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Water quality aspects from Spanish sites to support managed aquifer recharge (MAR) guidelines not based on maximum allowable concentration standards

Aspetti sulla qualità dell'acqua in siti spagnoli di ricarica delle falde in condizioni controllate per valutare le linee guida a livello nazionale per evitare l'utilizzo delle concentrazioni massime ammissibili.

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Riassunto

La maggior parte dei paesi che hanno linee guida tecniche o regolamenti per la ricarica artificiale o in condizioni controllate degli acquiferi (Managed Aquifer Recharge, MAR), che includono aspetti legati alla qualità dell'acqua, si basano sull'istituzione di standard o Concentrazioni Massime Ammissibili (CMA) per regolare la qualità dell'acqua percolata o iniettata in un acquifero. Il numero di parametri in queste linee guida varia considerevolmente (da 6 in Spagna a 156 negli USA) e spesso si applica a tutti gli acquiferi all'interno dei confini amministrativi (ad esempio, il territorio nazionale), indipendentemente dalla natura del mezzo ricevente, dalla profondità della falda acquifera e da altri fattori chiave.

Undici sistemi MAR in Spagna sono stati studiati (otto operativi e tre sperimentali, con un numero limitato di dati provenienti da tre siti), caratterizzando sia la qualità dell'acqua di ricarica che l'acqua risultante dai processi di interazione tra acqua di ricarica-suolo-zona insatura-zona satura dell'acquifero. In tutti i casi, si osserva un miglioramento della qualità delle acque sotterranee, anche se alcuni parametri nell'acqua di ricarica non sono conformi agli standard impiegati in alcuni paesi europei su cui si concentra questo articolo. L'articolo suggerisce che regolare i processi di ricarica in condizioni controllate per mezzo di CMA a livello nazionale lascia spazio per l'utilizzo di un approccio alternativo, specifico per ogni sito. Potrebbe essere raccomandabile stabilire standard locali a livello regionale o su scala di acquifero per riflettere meglio le diverse caratteristiche di qualità delle acque sotterranee. Secondo l'articolo, le autorità idriche potrebbero ricevere più poteri decisionali nel concedere autorizzazioni per la realizzazione di impianti di ricarica in condizioni controllate basate sulla qualità degli studi idrogeologici e di analisi di rischio per ciascuna richiesta. Questo aiuterebbe a ridurre l'applicazione del principio di precauzione nel concedere l'autorizzazione.

Abstract

Most countries that have technical guidelines or regulations for artificial recharge or managed aquifer recharge (MAR), that include water quality aspects are based on the establishment of standards or Maximum Allowable Concentrations (MACs) to regulate the quality of the water percolated or injected into an aquifer. The number of parameters in these guidelines vary considerably (from 6 in Spain to 156 in the USA) and often apply to all aquifers within administrative boundaries (e.g., national territory), regardless of the nature of the receiving medium, the depth of the water table, and other key factors.

Eleven MAR systems in Spain have been studied (eight operational and three experimental, with limited number of data from three sites), characterising both, the recharge water quality and the water resulting from the interaction processes recharge water-soil-unsaturated zone-saturated zone of the aquifer. In all cases, an improved effect on groundwater quality is observed, even though some parameters in the recharge water don't comply with the standards employed in some European countries, where this article focuses.

The article suggests that regulating water quality for MAR through MACs at national level gives room for another alternative approach specific for each site. It might be recommendable to establish local standards at the regional or aquifer-wide level to better reflect the diversity of groundwater occurrence. As per the article, sectoral water authorities could receive more decision-making power on granting permits for MAR based on the quality of the hydrogeological and risk studies for each request. This would help reduce the application of the precautionary principle when in granting permission.

Introduction

Managed Aquifer Recharge (MAR) englobes different techniques to store water in aquifers. Although MAR has been practiced since centuries ago (ENIP, 2021), in recent years it has gain momentum due to its capacity to adapt to climate change and buffer the impacts from extreme water-related events (e.g., droughts, floods) (Wendt et al., 2020).

Within this context, conducting MAR safely under clear guidelines and regulations is critical to avoid harm to human health, the environment, and socioeconomic assets. Moreover, the principle of "Do No Significant Harm" (DNSH) has been recently incorporated into the European Union (EU) regulations, such as the benchmark regulation (EC, 2021). These requirements usually translate into water quality standards for MAR that apply at the national level. It is remarkable that the Water Framework Directive, WFD (EC, 2000) advocates for water management at the River Basin District or River Basin level, and hence, does not require standards at EU level. In fact, the Groundwater Directive, GWD (2006/118/EC, EC, 2006) allows threshold values to be established at the national or river basin district (RBD) level to reflect different hydrogeological characteristics.

Some countries and organisations have moved forward with normative bodies concerning MAR. The first indirect pragmatic effort has come from the World Health Organization (WHO), which developed a framework to assess the risk to health entailed by MAR (WHO, 2006). In line with this, the European Union (EU) has provided a comprehensive regulatory framework to ensure the good status of surface water and groundwater through the WFD and GWD, among others. More recently, the Joint Research Center (JRC) has formulated a technical guidance document on the minimum water quality requirements for water reuse in agricultural irrigation, which indirectly involves MAR (JRC, 2017). Additionally, several authors and the Common Implementation Strategy (CIS) for the WFD have been working towards a guiding document on MAR for the EU (CIS, 2023, draft).

The Australian Guidelines for Water Recycling (NRMMC; EPHC; NHMRC, 2013) apply a scientific and risk-based approach focusing on identifying and managing risks.

The United States of America implemented the Underground Injection Control Regulations and Safe Drinking Water Act Provisions (USEPA, 1974). In this document, water pollution is prevented by setting maximum allowable concentrations (MACs) for a comprehensive list of contaminants, (USEPA, 2019; Maliva, 2020). Several USA states have enacted their own regulations for MAR, all of them more stringent than the national regulation. Some examples are Arizona (Arizona State Legislature, 1994), California (State of California, 1993;2012), Florida, (State of Florida, 1999), and Washington (Shaleen-Hansen, 2017). The American Association of Civil Engineers (ASCE) proposed one technical guidelines document for MAR (ASCE, 2001) later developed for a climate change scenario (ASCE, 2020).

Other countries have formulated their own MAR-related documents (Fernández et al., 2020; 2022a), either as mandatory regulations, including, Brazil, Chile, Italy, Mexico, South Africa (draft), Spain, The Netherlands (under review), USA, WFD (EU); or guidelines, such as Australia, China (draft), India (draft), Namibia (Regulation proposal), Thailand; or operator rules, as Belgium and Israel; or Technical guidance, as New Zealand.

Out of the 22 MAR-related documents, only 10 consider water quality standards and establish limitations in the form of MACs: WHO, Australia, USA, USA (ASCE), The Netherlands (Minister van Volkshuisvesting, 1993), Italy (MATTM, 2016), Mexico (Conagua, 2007;2009), Spain (the only one with different MACs for percolation (using gravity), and injection (under pressure and requiring electricity MAR systems) (BOE, 2007), India (Dillon et al., 2014), and Europe (JRC, 2017). They are all applicable at the national or at a higher level, regardless of the aquifer's characteristics and the unsaturated zone conditions.

Therefore, there are relatively few regulations on MAR worldwide. Some use maximum allowable concentration (MAC) standards for MAR source water to control pollution (Fernández et al., 2023).

The dichotomy between regulating MAR establishing fixed MACs for the water to be recharged, or other alternatives avoiding assessing limited quality standards parameters is a concurrent topic in the modern MAR state-of-the-art (Fernández et al., 2023). It is also the main stake of this article.

The current analysis explores whether using MACs would be a convenient approach, based on MAR sites in the Spanish territory. To this aim, water quality before and after percolation or injection (injection poses a higher risk, as recharge water is introduced directly into the saturated zone, impeding the important purification role of the unsaturated zone during percolation) is analysed and compared to MACs regulated in Italy, Spain and The Netherlands, the three European countries that have regulated MACs for MAR systems (listed in alphabetical order).

Consequently, the analysis focuses on the exploration of MAR water quality at MAR sites in Spain, the comparison between several quality parameters in these sites and some MAC-based water quality control standards, and additionally, other factors are compared, including the receptors of the water´s characteristics.

Thus, this analysis aims to determine if Spanish MAR sites would comply with existing regulations, and, accordingly, whether a MAC approach might satisfactorily prevent groundwater contamination, and simultaneously, allow MAR expansion.

Material and methods

Case studies description

A total of eleven Spanish MAR sites have been chosen and described based on the availability of chemical analysis data for both, recharge water and groundwater resulting from interaction in the receiving aquifer. However, some of this data is not publicly available and requires a request to the owners of the information. Unfortunately, not all owners have allowed the publication of the chemical analysis tables.

The location and main characteristics of these spots are summarised in Figure 1 and Table 1. These sites involved a wide variety of geographical contexts and use some of the different methods to conduct MAR, including water spreading, Aquifer Storage (AS), Aquifer Storage and Recovery (ASR), and Soil Aquifer Treatment (SAT) - MAR.

The following is a description of each site:

The Careos canals (Granada-Almeria)

The "acequias de careo" or careo channels are MAR canals of permeable bottom and water-spreading systems used in Sierra Nevada, Southern Spain, to increase water availability since the early Al-Andalus (8-10th century AD). They consist of dug canals at the headwater of the basins that collect, transport,

Fig. 1 - Evaluated MAR sites in Spain.

Fig. 1 - Siti MAR esaminati in Spagna.

Tab. 1 - Characteristics of the Spanish MAR sites evaluated (location, water source used for MAR, objective, MAR type, and hydrochemical facies for both, the recharge water (MAR), and groundwater (GW).

Tab. 1 - Caratteristiche dei siti MAR spagnoli valutati (ubicazione, fonte idrica utilizzata per la ricarica, obiettivo, tipo ricarica (MAR) e facies idrochimica per le acque di ricarica (MAR) e le acque sotterranee (GW).

