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# Pump-and-treat (P&T) vs groundwater circulation wells (GCW): Which approach delivers more sustainable and effective groundwater remediation?

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ABSTRACT

Pump-and-treat (P&T) is commonly used to remediate contaminated groundwater sites. The scientific community is currently engaged in a debate regarding the long-term effectiveness and sustainability of P&T for groundwater remediation. This work aims to provide a quantitative comparative analysis of the performance of an alternative system to traditional P&T, to support the development of sustainable groundwater remediation plans. Two industrial sites with unique geological frameworks and contamination with dense non-aqueous phase liquid (DNAPL) and arsenic (As) respectively, were selected for the study. At both locations, attempts were made for decades to clean up groundwater contamination by pump-and-treat. In response to persistently high levels of pollutants, groundwater circulation wells (GCWs) were installed to explore the possibility of accelerating the remediation process in unconsolidated and rock deposits. This comparative evaluation focuses on the different mobilization patterns observed, resulting variations in contaminant concentration, mass discharge, and volume of extracted groundwater. To facilitate the fusion of multi-source data, including geological, hydrological, hydraulic, and chemical information, and enable the continuous extraction of time-sensitive information, a geodatabase-supported conceptual site model (CSM) is utilized as a dynamic and interactive interface. This approach is used to assess the performance of GCW and P&T at the investigated sites. At Site 1, the GCW stimulated microbiological reductive dichlorination and mobilized significantly higher 1,2-DCE concentrations than P&T, despite recirculating a smaller volume of groundwater. At Site 2, As removal rate by GCW resulted generally higher than pumping wells. One conventional well mobilized higher masses of As in the early stages of P&T. This reflected the P&T's impact on accessible contaminant pools in early operational periods. P&T withdrew a significantly larger volume of groundwater than the GCW. The outcomes unveil the diverse contaminant removal behavior characterizing two distinct remediation strategies in different geological environments, revealing the dynamics and decontamination mechanisms that feature GCWs and P&T and emphasizing the limitations of traditional groundwater extraction systems in targeting aged pollution sources. GCWs have been shown to reduce remediation time, increase mass removal, and minimize the significant water consumption associated with P&T. These benefits pave the way for more sustainable groundwater remediation approaches in various hydrogeochemical scenarios.

## 1. Introduction

Pump-and-treat (P&T) undoubtedly represents the most widely adopted strategy for decontaminating polluted groundwater that contains dissolved chemicals such as industrial solvents, metals, fuel oil, and emerging contaminants (Bagatin et al., 2014; Mackay and Cherry, 1989; Majone et al., 2015; Sheng et al., 2018; Mukhopadhyay et al., 2022; Newell et al., 2021). Evidence gathered from long-term field implementations at different sites suggests that although P&T may be effective in the early phases of application, performance substantially drops with

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time (Naseri-Rad et al., 2021; Suthersan et al., 2015). The initial pump-and-treat stage is characterized by high pollutant removal rates. According to Brusseau and Guo (2014) and Guo et al. (2019), it reflects both the large amounts of pollutants stored in the contamination sources and the hydraulic gradient induced by the pump-and-treat system. A steady-state concentration "plateau" stage follows concentration reduction and plume shrinkage (Mackay and Cherry, 1989; Rivett et al., 2006). Several authors ascribe this asymptotic phase to the impact of wellfield hydraulics, poorly accessible pollutant mass due to permeability heterogeneity on mass transfer and mass removal. The well-known phenomena of pollutant desorption and back-diffusion from secondary sources within low-permeability media can continue for decades (Petrangeli Papini et al., 2016; Pierro et al., 2017; Tatti et al., 2018, 2019). As a result, considerable levels of residual contamination may persist by further treatment (Besha et al., 2018). Remediation of groundwater contamination typically proceeds slowly employing the common pump-and-treat approach, increasing both the costs and time to reach cleanup goals (Mackay and Cherry, 1989; Rivett et al., 2006). Careful consideration should also address the potential issues that might arise from the withdrawal of huge volumes of groundwater and the consumption of water resources compared to in situ technologies (Elmore and Graff, 2002; Elmore and De Angelis, 2004).

In light of this, a particular emphasis can be placed on groundwater circulation wells (IEG-GCW), a conservative in situ solution of zero groundwater discharge. The GCW technologies consist of a multi-screened vertical well. Groundwater is extracted from one aquifer horizon and after treatment recharged to another horizon (Ponsin et al., 2014; Wang et al., 2023). This induces the development of ellipsoidal groundwater circulation cells, enhancing vertical hydraulic gradients and boosting the mobilization and removal of pollutants (Herrling et al., 1991a, 1991b, 1993a, 1993b; Stamm, 1997; Xia et al., 2019). Contaminated groundwater is delivered to a treatment plant above ground, that can be configured flexibly depending on the pollutant, to reduce contaminant concentration before re-injection into the aquifer.

