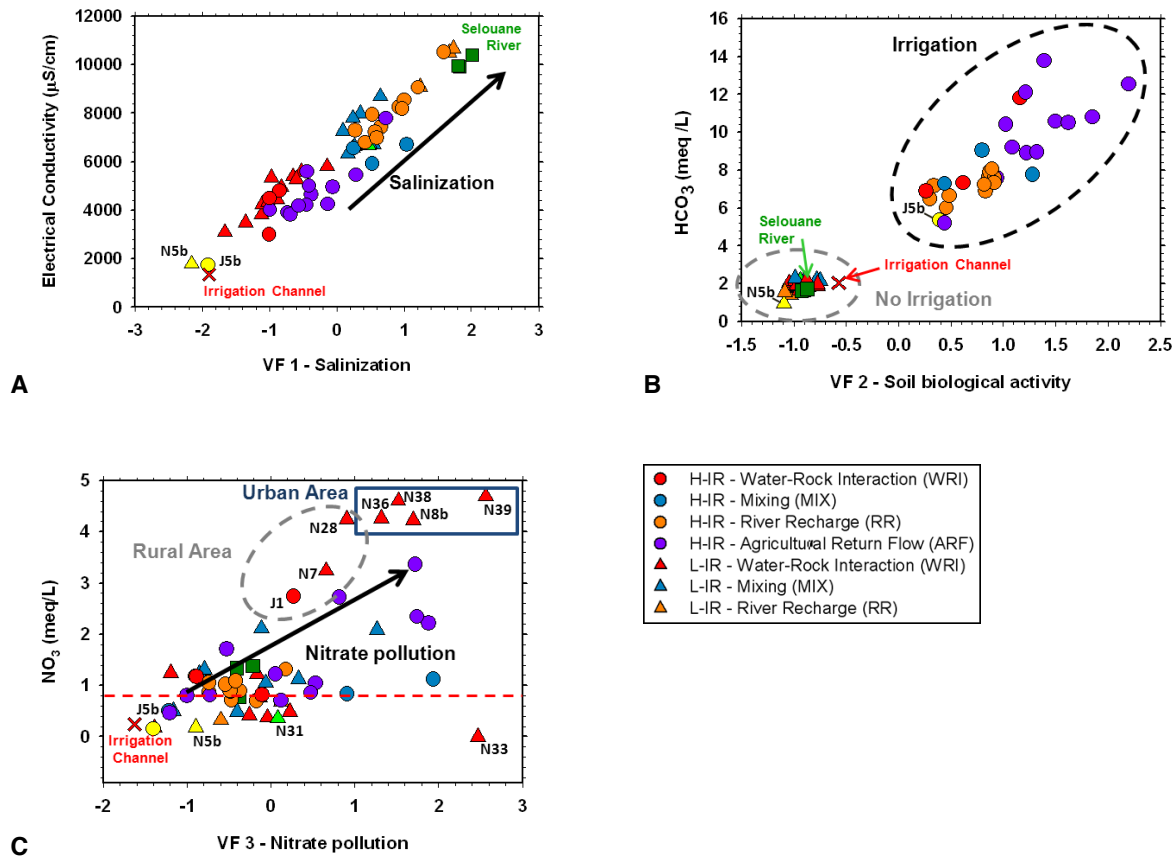


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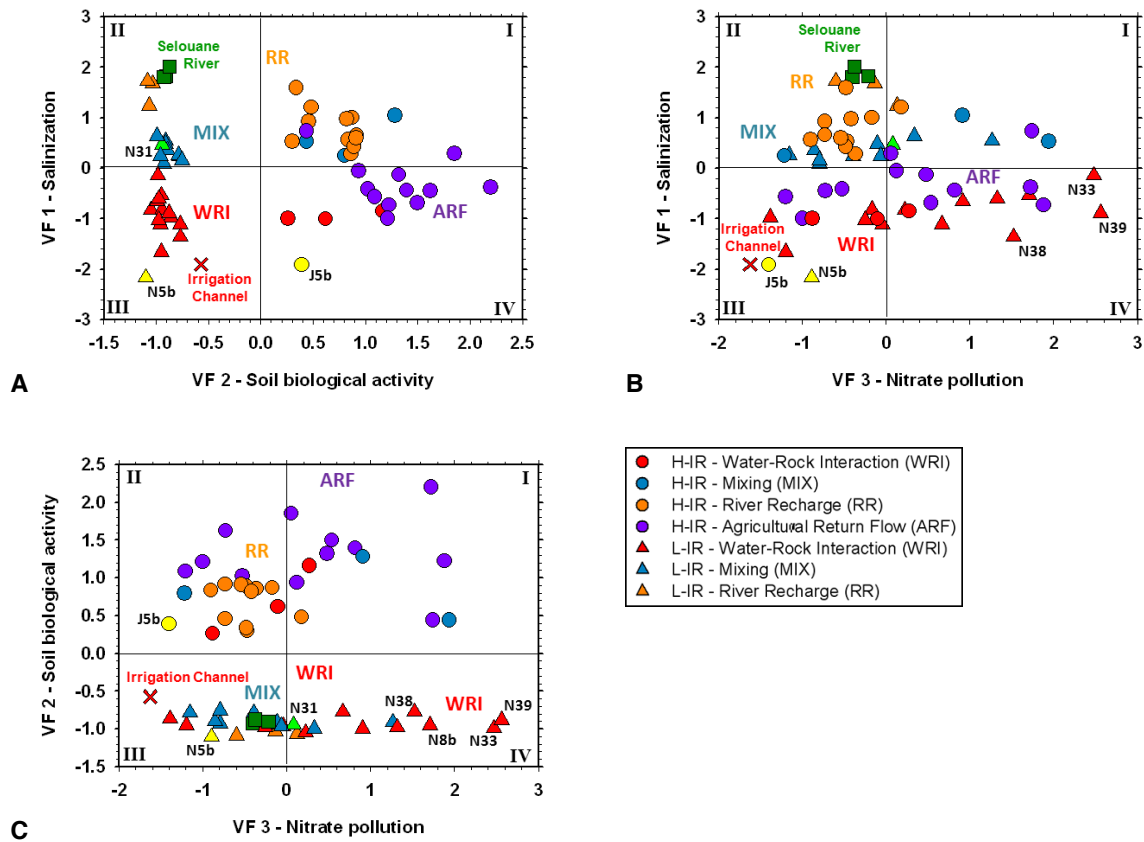