MAR site	Location (province, region)	Source water type	Objective	MAR type	Hydrochemical facies
Careos	Granada and Almeria (Andalusia)	Snowmelt	Improve water supply for agriculture and villages during the dry season	Water spreading	Ca-SO_4 and Ca-HCO_3 (MAR and GW)
Cobre de las Cruces Copper Mine	Seville (Andalusia)	Treated wastewater	Drain the open hole mine and reinjection nearby	Aquifer storage	Na-Cl (MAR)
Guadiana MAR site	Ciudad Real (Castile La Mancha)	River water	Irrigation and recover of environmental assets	Aquifer storage	Ca-Na-CO_3 (MAR) Na-CO_3 and Na-HCO_3 (GW)
Canal Isabel II ASR site	Madrid (Madrid)	Treated water	Strategically store water in case of need	Aquifer storage, (transfer), & recovery [AS(T)R]	Ca-HCO_3 (MAR) Ca-HCO_3 to Na-HCO_3 (GW)
Los Arenales MAR sites 1-Santiuste 2-El Carracillo 3-Pedrajas-Alcazarén	Segovia and Valladolid (Castile and Leon)	River water (1,2), and runoff, river and WWTP (3)	Reverse groundwater depletion and sustain irrigation	Water spreading	Ca-HCO_3 (MAR and GW, a & b). Ca-SO_4 (MAR and GW, c)
Arabayona MAR site	Arabayona (Castile and Leon)	Drainage	Evacuation of flooding water to a neighbour aquifer	Rainwater harvesting	Ca-HCO_3 (MAR and GW)
Zorrilla urban water buffer	Valladolid (Castile and Leon)	Rainwater	Experimental site to use urban runoff for irrigation of the stadium's green	Aquifer storage and recovery (ASR)	Ca-HCO_3 (MAR) Mg-Ca-HCO_3 (GW)
Sant Vicenç dels Horts MAR site	Sant Vicenç dels Horts, Barcelona (Catalonia)	Treated wastewater	Increase the irrigation guarantee	Soil aquifer treatment (SAT)	Ca-HCO_3 (MAR and GW)
Port de La Selva SAT-MAR site	El Port de la Selva, Girona (Catalonia)	Treated wastewater	Water reuse to cover peak demands	Soil aquifer treatment (SAT-MAR)	Na-Ca-HCO_3-SO_4 (MAR) Ca-Na-HCO_3 (GW) CECs
Majorca experimental SAT-MAR	Majorca (Balearic Islands)	Treated wastewater	Irrigation and strategic reservoir for peak demands	Water spreading with reclaimed water (SAT-MAR)	Na-Cl and Na-Cl-HCO_3 (MAR) Ca-Cl, Ca-SO_4, Na-Cl, and Na-HCO_3 (GW)
Tenerife SAT-MAR pilot	Tenerife (Canary Islands)	Treated wastewater	Incipient experimental site to diversify water storage systems	Recharge with reclaimed water (SAT-MAR)	Mg-Na-Cl-HCO_3 (MAR) Na-Cl-HCO_3 (GW)

and enhance snowmelt and runoff recharge into the underlying fractured aquifer (Figs. 2, 1 a-b). This recharge occurs predominantly in spring and increases groundwater discharge into the lowlands during the dry season. The total volume infiltrated is up to 3.5 Mm^3/year (Fernández et al., 2005a).

The role of the Careos in the hydrological cycle of the Bérchules River Watershed extends over an area of about 68 km^2 on the southern edge of Sierra Nevada (Barberá et al., 2018). The authors integrated different approaches and focused on analysing the hydrochemistry of major ions, chemical components, and water isotopes. Water samples were collected from wells, springs, and surface water points in two campaigns during the snowy (January-February) and the snowmelt (May-June) seasons of 2015. Snow samples were analysed in April 2015 (Barberá et al., 2018; Jódar et al., 2022).

Overall, water in Bérchules has calcium-bicarbonate and calcium-magnesium-bicarbonate type facies, with fewer occurrences of calcium-magnesium-bicarbonate-sulphate and sodium-calcium-bicarbonate types. Groundwater electric conductivities range between 19 and 1,188 µS/cm with an average of 111 µS/cm, and shows low mineralisation level in the uplands (≤ 36 µS/cm). The type of existing aquifers is fissured, with schists, shales and slates as main lithologies. The mineralisation of groundwater is due to two main processes: the concentration of solutes such as Na, Ca, Cl, and SO_4, due to evaporation and chemical reactions between the recharged water and the porous medium, namely hydrolysis (e.g., albite, anorthite, and K-feldspar) and dissolution (calcite and dolomite). There is also some input of CO_2 to water from biogenic sources in the soil and the atmosphere. The study of temperature gradients, isotopes, and conservative chloride concentrations led to the conclusion that nearly 78% of basin discharge corresponds to groundwater and that 21% of annual precipitation results in recharge (Barberá et al., 2018). MAR in these areas has considerably increased recharge, since the characteristic steep slopes, and low-permeable lithologies could not account for the high percentage of precipitation converted into groundwater (Barberá et al., 2018; Jódar et al., 2022).

Snowmelt and groundwater are predominantly Ca-SO_4 and Ca-HCO_3 facies (Fig. 4: 1), although some particular springs and wells have different hydrofacies.

The source water for MAR in this MAR scheme is high-quality and does not require any barrier to reduce the risk of water pollution. Also, the referenced literature doesn't mention any geogenic contaminant that could be mobilised by MAR and poses a risk for later human use or the environment.

The Cobre las Cruces copper mine (Seville)

The Cobre de Las Cruces Copper Mine is an open pit mine in South-western Spain (Figs. 2, 2 a-b). The mine has intersected the Niebla-Posadas granular aquifer, integrated by conglomerates, gravel, sand and sandstone with interbedded clay layers of Miocene age, and therefore, requires a complex drainage and re-injection systems to dewater a large area. The drainage system comprises 32 active extraction wells. Before the re-injecting of the abstracted water by means of 28 injection wells in an outer ring, water is treated through reverse osmosis to remove metals and other water constituents. The drainage and re-injection system transports an annual volume of around 3.2 Mm^3 (Baquero et al., 2016). The re-injected volume is about 1.85 Mm^3/year.

At the mine site, the aquifer is confined by a marl layer whose thickness varies between 120-150 m. The native groundwater is almost not renewable and is a mix of two end-members, one of which is highly saline cognate water that probably remained since the transgression of the Tortonian Sea. Groundwater quality varies spatially. As it travels from the recharge zone in the northern fringe of the aquifers to the south, the concentration of As, NH_4, and B increases through natural processes, predominantly that involve organic matter, minerals in the porous medium, and mixing of waters. In some parts of the aquifer, some constituents' concentration exceeds drinking and irrigation water quality.

In the recharge zones, nitrate concentration and sulphate are high due to agricultural activities and environmental enrichment, respectively. Nitrate concentrations decrease as groundwater travels southward and disappears once the aquifer becomes confined. The re-injected water must be treated to comply with the regional water authority's requirements, due to certain pollutants of natural origin being above desired levels. This treatment takes place in a waste water treatment plant (WWTP). The resulting water also loses calcium and magnesium (Baquero et al., 2016). Regarding hydrofacies, the native groundwater and the treated groundwater belong to the Na-Cl type (Fig. 4: 2).

The Guadiana MAR canal (Ciudad Real)

This MAR site utilises intermittent river water surpluses to recharge it into a karstic unconfined aquifer. It consists of a series of wells placed on the river bank of the Guadiana Canal that capture river water during high stages (especially in winter) and percolate it (Figs. 2, 3 a-b) into a karstic and granular aquifer, comprising tertiary limestone and detrital plio-quaternary volcanic sediments. The aquifer is heterogeneous, with permeabilities that range between 50 and 20,000 m/day. Water tables are reached at a depth between 50 and 30 m. The final use of the water stored through MAR is irrigation demands along the Guadiana Canal, and the restoration of degraded wetlands in the Daimiel National Park (Fernández, 2015).

Groundwater in the area can be of poor quality in some wells, especially regarding to nitrate and nitrite concentration, likely as a result of agriculture in the region, and the presence of reduction environments due to shale layers, respectively. The main water quality issue in this site is the presence of nitrites above the Spanish regulation for MAR, i.e. Royal Decree 1620/2007 (BOE, 2007), mainly due to the MAR water collecting method (drainage of the mine). Nonetheless, the quality of this water is often better than the native groundwater, which implies that MAR helps dilute pollutants (Fernández, 2015).

Water source hydrofacies correspond to Ca-Na-CO_3 while groundwater's to Na-CO_3, predominantly, and Na-HCO_3 in at least one well (Figs. 4, 3).

The Canal de Isabel II ASR sites (Madrid)

Canal de Isabel II (CYII) is the Madrid region's main water supply and wastewater treatment organisation. It relies predominantly on dam storage to meet water demand and can extract up to 70 Mm^3 of groundwater from the multi-layer Tertiary Detrital aquifer of Madrid (TDAM) in case of emergency (e.g., prolonged drought). In this context, CYII has conducted aquifer storage and recovery (ASR) tests at three sites to replenish groundwater storage in the TDAM (Nogueras et al., 2019), namely, Casilla Valverde Bis, La Cabaña Bis, and FE-1R (Figs. 2, 4 a-b).

The wells for extraction reach depths around 700 m, while the MAR recharge infrastructure does not exceed 400 m. MAR trials have been conducted to assess the impact of ASR on water quality, quantity, and some design criteria for optimal performance (Nogueras et al., 2019; Sánchez and Gutiérrez, 2019).

The injected water meets the Spanish criteria for drinking water set forth by Royal Decree 140/2003 (BOE, 2003) for drinking water standards. Groundwater levels rise by about 8-10 m during recharge. The conductivity of groundwater (300 µS/cm) drops due to mixing with the recharged water (90-100 µS/cm). Trihalomethanes (THMs) can be found in the aquifer due to the injection of chlorinated drinking water, and show potential as a tracer to determine the distribution of MAR water in the aquifer (Table 2). However, significant changes in the quality of the native groundwater were not found due to the implementation of the ASR system. The recovered water meets regulated MACs (Table 2) (Nogueras et al., 2019; Sánchez and Gutiérrez, 2019). It is worth to mention that the produced THMs pose a potential risk, since some THMs are not regulated in the MACs presented in Annex 2 (e.g., cloroform and bromoform).

The water quality of the TDAM shows spatial variation. It changes with depth from Ca-HCO_3 to Na-HCO_3 hydrofacies (Fig. 3, 4).

The Los Arenales MAR sites (Segovia and Valladolid)

The Los Arenales MAR sites consists of three large-scale systems that replenish an intensively exploited aquifer, namely, Los Arenales aquifer. They are located on the Spanish side of the Duero River basin, Central Spain. They are distributed in three main regions: Santiuste, El Carracillo, and Pedrajas-Alcazarén.

These constructions were a response to the considerable decline in groundwater levels experienced in the southern region of the Duero River basin, due to massive groundwater abstractions for irrigation. They also seek to ensure irrigation demands in the context of over-allocated water resources.

These MAR systems rely on a combination of infiltration basins, infiltration canals, artificial wetlands, and wells to recharge an unconfined quaternary sandy aquifer that has, in some parts, direct connection with layers of the deep tertiary granular semiconfined and confined aquifer of the Central Duero basin (Fernández and López-Gunn, 2021).

All these systems share "MAR-based" solutions to address aquifer-intensive use that are characterised by five common features: i) passive systems that do not require electricity for MAR activity, relying instead on gravity); ii) intermittent recharge, i.e. it takes place when there is high flow in the rivers from which water is diverted; iii) regulated MAR system that is integrated into the whole IWRM scheme; iv) legally regulated through temporal water permits with specific characteristics, and v) integrated, with the interconnection of all the water management options of surface and groundwater origin. The Los Arenales MAR Sites have yielded an average recharge of about 4.8 Mm^3/year between 2002 and 2020 (Fernández and López-Gunn, 2021).

a- Santiuste basin

The components of the MAR system are about 27 km of MAR canals, five infiltration ponds, three artificial wetlands, an inverse riverbank filtration (RBF) system, and three high-diameter infiltration wells (Fernández 2005b; Fernández and López-Gunn, 2021).

The water source is taken from the Voltoya River's surpluses, which are granted only when river stages are above

Tab. 2 - Water quality during the main stages of the ASR trail schemes. Taken from Nogueras et al. (2019). All of them met the Royal Decree 140/2003 about water quality thresholds. The distance between the pumps from the injection (and eventually extraction) to the extraction (exclusively) downstream borehole is about 500 m.

Tab. 2 - Qualità dell'acqua durante le fasi principali dei processi ASR. Tratto da Nogueras et al. (2019). Tutti rispettavano il Regio Decreto 140/2003 sui limiti di qualità dell'acqua. La distanza tra le pompe del pozzo di iniezione (ed eventualmente estrazione) al pozzo di estrazione (esclusivamente) a valle è di circa 500 m.

	Casilla Valverde Bis			FE-1 R		
Parameter	Before MAR	Recovery water from the recharge borehole	Water extracted in the down-flow extraction borehole	Before MAR	Recovery water from the recharge borehole	Water extracted in the down-flow extraction borehole
Conductivity (µS/cm)	219	149	317	260	220	431
pH	7.8	8.78	8	7.49	7.26	8.15
As (µg/L)	7.5	< 2.5	17	7.2	< 2.5	36.1
THMs (µg/L)	0	35	1.7	0	11.9	2.1
Nitrate (mg/L)	2.8	0.3	3.4	4.8	5.6	2.6

Fig. 2 - Pictures of the characterised MAR sites in Spain (part 1).
1. Careo canals in the Bérchules River watershed, Sierra Nevada (a, b). 2. The Cobre Las Cruces copper mine, general view (a), and re-injection well (b). 3. The Guadiana MAR site: Peñarroya dam heading the MAR canal (a), and MAR well used to recharge river water into the karstic mudstone aquifer (b). 4. The Canal de Isabel II FE-1R ASTR Sites (Madrid). FE-1R borehole for injection (a), and Fuencarral water supply and MAR system scheme (b). 5. Los Arenales MAR sites: 5.1 Santiuste basin MAR site. Valve at the heading of the MAR channel (a), and infiltration-stagnation basin in Santiuste (b). 5.2 El Carracillo MAR site. La Laguna del Señor, an infiltration basin in the MAR system heading (a), and infiltration basin in Gomezserracín (b). 5.3 Pedrajas-Alcazarén SAT-MAR. Valve 3 of the SAT-MAR conduction system (a), and infiltration channel (b).
** Photos of the first author, except 2A (courtesy of CLC), and 3B (courtesy of Tragsa).*

Fig. 2 - Immagini dei siti MAR caratterizzati in Spagna (parte 1).
Canali di Careo nel bacino del fiume Bérchules, Sierra Nevada (a, b). 2. La miniera di rame Cobre Las Cruces, vista generale (a), e pozzo di reiniezione (b). 3. Il sito MAR di Guadiana: diga di Peñarroya all'inizio del canale MAR (a), e pozzo MAR utilizzato per ricaricare l'acqua del fiume nell'acquifero carsico (b). 4. I siti FE-1R del Canal de Isabel II ASTR (Madrid). Pozzo FE-1R per iniezione (a), e schema del sistema di approvvigionamento idrico e MAR di Fuencarral (b). 5. Siti MAR di Los Arenales: 5.1 Sito MAR del bacino di Santiuste. Valvola all'inizio del canale MAR (a), e bacino di infiltrazione-decantazione a Santiuste (b). 5.2 Sito MAR di El Carracillo. La Laguna del Señor, un bacino di infiltrazione all'inizio del sistema MAR (a), e bacino di infiltrazione a Gomezserracín (b). 5.3 SAT-MAR di Pedrajas-Alcazarén. Valvola 3 del sistema di conduzione SAT-MAR (a), e canale di infiltrazione (b).
Foto del primo autore, tranne 2A (cortesia di CLC), e 3B (cortesia di Tragsa).

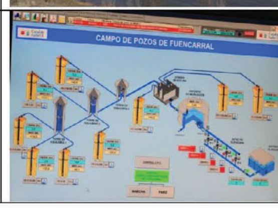
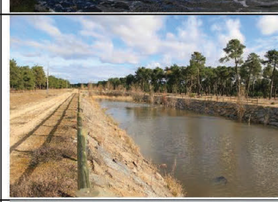
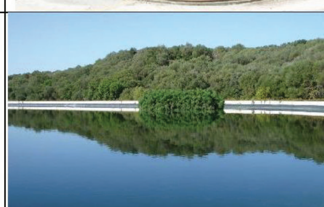
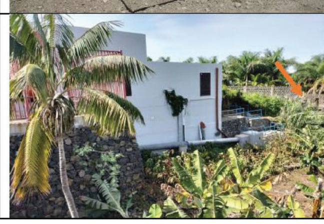
Nº	MAR SITE	a	b
1	**The Careos canals (Granada-Almeria)**		
2	**The Cobre de Las Cruces copper mine (Seville)**		
3	**The Guadiana MAR canal (Ciudad Real)**		
4	**The Canal de Isabel II, FE-1R ASR, Fuencarral site (Madrid)**		
5	**LOS ARENALES**		
5.1	**Santiuste basin (Segovia)**		
5.2	**El Carracillo MAR system (Segovia and Valladolid)**		
5.3	**Pedrajas-Alcazarén SAT-MAR system (Valladolid)**		

Fig. 3 - Pictures of the characterised MAR sites in Spain (part 2).
6. The Arabayona MAR site. Example of water logging issues experienced in the area due to poor drainage, where water is conducted to perimeter irrigation canals (a), and MAR-canal built to infiltrate drained water into a neighbour aquifer (b). Modified from Fernández and Paredes (2022b). 7. The urban water buffer Zorrilla (Valladolid), taken from https://www.fieldfactors.com (a), and piezometer construction in the parking lot (b). 8. The Sant Vicenç dels Horst MAR site. Location and images of the diversion from the Llobregat River (a), and infiltration pond (b). 9. The Port de La Selva SAT-MAR Site, WWTP secondary treatment (a), and tertiary unit (b). https://demeau-fp7.eu/sites/files/SVH.png
10. Majorca experimental SAT-MAR site. Pond to store water from the Maria de Salut wastewater treatment plant (a), irrigation system at one of the plots studied where MAR tests were conducted by over-irrigation, and position of the piezometer downwards (b) marked with an orange arrow. 11. Tenerife experimental SAT-MAR site. WWTP (EDRAR) Noreste (a), and MAR site using a percolation well marked with an orange arrow (b).
Photos of the first author, except 7 A (courtesy of Field Factors), and 9 A-B (courtesy of Amphos 21).

Fig. 3 - Immagini dei siti MAR caratterizzati in Spagna (parte 2).
6. Il sito MAR di Arabayona. Esempio di problemi di ristagno d'acqua sperimentati nella zona a causa di un cattivo drenaggio, dove l'acqua viene condotta nei canali per l'irrigazione perimetrale (a), e canale MAR costruito per infiltrare l'acqua drenata in un acquifero vicino (b). Modificato da Fernández e Paredes (2022b). 7. Il serbatoio urbano Zorrilla (Valladolid), tratto da https://www.fieldfactors.com (a), e costruzione del piezometro nel parcheggio (b). 8. Il sito MAR di Sant Vicenç dels Horst. Posizione e immagini della derivazione dal fiume Llobregat (a), e laghetto di infiltrazione (b). 9. Il sito SAT-MAR di Port de La Selva, trattamento secondario delle acque reflue (a), e unità terziaria (b). https://demeau-fp7.eu/sites/files/SVH.png
Sito sperimentale SAT-MAR di Maiorca. Stagno per immagazzinare l'acqua proveniente dalla centrale di trattamento delle acque reflue di Maria de Salut (a), sistema di irrigazione in una delle parcelle studiate dove sono stati condotti test MAR tramite sovra-irrigazione, e posizione del piezometro verso il basso (b) contrassegnato con una freccia arancione. 11. Sito sperimentale SAT-MAR di Tenerife. Centrale di trattamento delle acque reflue (EDRAR) Noreste (a), e sito MAR che utilizza un pozzo di percolazione contrassegnato con una freccia arancione (b).
Foto del primo autore, tranne 7 A (cortesia di Field Factors), e 9 A-B (cortesia di Amphos 21).

Nº	MAR site	a	b
6	**The Arabayona MAR site (Salamanca)**		
7	**The Zorrilla urban water buffer (Valladolid)**		
8	**The Sant Vicenç dels Horst MAR site (Barcelona)**		
9	**The Port de La Selva SAT-MAR site (Girona)**		
10	**Majorca Island. Experimental SAT-MAR site (Balearic Islands)**		
11	**The Tenerife SAT-MAR pilot (Canary Islands)**		

a minimum ecological flow (about 1,000 L/s). The recharge period varies depending on precipitations, comprising some rainy winter months, usually from December 1st to April 30th.

Water is usually good quality because Voltoya River proceeds from the granitic mountains in the Central Massif. The water has been analysed since 2001, with plenty of piezometers integrated into a monitoring network (Fernández, 2005b) (Figs. 2, 5.1 a-b).

b- El Carracillo MAR system

The system integrates a fish-bone pipeline network as a 19.2 km aqueduct from the Cega River (Salto de Abajo site) to 14 distribution points, either in infiltration ponds or to the heads of MAR canals. Several MAR techniques are used, including 16 infiltration ponds, 17 km of MAR canals, two spreading basins, and three artificial wetlands. The scheme includes reused abandoned wells and sand pits.

The system manages relatively high quality since the water intake is located at a relatively high altitude in plutonic bedrock before water use could threaten water quality integrity. The system also counts on a piezometer monitoring network (Fernández and López-Gunn, 2021; Fernández and San Sebastián, 2021) (Figs. 2, 5.2 a-b).

c- Pedrajas-Alcazarén SAT-MAR system

The Pedrajas-Alcazarén MAR system is novel concerning previous experiences in water intake diversification, originating from 3 different sources: a river diversion from Pirón River, a WWTP with advanced secondary treatment, and a ditch to convey runoff from the village rooftops to a connection point where the MAR canal starts.