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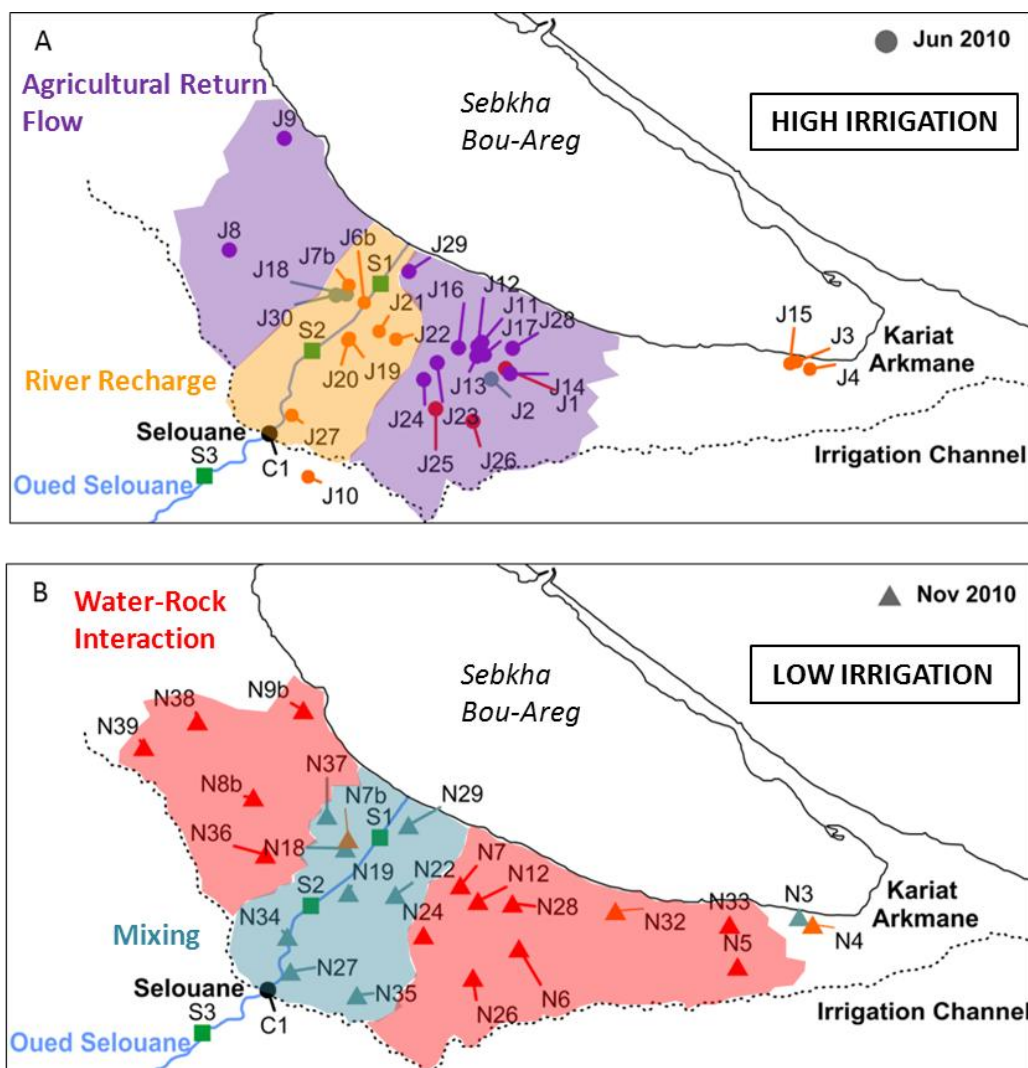


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