The components of the system are the SAT-MAR or a combination of a WWTP and a MAR system, a 2 km long pipeline, 5.5 km of infiltration canals, an RBF system, and two infiltration ponds (Fernández and López-Gunn, 2021) (Fig. 2, 5.3 a-b).

The main concern related to this water source is total organic carbon (TOC) concentration above most of the standard limits reported in 2012, at the beginning of the operations (Fernández et al., 2016). It was solved improving the WWTP efficiency. The second eventual damage is the appearance of contaminants of emerging concern (CECs).

In Santiuste and El Carracillo sites, the predominant water facies are Ca-HCO_3, while in Pedrajas-Alcazarén SAT-MAR, the main water type is Ca-SO_4 (Figs. 3, 5) (Fernández et al., 2016).

The Arabayona MAR site (Salamanca)

The Arabayona MAR site drains water from a flooded area (dewatered former wetlands) to a MAR channel placed beyond a threshold, where water infiltrates an underlying tertiary unconfined aquifer comprising conglomerates, sandstone, silt, and mud.

Arabayona irrigation district includes an area with topographic and environmental conditions that favour inundation, mainly due to precipitation, high groundwater tables, and irrigation, which frequently results in crop damage. To deal with this issue, drained water passes through secondary sewer systems, that also work as runoff traps, on the way to an infiltration channel (Fernández and Paredes, 2022b) (Fig. 3, 6 a-b).

Water is generally of good quality (Fig. 4, 6). The main exception is the high concentration of nitrates due to agriculture. Water hydrofacies is Ca-HCO_3 for all, drained, MAR, and groundwater.

The Zorrilla urban water buffer (Valladolid)

This pilot MAR scheme is located at the José Zorrilla football Stadium in Valladolid. Initially conducted some experiments collecting rainwater from the parking area, which is infiltrated underground for later recovery (ASR system) and reused as a source of irrigation for the football court. A gutter system collects and directs the water to a storage tank. Subsequently, water is conveyed to a biofilter with vegetation that improves water quality before injection underground. Finally, when required, water is pumped and used to irrigate the stadium. This scheme can meet up to 20% of the stadium irrigation needs (Versteeg et al., 2021) (Fig. 3, 7 a-b).

Runoff water in this site is Ca-HCO_3 type, and groundwater belongs to Mg-Ca-HCO_3 hydrofacies (Fig. 4, 7). The main water quality concern is alkalinity for irrigation of the green, and CECs presence, especially HCs.

The activity is still expecting the final permission from water authorities.

The Sant Vicenç dels Horst MAR site (Barcelona)

This MAR site is located in the Llobregat area and the vicinity of the municipality of Sant Vicenç dels Horst, Catalunya. Water from the Llobregat River is conducted to a decantation pond with an area of about 5,600 m^2. Subsequently, the water is taken to an infiltration pond (4,000 m^2), where water percolates into an unconfined aquifer a few meters thick (and up to 10 m). The main purpose of this MAR system is to increase groundwater storage at the local scale. Yearly recharge volumes are in the order of 1.2 Mm^3/y (Fajnorová et al., 2021) (Fig. 3, 8 a-b).

In 2011, the infiltration pond was upgraded with an organic layer of vegetal composts to enhance the removal of certain water constituents, through processes such as adsorption and degradation.

In this site, MAR water source and groundwater have a very similar proportion of major ions and the predominant hydrofacies is Ca-HCO_3 (Fig. 4, 8).

The Port de La Selva SAT-MAR site (Girona)

The town of El Port de La Selva WWTP uses a Soil Aquifer Treatment (SAT) scheme to improve the quality of reclaimed water for urban water supply. The scheme involves recharging tertiary-treated wastewater into granular unconfined aquifer 13–14 m thick composed of poorly sorted and poorly rounded metamorphic rocks in gravel and block size, embedded in a matrix of sand and silt, through three infiltration basins (Fig. 3, 9b). The hydraulic conductivies ranges between 4 and 600 m/d. The distance between the closest infiltration pond and piezometers 2 and 4 is about 30 and 80 m downstream, respectively (Fajnorová et al., 2021). The SAT-MAR system was modelled using the code FEFLOW.

In winter, primary effluent is treated to reduce total nitrogen below 10 mg/l before it is directed to the tertiary treatment plant comprising a dual media filter (granular activated carbon filter and UV disinfection systems). The final effluent is conveyed to the infiltration basins where managed aquifer recharge (MAR) takes placeOperations started in 2015. The intentional recharge uses three infiltration basins with a combined area of 439 m^2, which operate following wet and dry cycles. Changes in groundwater quality are monitored through a small network of three piezometers located down-gradient of the infiltration sites. The aquifer consists of gravel deposits embedded in a sand and silt matrix, about 13-14 m thick (Amphos 21, 2016; Fajnorová et al., 2021).

The SAT-MAR system in Port de La Selva combined with attenuation processes in the aquifer also helps to reduce the concentration of several water constituents in the recharge water after traveling through the aquifer, e.g., dissolved organic carbon, chloride, sulphate, and dissolved oxygen (Fig. 4, 9 and Table 3). Although SAT is not really a safe MAR practice, and definitely against WFD provisions, the conditions of the ground in this area have probed to reduce bacteria, enteric viruses, and phage presence (Table 3).

Water quality is a major concern in this area due to the presence of microorganisms, antibiotics, and contaminants of emerging concern. As per the previous study, 15 contaminants of emerging concern (CECs) were detected in groundwater above the safe levels for health and drinking purposes. However, efforts are being made to improve the purification capacity of the wastewater treatment plant in order to address these concerns. The effluent used for MAR is Na-Ca-HCO_3-SO_4 type, while groundwater from two piezometers belongs to Ca-Na-HCO_3 hydrofacies (Fig. 4, 9). Some cations have been assessed using a hydrochemical calculator.

Majorca. Experimental SAT-MAR site (Balearic Islands)

In this site, an experiment was undertaken, consisting of over-irrigating crops to recharge an underlying granular aquifer fossilizing another karstic mudstone aquifer, via irrigation returns. The agricultural area in which this experiment took place is distributed between the municipalities of Maria de la Salut, Sineu, and Ariany. It is limited to the east by the road from Petra to Santa Margalida (Ma-3340), and to the south by the Ma-3301 road. This site consists of small or very small plots of land (EARSAC, 2019).

The irrigation system uses private wells that pump water at a corner of the plots and distribute it by gravity. The total area is 160 ha. The crops grown are mainly fodder crops, cereals, almonds, vegetables, and, to a lesser extent, some fruits and citrus fruits.

Between 2013 and 2018, the experiment took place employing “stimulated recharge” by applying a dose of irrigation above the crop’s necessities with reclaimed water proceeding from a Maria de la Salut wastewater treatment plant and storage pond (Fig. 3, 10 a), recharging the site by over-irrigation (Fig. 3, 10 b). The excess water reaching the aquifer and the interactions in the saturated and unsaturated zone were analysed through a well located at a lower hydraulic level used as a piezometer about 50 m far, and more distant wells.

Sequential analyses were conducted over five years, enabling the study of the interaction processes between reclaimed water and the receiving medium and water crops. They demonstrated that the system began to function in a permanent regime after five years of irrigation, in terms of both, groundwater quantity and quality (EARSAC, 2019).

The treated wastewater corresponds to Na-Cl and Na-Cl-HCO_3 hydrofacies, while the native groundwater varies considerably, showing Ca-Cl, Ca-SO_4, Na-Cl, and Na-HCO_3 in the closest piezometer (Fig. 4, 10).

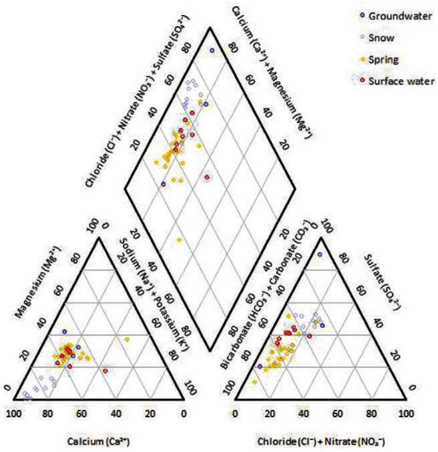
Tab. 3 - Port de la Selva. Summary of laboratory analyses results. Modified from Amphos 21, 2016.

Tab. 3 - Porto della Selva. Riepilogo dei risultati delle analisi di laboratorio. Modificato da Amphos 21, 2016.

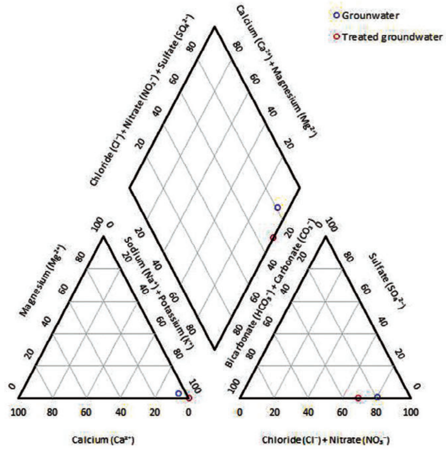
Parameter	Reclaimed water 14/11/2016	Reclaimed water 14/12/2016	Piezometer 24/10/2016	Piezometer 4 14/11/2016)
TOC, mg/L	7.6	2.1	< 2.0	< 2.0
Total plaguicides organhalogenates	< 0.010 µg/L	< 0.010 µg/L	< 0.010 µg/L	< 0.010 µg/L
Total plaguicides organophosphates	< 0.010 µg/L	< 0.010 µg/L	< 0.010 µg/L	< 0.010 µg/L
Triazines	Total< 0.010 µg/L Atraton 0.010 µg/L (0.1 µg/L) Terbutryn 0.037 µg/L (0.1 µg/L)	Total< 0.010 µg/L	Total< 0.010 µg/L	Total< 0.010 µg/L
Total HC	< 0.010 µg/L Napthalene 0.022 µg/L (-)	< 0.010 µg/L	< 0.010 µg/L Napthalene 0.020 µg/L (-)	< 0.010 µg/L
Volatile organic compounds	Sum THMs< 1.5 µg/L (100 µg/L)	Sum THMs< 1.5 µg/L (100 µg/L)	Sum THMs< 1.5 µg/L (100 µg/L)	Sum THMs< 1.5 µg/L (100 µg/L)

Fig. 4 - Piper-Hill-Langelier hydrograms (Piper, 1944) for water analysis of the MAR sites. The legends indicate whether a point corresponds to MAR or source water (before MAR), or groundwater (after MAR). All hydrograms are own elaboration using data either from references, or borrowed by data owners expressed in Table 4.

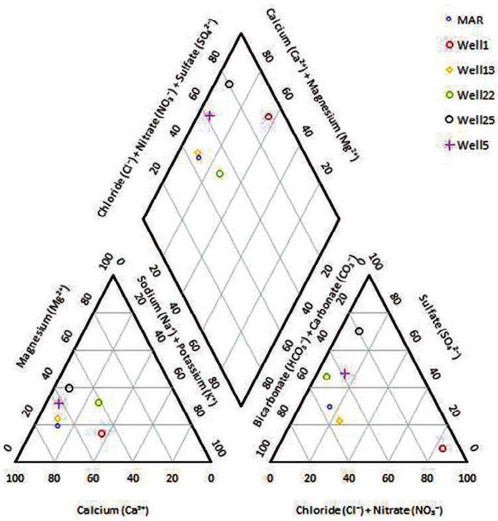
Fig. 4 - Idrogrammi di Piper-Hill-Langelier (Piper, 1944) per l'analisi dell'acqua dei siti MAR. Le legende indicano se un punto corrisponde a acqua MAR o acqua sorgiva (prima del MAR), o acqua sotterranea (dopo l'impianto MAR). Tutti gli idrogrammi sono elaborati utilizzando dati provenienti sia dalla bibliografia, sia concessi dai proprietari dei dati espressi nella Tabella 4.



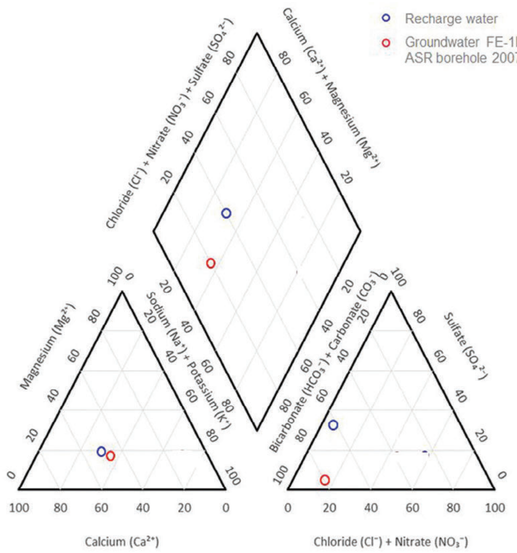
N°1 - MAR Location: The Careos canals (Granada-Almeria).



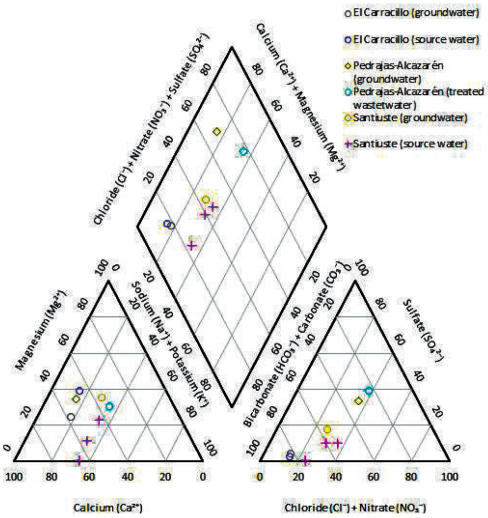
N°2 - MAR Location: The Cobre las Cruces copper mine (Seville).



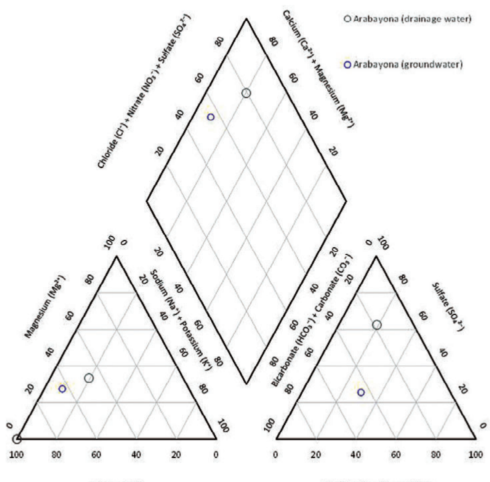
N°3 - MAR Location: The Guadiana MAR canal (Ciudad Real).



N°4 - MAR Location: The Canal Isabel II Fuencarral ASR site (Madrid).



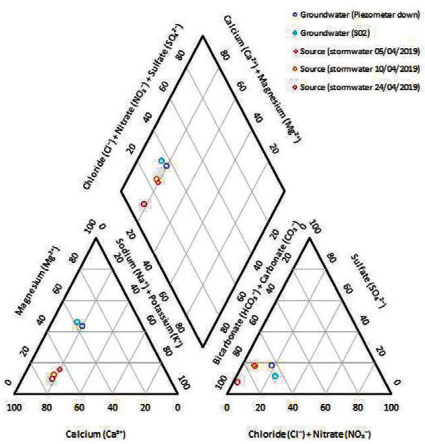
N°5 - MAR Location: The Los Arenales MAR sites:
a-Santiuste basin (Segovia)
b-El Carracillo (Segovia and Valladolid)
c-Pedrajas-Alcazarén (Valladolid)



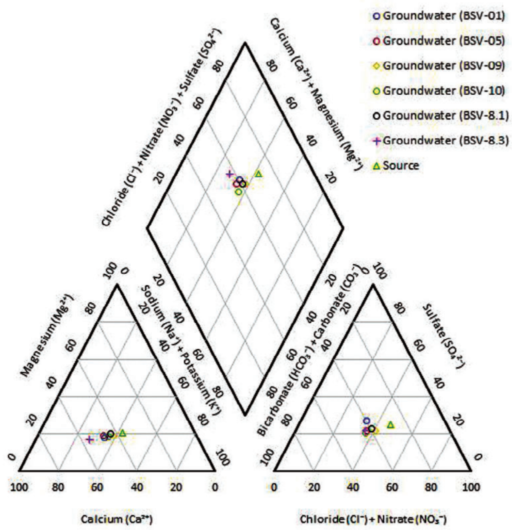
N°6 - MAR Location: The Arabayona MAR site (Salamanca)

Fig. 4 - Piper-Hill-Langelier hydrograms (Piper, 1944) for water analysis of the MAR sites. The legends indicate whether a point corresponds to MAR or source water (before MAR), or groundwater (after MAR). All hydrograms are own elaboration using data either from references, or borrowed by data owners expressed in Table 4.

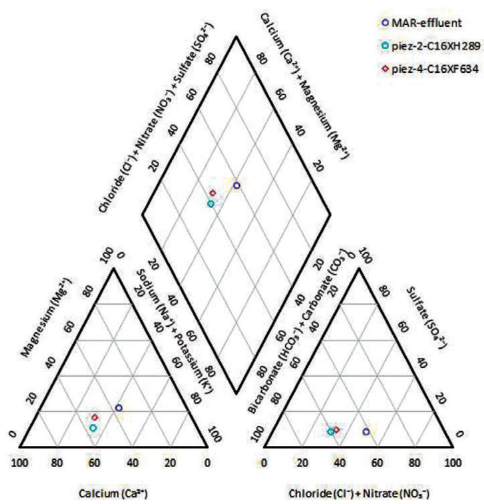
Fig. 4 - Idrogrammi di Piper-Hill-Langelier (Piper, 1944) per l'analisi dell'acqua dei siti MAR. Le legende indicano se un punto corrisponde a acqua MAR o acqua sorgiva (prima del MAR), o acqua sotterranea (dopo l'impianto MAR). Tutti gli idrogrammi sono elaborati utilizzando dati provenienti sia dalla bibliografia, sia concessi dai proprietari dei dati espressi nella Tabella 4.



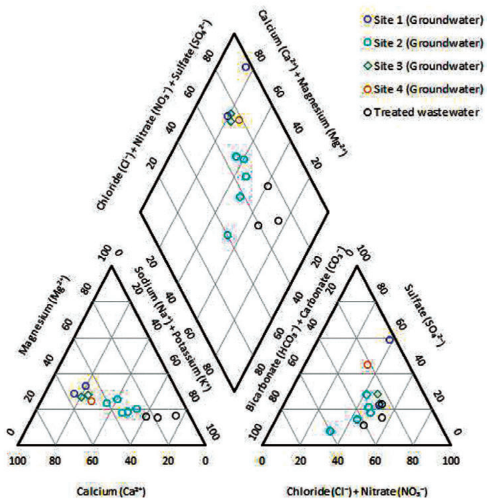
N°7 - MAR Location: The Zorrilla urban water buffer (Valladolid).



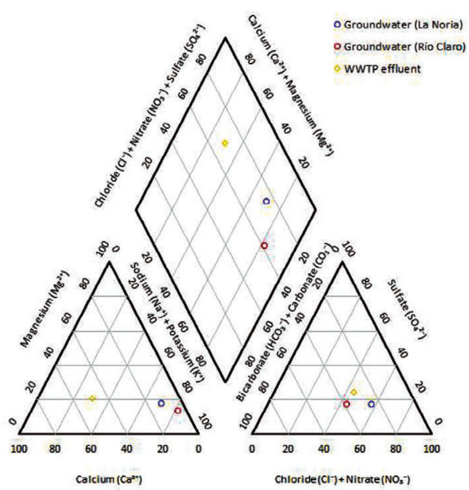
N°8 - MAR Location: The Sant Vicenç dels Horst MAR site (Barcelona).



N°9 - MAR Location: The Port de La Selva SAT-MAR site (Girona).



N°10 - MAR Location: The Majorca experimental SAT-MAR site (Balearic Islands).



N°11 - MAR Location: The Tenerife SAT-MAR pilot (Canary Islands).

The Tenerife SAT-MAR pilot (Canary Islands)

A new experiment on Tenerife Island conducted by the Consejo Insular de Aguas de Tenerife (CIATF) is currently taking place, consisting of the surplus of the wastewater treatment plant Valle de Guerra (Northeast of the Island) (Fig. 3, 11 a), to be percolated into fractured basalts formations through a dug-well (Fig. 3, 11 b). The objective is to study the behaviour of the receiving medium and the interaction processes between reclaimed water and the basaltic aquifer. Also, this site aims to advance the knowledge of groundwater movement through volcanic fractured aquifers, which behave as a heterogeneous and anisotropic medium.

The water is percolated through a 20 m deep, and 1.20 m diameter dug well. The project will run for at least one year. The water quality evolution will be tested in two exploitation wells located downwards according to the groundwater flow gradient, namely, Río Claro and La Noria wells, used for the irrigation of banana trees.

In this MAR site, the recharge water is classified as Mg-Cl-HCO_3, and the groundwater belongs to Na-Cl-HCO_3 hydrofacies (Fig. 4, 11). Some cations have been assessed using a hydrochemical calculator.

This pilot is still in construction. The results of this MAR trial will be specified at the end of 2024.

Figures 2 and 3 provide a graphical understanding of these MAR systems; Figure 4 presents the Piper-Hill-Langelier hydrograms plotting the input water in these MAR systems, and the piezometers or wells nearby. It is worth mentioning that all the monitoring piezometers and wells are below 1 km of distance (except for the case 11, where wells are about 1,200 m distant. In all cases, there are water-points to monitor the groundwater quality evolution in lower sites downstrem, according to the groundwater flow. There have been represented analyses from 27 percolated or injected waters, and 35 after interaction processes have taken place in the aquifer (Table 5). Three sites (2, 4 and 6) count only with two analysis, one from a recharge water sample, and another in the observation piezometer downstream. The dissociated hydrograms are displayed in the figures 5 (input water), and 6 (water collected from monitoring points).

The source tables exposing the chemical analysis tables from those sites where owners have given publication permission are exposed in supplementary files, annex 1 (chemical analysis tables). N/P means that permission to publish has not been received, and interested readers should contact the owners for specific tables. It is also worth remarking that all these MAR sites presented their corresponding environmental impact study, according to the mandatory requirement expressed in the Royal Decree 445/2003 (BOE, 2003). Permissions or allowances for MAR require the approval of both, the substantive body (Water authorities), and the environmental body. All these sites obtained their corresponding authorisation, and operate according to law. In this sense, the river basin confederation concerned is in charge of monitoring each MAR activity, following the qualitative evolution of the groundwater in nearby piezometers included in their monitoring network, to ensure that there is no groundwater quality deterioration. Most of the monitored parameters include macro and micro constituents, heavy metals, bacterial content, some pharmaceutical compounds, and even contaminants of emerging concern (CEC), including some of the previous group of substances. None that of the eleven MAR sites have had legal impediments, except the Pedrajas-Alcazarén SAT-MAR (5c), when in 2012 the water authorities (CHD) detected a concentration of total organic carbon exceeding the limits in the closest piezometer. The WWTP improved the purification procedure by investing in a new treatment line, and the impact was solved in a matter of months. None has received other water quality constraints since their operation began.

Variability of water quality across MAR sites

The water quality parameters obtained for each MAR site are shown in Table 4, and the number of water quality analysis and their source are presented in Table 5.

The water quality of the selected Spanish MAR sites displays a wide range of hydrochemical facies comprising the main water types, including, Ca-SO_4, Ca-HCO_3, and Na-SO_4 (Fig. 4). The only hydrofacies rarely represented is Na-Cl, which is exclusively found in the Cobre las Cruces mine (drained groundwater utilized for MAR). Overall, water samples from a particular site (recharge water and recharged groundwater) tend to fall within the same hydrofacies, except for the Los Arenales MAR sites, which show a relatively wider distribution in the Piper-Hill-Langelier hydrogram, due to the extensive geographical area, and the use of different water sources. Similarly, the Guadiana canal, with a length of over 30 km, flows over different lithologies, which accounts for its spread in both all hydrograms (Figs. 5 and 6).

The hydrofacies of native groundwater at Spanish MAR sites are distributed across all domains of a Piper-Hill-Langelier hydrogram similar to the hydrofacies of MAR water sources, reflecting a considerable variability in the proportion of primary ions (Fig. 6). Nonetheless, magnesium cation is never prevalent in both, MAR water sources (Fig. 5), and groundwater in piezometers and wells nearby (Fig. 6).

Systems relying on wastewater, i.e. Pedrajas-Alcazarén (advanced secondary treatment), Port de La Selva, Majorca, and Tenerife SAT-MAR systems (tertiary treatment) have a permanent control and monitoring of the microorganisms presence, and contaminants of emerging concern (CECs), including pharmaceutical compounds´ evolution. Therefore, effluents often meets the required quality, improved thanks to their travel through the saturated and unsaturated zones. Usually, WWTP from big cities include a variable number of CECs (sites 9 and 10), whilst in rural areas the CECs have a negligible presence. This is a matter of concern, and continuous efforts are being made to enhance the purification capacity of the WWTP.

MAR sites relying on river water and snowmelt collected near orogenic barriers, such as the careos canals and the

Tab. 4 - Parameters considered in the water quality analysis of each MAR site. This table exclude parameters below detection limits.
Tab. 4 - Parametri considerati nell'analisi della qualità dell'acqua di ciascun sito MAR. Questa tabella esclude i parametri al di sotto dei limiti di rilevamento.

PARAMETER	1-Careos	2-Cobre las Cruces	3-Guadiana	4-Canal Isabel II	5-Los Arenales	6-Arabayona	7-Valladolid UWB	8-Sant Vicenç Horts	9-Port La Selva	10-Majorca	11-Tenerife
Alkalinity, total	X	X	-	-	X	-	-	X	X	-	-
Chemical Oxygen Demand (COD)	-	-	-	-	-	-	-	-	-	-	X
Conductivity (µS/cm)	X	X	X	X	X	X	X	X	X	-	X
Dissolved Organic Carbon (DOC)	-	-	-	-	-	-	-	-	-	-	-
Max. pH	X	X	X	X	X	X	X	X	X	-	X
Temperature (ºC)	X	-	-	-	X	-	X	-	X	-	-
Total Dissolved Solids (TDS)	-	X	-	-	X	-	-	-	-	-	-
Total nitrogen (N)	-	-	-	-	-	-	-	-	-	-	X
Total Organic Carbon (TOC)	X	-	-	-	-	-	-	X	X	-	-
Total phosphorus (P)	-	-	-	-	-	X	X	X	-	-	X
Total Suspended Solids (TSS)	-	-	-	-	X	-	X	-	-	-	-
Turbidity (NTU)	-	-	-	-	X	-	-	-	X	-	-
Calcium (Ca) hardness in ºF or	X	-	X	-	X	-	X	X	-	X	-
Magnesium (Mg)	X	-	X	-	X	-	X	X	-	X	X
Sodium (Na)	X	X	X	-	X	-	X	X	X	X	-
Chloride (Cl-)	X	X	X	-	X	-	-	X	X	X	-
Sulphate (SO_4^{2-})	X	-	X	-	X	-	X	X	X	X	-
Fluoride	-	X	-	-	-	-	-	-	-	-	-
Nitrite-Nitrate (both as N)	-	-	-	-	-	-	-	-	X	-	-
Nitrate (NO_3^-)	X	-	X	X	X	X	X	X	X	X	X
Nitrite (NO_2^-)	-	-	X	-	X	X	-	X	-	-	-
Ammonia (NH_4^+)	-	-	-	-	X	-	X	X	X	-	X
Phosphates	-	-	X	-	X	-	-	-	-	-	X
Boron (B)	-	X	X	-	-	-	-	-	X	-	-
Cyanide (CN-)	-	-	-	-	-	-	-	-	X	-	-
Faecal Coliforms (f.c /100 ml)	-	-	-	-	-	-	-	-	X	-	-
E.coli (UFC/100 mL)	-	-	-	-	-	-	X	-	-	-	-
Aluminium (Al)	-	-	-	-	-	-	-	X	-	-	-
Antimony (Sb)	-	-	-	-	-	-	-	-	-	-	-
Arsenic (As)6	-	X	-	X	-	-	-	-	-	-	-
Barium (Ba)	-	X	-	-	-	-	-	-	-	-	-
Cadmium (Cd)	-	-	-	-	-	-	-	-	-	-	-
Chromium total (Cr)	-	-	-	-	-	-	-	-	-	-	-
Copper (Cu)	-	X	-	-	-	-	-	-	X	-	-
Iron (Fe)	-	-	-	-	X	-	X	X	-	-	X
Lead (Pb)	-	-	-	-	-	-	-	-	X	-	-
Manganese (Mn)	-	-	-	-	-	-	-	X	X	-	-
Mercury (Hg)	-	-	-	-	-	-	-	-	-	-	-
Nickel (Ni)	-	-	-	-	-	-	-	X	-	-	-
Zinc (Zn)	-	-	-	-	-	-	X	-	-	-	-
Fats and oils	-	-	-	-	-	-	X	-	-	-	-
Naphthalene	-	-	-	-	-	-	-	-	X	-	-

Tab. 5 - Number of water quality analyses analysed per site, including the water source for MAR (recharge water), and nearby piezometers (recharged groundwater). Experimental sites are those related to R&D projects with a determined duration.

Tab. 5 - Numero di analisi della qualità dell'acqua analizzate per sito, inclusa la fonte d'acqua per MAR (acqua di ricarica) e piezometri vicini (acque sotterranee ricaricate). I siti sperimentali sono quelli relativi a progetti di ricerca e sviluppo con una durata determinata.

MAR site	Recharge water. Nº of aliquots	Recharged groundwater. Nº of aliquots	Source
1-Careos	9	3	Barberá et al. (2018)
2-Cobre las Cruces Copper Mine	1	1	Baquero et al. (2016)
3-Guadiana MAR site	1	5	Fernández (2015)
4-Canal Isabel II ASR Sites	1	1	Nogueras et al. (2019)
5-Los Arenales MAR sites	5	3	Fernández Escalante et al., Tragsa (2005b; 2016; 2021a, 2021b)
6-Arabayona MAR site	1	1	Fernández and Paredes (2022b)
7-Urban water buffer Zorrilla (experimental)	3	2	Field Factors, Fajnorová et al. (2021)
8-Sant Vicenç dels Horts MAR site	1	6	Dessin project, Amphos 21 (2016)
9- Port de La Selva SAT-MAR site	1	2	Demoware project, Amphos 21 (2016)
10-Majorca experimental SAT-MAR	3	9	EARSAC, Tragsa (2019)
11-Tenerife experimental SAT-MAR	1	2	CIATF (on going)
Total	27	35	

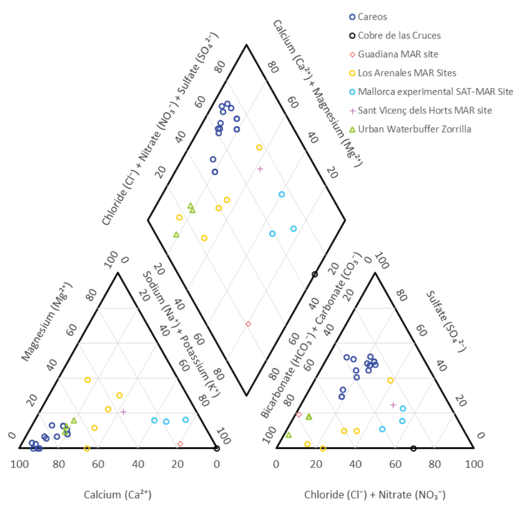


Fig. 5 - Piper-Hill-Langelier hydrogram plotting MAR water sources from selected MAR sites in Spain.

Fig. 5 - Idrogramma Piper-Hill-Langelier che rappresente le fonti d'acqua MAR da siti MAR selezionati in Spagna.

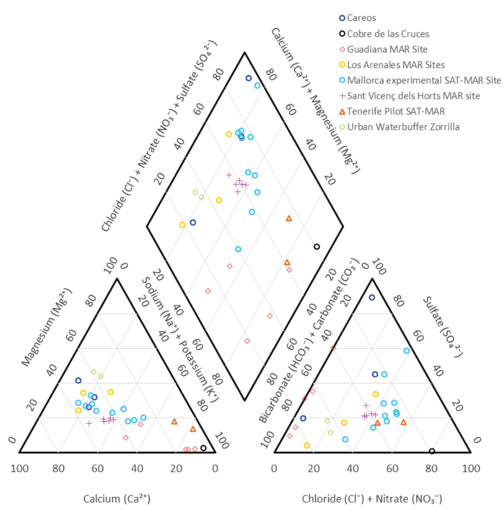


Fig. 6 - Piper-Hill-Langelier hydrogram including recharged and native groundwater quality nearby selected MAR sites in Spain.

Fig. 6 - Idrogramma Piper-Hill-Langelier che include la qualità delle acque sotterranee native e di ricarica vicino ai siti MAR selezionati in Spagna.

Los Arenales (Santiuste and El Carracillo), do not face water quality issues. This is likely due to the little chance water has to interact with anthropogenic or geogenic sources of contaminants. On the other hand, the Guadiana MAR channel and Sant Vicenç dels Horts sites exemplify the situation in which anthropogenic activity in upstream parts of the river has increased the concentration of some water constituents that might become a concern (attested by an electrical conductivity of 746 µS/cm and 1,525 µS/cm, respectively).

The Majorca SAT-MAR experimental site presents varied hydrofacies in groundwater wells that pass through different lithologies at varying depths.

Comparison of MAR water sources with MAC-based standards

The analysis of compliance of the water used for MAR with national MAC regulations is subdivided into the countries' legislation analysed. These input waters are before the point of compliance (POC) of the WFD, or just on the POC in case of direct injection. On the other side of the POC, MACs apply at the level of the groundwater body and reflect unsaturated zone capacity and interaction processes in the saturated zone as well.

Italian standard

The MAC standard for Italy (MATTM, 2016), considering the hydrodynamic approach of this regulation (Lippera et al., 2023), has been breached 21 times in the 11 MAR Spanish sites presented before (chemistry tables in annex 1). Chloride and ammonia are the parameters more times exceeded (six times), followed by nickel (four times). The threshold of the latter parameter has been topped in 100% of the MAR water source samples (Table 6). The exceeded column expressed in percentage (column 4) might be more expressive than the number of times above a certain value is surpassed.

Italy's regulatory standard would have prevented MAR implementation in six sites: Cobre las Cruces copper mine, the Zorrilla urban water buffer, Sant Vicenç dels Horst, Port de La Selva, Majorca, and Tenerife experimental SAT-MAR sites. Note that these sites are legally operating, and contamination to the native groundwater has not been detected or reported by promotors or water/environmental authorities. On the contrary, it results beneficial for the receiving aquifer, as water quality is, in general, better (according to the Annex 2's tables).

Tab. 6 - Analysis of exceedance of MACs in the 11 MAR sites using the standard for Italy. N represents the number of comparisons between a MAC and the value measured in a MAR water source. The exceeded columns represent the times a given MAC is surpassed in number (N) and percentage (%).

Tab. 6 - Analisi del superamento dei CMA negli 11 siti MAR utilizzando lo standard per l'Italia. N rappresenta il numero di confronti tra un CMA e il valore misurato in una fonte d'acqua MAR. Le colonne superate rappresentano le volte in cui un dato CMA viene superato in numero (N) e percentuale (%).

Parameters	Total (N)	Exceeded (N)	Exceeded (%)
Conductivity (μS/cm)	27	0	0%
Chloride (Cl^-)	27	6	22%
Sulphate (SO_4^{2-})	28	0	0%
Fluoride	1	1	100%
Nitrate (NO_3^-)	31	1	3%
Nitrite (NO_2^-)	6	0	0%
Ammonia (NH_4^+)	11	6	55%
Boron (B)	4	1	25%
Arsenic (As)[6]	4	2	50%
Lead (Pb)	1	0	0%
Nickel (Ni)	4	4	100%
Total	**144**	**21**	**15%**

Spanish standards

For direct injection, the most commonly breached parameter in the Spanish standard (BOE, 2007), is nitrate, which exceeded the standard (established at 25 mg NO_3/L) about a 23 %. Nonetheless, total nitrogen, E. coli are exceeded more frequently in terms of percentage (100%) and total suspended solids (75%) (Table 7). This MAC standard would preclude operations in six MAR sites: the Guadiana MAR site, the Pedrajas-Alcazarén site of the Los Arenales aquifer, the Urban water buffer Zorrilla, the Sant Vicenç dels Horst MAR Site, the Majorca and the Tenerife experimental SAT-MAR sites. Note that this standard is one of the most restrictive for MAR water quality, as it deals with systems directly injecting water in the aquifer (although only the Canal de Isabel II and the Zorrilla systems conduct an "injection" in the most rigid sense of the word, what entails a sealed circuit, a pump, and electricity supply).

Tab. 7 - Analysis of exceedance of MACs in the 11 MAR sites using the standard for direct injection for Spain from available data. N represents the number of comparisons between a MAC and the value measured in a MAR water source.

Tab. 7 - Analisi del superamento dei CMA negli 11 siti MAR utilizzando lo standard per l'iniezione diretta per la Spagna dai dati disponibili. N rappresenta il numero di confronti tra un CMA e il valore misurato in una fonte d'acqua MAR.

Parameters	Total (N)	Exceeded (N)	Exceeded (%)
Total nitrogen (N)	1	1	100%
Total Suspended Solids (TSS)	4	3	75%
Turbidity (NTU)	3	1	33%
Nitrate (NO_3^-)	31	7	23%
E.coli (UFC/100 mL)	1	1	100%
Total	**40**	**13**	**33%**

The other standard of the Spanish regulation, which addresses percolation (water is recharged by gravity, and it crosses the vadose zone), is consequently less restrictive (BOE, 2007), also referred to the Art. 257 to 259 of the RD 849/1986 (BOE, 1986) about spill authorizations. It shows nitrate as the parameter that exceeded more times the limit (two times) (Table 8). However, in terms of percentage, nitrate is rarely exceeded (6%). This standard would put in stake operations at three MAR sites: Pedrajas-Alcazarén, the Urban Water buffer Zorrilla, and the Tenerife SAT-MAR site.

Tab. 8 - Analysis of exceedance of MACs in the 11 MAR sites using the standard for percolation for Spain. N represents the number of comparisons between a MAC and the value measured for a MAR water source.

Tab. 8 - Analisi del superamento dei CMA negli 11 siti MAR utilizzando lo standard di percolazione per la Spagna. N rappresenta il numero di confronti tra un CMA e il valore misurato per una fonte d'acqua MAR.

Parameters	Total (N)	Exceeded (N)	Exceeded (%)
Total nitrogen (N)	1	1	100%
Total Suspended Solids (TSS)	4	0	0%
Nitrate (NO_3^-)	31	2	6%
E.coli (UFC/100 mL)	1	1	100%
Total	**37**	**4**	**11%**

Dutch standard

The standard from The Netherlands for MAR water (Minister van Volkshuisvesting, 1993), is exceeded for most parameters, except lead and naphthalene. The most breached MACs were nitrate (eight times), chloride (eight times), sodium (six times), and nickel (four times) (Table 9). The application of this standard would have prevented MAR implementation at seven sites: the Cobre las Cruces copper mine, the Guadiana MAR site, the Pedrajas-Alcazarén site from the Los Arenales aquifer, the Zorrilla urban water buffer, the Sant Vicenç dels Horst MAR site, the El Port de La Selva SAT-MAR, the Majorca SAT-MAR, and the Tenerife SAT-MAR.

Tab. 9 - Analysis of exceedance of MACs in the 11 MAR sites using the standard for The Netherlands. N represents the number of comparisons between a MAC and the value measured for a MAR water source.

Tab. 9 - Analisi del superamento dei CMA nei 11 siti MAR utilizzando lo standard per i Paesi Bassi. N rappresenta il numero di confronti tra un CMA e il valore misurato per una fonte di acqua MAR.

Parameters	Total (N)	Exceeded (N)	Exceeded (%)
Sodium (Na)	27	6	22%
Chloride (Cl^-)	27	8	30%
Sulphate (SO_4^{2-})	28	5	18%
Fluoride	1	1	100%
Nitrate (NO_3^-)	31	8	26%
Ammonia (NH_4^+)	11	2	18%
Phosphates	5	3	60%
Cyanide (CN^-)	1	1	100%
Arsenic (As)[6]	4	2	50%
Barium (Ba)	1	1	100%
Copper (Cu)	3	1	33%
Lead (Pb)	1	0	0%
Nickel (Ni)	4	4	100%
Zinc (Zn)	1	1	100%
Naphthalene	1	0	0%
Total	146	43	29%

The MAC that posed more problems to the Spanish MAR sites was nitrate, due to the agricultural context in which many are involved directly or indirectly. Other frequently exceeded parameters were chloride, probably related to the proximity to the sea at many sites, and nickel. The Spanish standard for direct injection was the most restrictive MAC standard in terms of the total percentage of parameters breached (i.e., 33%). Nonetheless, as stated above, this standard is aimed at particular projects and assumes no treatment in the vadose zone takes place. Consequently, it has more stringent thresholds, and it has expectations to be imminently revised, (BOE, 2023).

The most restrictive MAC standard regarding the number of sites that wouldn't meet the requirements is the Dutch, which would preclude operations at seven locations (not considering the impact of the unsaturated zone for the percolation MAR systems). This standard also had the second-highest rate of parameter rejections (i.e., 29%).

Discussion

A sound standard based on MACs to control water contamination during MAR should be able to consider exemptions due to the existing water quality, when MAR proves to be beneficial. Probably, it shouldn't pose too stringent limits that could deter the implementation of successful MAR sites, in case they fulfil the water and environmental regulations.

With the focus on Spain, where the eleven MAR sites have been presented, characterizing their water quality. The analysis of major constituents and hydrofacies shows the wide range of source water quality for MAR, used to recharge varied geological receiving aquifers.

There have been some advances in terms of MAR regulations, in a permanent adaptation to the climate change context. Previously, MAR was considered a mechanism for water disposal. The recent amendment to the Spanish Water Act, i.e. Royal Decree 665/2023, RDPH (BOE, 2023), published on August 31st, 2023, ascribes the term managed aquifer recharge to the traditional concept of artificial recharge (Art. 273-1k), declares that MAR will not be considered a spill in Spain any longer (Art. 273-1), and declares: "Any surplus volume of water of appropriate quality shall be capable of being used for the artificial recharge of aquifers..." (Art. 273-3). This recent modification to the Spanish Water Act (BOE, 2023), also requires a justification of the need for the recharge and the destination of the stored water, including: a detailed hydrogeological report (a); a feasibility and compatibility report with the water bodies, including the origin, qualitative characteristics of the recharge water, accreditation of availability, and verification of non-affection to the associated environment (b); a detailed description of the recharge system and the associated works, installations, infrastructure, maintenance and control, etc. (c); volume of water to be recharged and forecast of the effects, including interaction with pre-existing piezometric levels (d); and proof of availability of land (e).

Despite the new modification of the Water Act, the MACs considered in the Royal Decree 1620/2007 (BOE, 2007) have not been modified yet, probably waiting for final Pan-European MAR guidelines, which are currently drafted by the CIS (CIS, 2023). This means that a MAC-based approach still remains valid for the entire Spanish territory, whilst another approach is feasible.

As most of the exposed MAR sites received the corresponding permission or allowance from water and environmental authorities, who monitor the evolution of each system, they have been operative for years improving groundwater quality (in isolated cases thanks to the effect of the unsaturated zone as well). They also count on great social acceptance from the stakeholders and end-users. Probably, the MACs-based regulations are not the most appropriate to solve the MAR water quality challenge. For example, if the water available to recharge exceeds slightly the nitrates limit of 50 ppm, and the mean concentration in the receiving aquifer water exceeds 100 ppm, as it happens in some spots of the Guadiana channel, the recharge would be positive in environmental terms. Despite this fact, it might be banned from a legislative point of view. Also, MAR with low-quality water may be used to improve the quality of aquifers where groundwater quality is still lower. It is worth mentioning that the assessed concentrations will be modified by direct dilution, dispersion, diffusion, sorption, and biodegradation, at least.

Some of the exposed sites have a small number of data, what could be a limitation of the study, although all of them counts on, at least, one analysis of the input water, and another from a downstream piezometer, what fulfils legislative requirements (BOE, 2023).

Alternatives to source water quality standards relying on MACs include a risk-based approach, in which all potential risks entailed in MAR operations are assessed, and measures are designed so that the resulting residual risk is acceptable. This approach also applies to contaminants and, consequently, safeguards water quality in a meaningful way for the local context and hydrogeological conditions. The risk-based approach is the cornerstone of guiding and regulative documents entailing MAR, such as the Australian Guidelines for Water Recycling (NRMMC, EPHC, NHMRC, 2013), and the WHO Guidelines for the safe use of wastewater, excreta and greywater (WHO, 2006), and are considered a sound way to regulate MAR operations (Zheng et al., 2023).

In some SAT-MAR cases, the use of the aquifer to improve the quality of the effluents (SAT) is not aligned with WFD requirements. This could lead to building up an argument that the saturated zone can be considered as well (so, going beyond the WFD) subject to ensuring safety. The best direction is improving the purification capacity of WWTPs.

The monitored and intentional recharge (MIR) concept (Fernández et al., 2023) proposes a list of minimum blocks or elements to consider when drafting guidelines for MAR. The concept has been built based on the existing regulations for artificial recharge. The nine blocks that integrate the MIR conceptual model are:

1. Water sources for MAR
2. Environmental conditions in which MAR activities take place, including the climate, aquifer type, geology, surface water basin, groundwater body, and depurative capacity
3. MAR technology
4. MAR sensors for data gathering, allowing to characterise the environmental conditions and monitoring the system
5. Guidelines for hydrodynamic monitoring of the water (quantity and quality), which include aspects on sampling frequency and location
6. The final use of the recovered water, including irrigation, water supply, and hydraulic barriers against seawater intrusion, among others
7. Analytical aspects, with recommendations on the scope and scale of the maximum allowed concentrations of potential contaminants, including pharmaceutical compounds and other CECs.
8. Risk assessment, elaborating on some of the risks to assess when conducting MAR operations, considering dependent habitats, taking special care in fulfilling the demands of environmental impact regulations.
9. Other topics that do not fit in the previous blocks are standardisation and interoperability, new contaminants of emerging concern (CECs), economic aspects, public participation, active stakeholder engagement, etc.

Interaction process modelling should be incorporated into usual tools for regulators. The possibility of using the effect of the unsaturated zone to improve recharge water quality must be studied carefully.

Currently, the precautionary principle, reflected in some MAR guidelines and regulations at the European level has been considerably stringent. A novel approach might be needed to protect water quality while allowing for adaptation to climate change. It is probable that there is no need for new regulations. Perhaps a wise modification of the WFD, as partly achieved through the 2020/741 regulation (EC, 2021a), which sets minimum requirements for water quality according to final uses (in this case, agricultural irrigation), might suffice. It is noticeable that the WFD already allows for water management at the level of the river basin disaggregated from the national level, therefore, the context at the European level is appropriate.

Another aspect of great importance is the synergy between water reuse and MAR since, in many cases, treated wastewater is recharged to purify water further before final use. The potential for water reuse in the European context, and specifically in Spain, is considerable. Consequently, MAR regulations should also bear in consideration to reuse actions aimed at achieving the good status of water bodies (revision and adaptation of RD 1620/2007 to European Regulation 2020/741, and to RD 665/2023).

Conclusions

The 11 Spanish Managed Aquifer Recharge (MAR) sites studied employ different methods and processes. They also rely on water sources that entail multiple quality challenges, ranging from nitrates to heavy metals and contaminants of emerging concern. The chemical analyses from these water sources cover all Piper-Hill-Langelier hydrogram. The sites also utilise conventional and unconventional sources of water, including treated groundwater (Figs. 2, 2 a-b), treated wastewater (Figs. 3, 9-10-11), and even potable water (Fig. 2, 4 a-b). This high variability may suggests the difficulties to establish limits for water quality parameters that fit all so varied MAR sites.

The different origins and the sources of water quality are typically different from site to site and country to country. What is foreseen is to establish maximum values, so that groundwater does not become contaminated due to MAR for each specific context, i.e. aquifer-wide level. Exceptions might be considered in cases 1) where the aquifer is already contaminated and the source water has better quality (as the example given in this paper for nitrates), and 2) where the foreseen treatment (e.g., SAT-MAR) proves that the resulting infiltration water will have concentrations below the MACs.

MAR sites in which the water intake occurs near headwaters or in watersheds with a relatively low anthropic intervention (e.g., careos), have the least water quality problems. On the other hand, sites sourcing water from highly intervened water bodies, especially those affected by urban effluents, wastewater treatment plants, or agriculture, pose the highest risk of water pollution if not adequately addressed.

The self-purification capacity for each receiving medium is different and must be considered, as long as its capacity is not exceeded, which depends, not only on the initial water quality, but also on the pre-treatment and post-treatment processes, the characteristics of the receiving medium (e.g., granular aquifers poses a higher self-purification than hard rocks).

Within this framework and based on the results, it seems difficult to propose a single MAC regulation without imposing too stringent limits on water quality, to the point of jeopardizing future useful MAR implementations, when the exposed systems: 1) comply with the legislated mandatory environmental requirements, 2) have received their corresponding permission or allowance from water and environmental authorities, and 3) groundwater quality evolution is monitored permanently. This is the case of some MAR systems operating for even 22 years (5a) (e.g. Santiuste basin) without relevant constraints, and they count on a general social acceptance, satisfying end-users. The comparison of water quality analysis from the studied sites with the maximum allowable concentrations (MACs) regulated in Italy, Spain, and The Netherlands, shows that infiltration water of some MAR sites breached some MACs, even though the data analyses from the nearby piezometers monitored by water authorities demonstrate that they are not polluting or deteriorating local water resources, but on the contrary, some water quality improvements have been reported. This evidence suggests that national or regional MACs are likely not the best way to prevent contamination while conducting MAR. In the case of establishing MACs to control water quality in the MAR water source, limits should be imposed at the local or "aquifer-wide" level, despite the risk of hampering potential MAR implementations. A MAC approach to control source water quality for MAR seems inappropriate considering the various water sources, lithologies of the receiving aquifers (and therefore different capacities of biodegradation, adsorption, etc.), and environmental conditions (e.g., redox conditions and pH). Therefore, a single threshold list even drafted for aquifer-wide scale may deter necessary deployments. Besides, MACs could restrict operations at sites that are currently legally operating without contaminating aquifers. For instance, the Guadiana MAR site couldn't be implemented under Italian and Dutch standards because the water source breaches some MACs. Nonetheless, in this site, the water percolated has a higher quality than the native groundwater (which does not meet the MAC standards either), and is helping to improve the general aquifer water quality, monitored each quarter by water authorities (CHG).

In summary, it seems recommendable a detailed study should be conducted for each specific case when granting a MAR permission or allowance.

The establishment of MACs in any regulation should also indicate what the final risk is, considering that each risk assessment is site-specific. A risk-based approach and conceptual models for formulating guidelines, such as the monitored and intentional recharge (MIR) concept, may considerably help in implementing MAR systems worldwide.

The need to regulate MAR adequately is imperative as, today, MAR is optional, but in the future, it might become necessary.

Supplementary Materialss

ANNEX 1: Chemical analysis tables.
ANNEX 2: Maximum allowable concentration for MAR source water in Italy, Spain, and The Netherlands legislation.
Online on: www.acquesotterranee.net

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Data sources

All data (unless specified) are in Tragsa's repository. Photos (unless specified) are of the first author.

Competing interest

The authors declare no conflict of interest. The funders had no role in the design of the study, in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

Author contributions

Conceptualisation, Enrique Fernández Escalante (EFE) and José Henao Casas (JHC); methodology, EFE and JHC; formal analysis, JHC and EFE; investigation, all; resources and data acquisition, EFE; data curation, all; writing- original draft preparation, JHC and EFE; writing-review and editing, all; visualisation, all; supervision, all; project administration, Rodrigo Calero Gil (RCG) and EFE; funding acquisition, EFE.

Additional information

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