The ability of the recirculation system to mobilize secondary sources of contamination adsorbed to the fine material in the saturated domain of a dense non-aqueous phase liquid (DNAPL) - contaminated aquifer is detailed in previous works (Ciampi et al., 2019a, 2021a; Tatti et al., 2019). Utilizing an electron donor system simultaneously empowers biological reductive dechlorination (BRD) and pollutant mobilization to be combined (Petrangeli Papini et al., 2016; Pierro et al., 2017). Ciampi et al. (2023) reveal the possibility of recirculating significant groundwater flow rates in a calcareous bedrock via a GCW, attacking pools of arsenic (As) trapped in the fractured medium. GCW technique was also tested to improve the distribution of a biostimulant in a heterogeneous aquifer for reductive dehalogenation and create an in-situ bioreactor for the enhanced treatment of chlorinated aliphatic hydrocarbons (CAHs) (Ciampi et al., 2022c). Particular recirculation systems can also be adapted to limited aquifer thicknesses, where drawdown would result in aquifer desaturation (Ciampi et al., 2022b).

Within the context of contaminated site projects, a data-driven, multi-source conceptual site model (CSM) can harmonize geological and hydrochemical information during different remediation stages, supporting the remedial design to suit the physicochemical conditions and unmasking the decontamination mechanics induced by the remedial actions (Ciampi et al., 2019b, 2021b, 2022a; Suthersan et al., 2016). Geodatabase-driven CSM exhibits the potential to act as a decision-support tool for assessing the sustainability of remediation alternatives through the use of an adaptive site management strategy and the application of computational techniques and technological performance evaluation tools (Huysegoms and Cappuyns, 2017; Price et al., 2017; Truex et al., 2017).

This paper presents a comparative performance evaluation of two groundwater remediation technologies, such as P&T and GCWs. The benchmarking focuses on the abatement capacity of pollutant concentrations/masses and groundwater volumes employed in long-term treatments. A comparison of this kind is currently lacking in the contemporary literature and may provide relevant insights into the pollutant removal behavior of different strategies adopted for groundwater pollution source control/degradation, orienting future decision-maker's choices toward developing sustainable remediation plans for groundwater, in balance with natural resource conservation considerations and limiting the adoption of remediation strategies that may not be protective of the groundwater resource (Connor et al., 2017).

This study focuses on two aged polluted industrial sites with distinct geological and pollution situations. Granular sediments and chlorinated hydrocarbons with few chlorine atoms such as 1,2-dichloroethylene (1,2-DCE) and vinyl chloride (VC) polluting groundwater are distinguishing characteristics of Site 1. Industrial degreasing processes utilized chlorinated solvents to prepare metal components for spray painting until 1987 (Petrangeli Papini et al., 2016; Pierro et al., 2017). A calcareous bedrock and historical sources of arsenic contamination in the aquifer characterize Site 2 (Ciampi et al., 2023). Arsenic oxides accumulated in the fractured aquifer as a result of the 1976 explosion of an industrial plant (Liberti and Polemio, 1981; Gianicolo et al., 2019; Mangia et al., 2018). Both sites feature decades of P&T efforts to mitigate groundwater pollution. After the persistence of significant pollution concentrations in groundwater and the prolonged withdrawal of water resources for traditional treatment, GCWs were installed. A benchmarking analysis, which includes an evaluation of the discharge rates and volumes of groundwater withdrawn from pumping wells and recirculated by GCWs, along with a comparison of mobilized concentrations, mass balances of pollutants removed from recirculation wells, and treatment plant associated with P&T systems over time, has the goal of (i) assessing the sustainability and performance of the two technologies in removing and treating secondary sources of pollutants; (ii) emphasizing the different pollutant removal behavior that characterizes distinct remediation strategies in different geological environments; and (iii) highlighting the limitations of traditional groundwater extraction and containment systems in impacting aged contaminants.

## 2. Materials and methods

### 2.1. Overview of Site's 1 and 2 geology and remediation history

This paper builds upon previous studies (Ciampi et al., 2019a, 2021a, 2023) that discuss the conceptual site models (CSMs) and remediation actions taken at industrial plants. Site 1 is located in the Po Plain region of northern Italy (see Fig. 1a), characterized by fluvial sediments, conoids, and alluvial deposits (Meisina et al., 2022). The CSM presented by Ciampi et al. (2019a) shows that the facility rests on alternating layers of sand, silt, gravel, and clay that extend to a depth of around 30 m. The underlying clays act as an aquitard, isolating water circulation in the sediments above and creating a single aquifer body. This model was developed using geological data from 56 drilled boreholes, and 106 groundwater monitoring points have been established at different depths along the saturated vertical, as detailed in Ciampi et al., (2021a). The average depth to the water table is approximately 1.9 m, and the groundwater flows from south to north. Several remediation measures have been implemented at Site 1 over the years to address the issue of chlorinated solvents in the groundwater. Since 2006, a pump-and-treat (P&T) system with 34 pumping wells has been gradually installed, and the current flow rate is around 53 $m^3$ $h^{-1}$. Next, a GCW was installed and operated in two distinct phases (Fig. 1b). These periods are detailed in the study by Ciampi et al. (2021a) and correspond to the implementation of the GCW at the pilot scale in 2014 (phase I) and the subsequent upscaling of the intervention in 2019 (phase II).

Site 2 is located along the Adriatic Sea in southern Italy (see Fig. 2a) and surrounded by fractured limestones from the Cretaceous period (Larsen et al., 2010). The complex geology of the area, as elucidated by Ciampi et al. (2023), includes a sequence of filling materials, conglomerates, breccias, and a limestone aquifer that extends to a depth of 80 m.

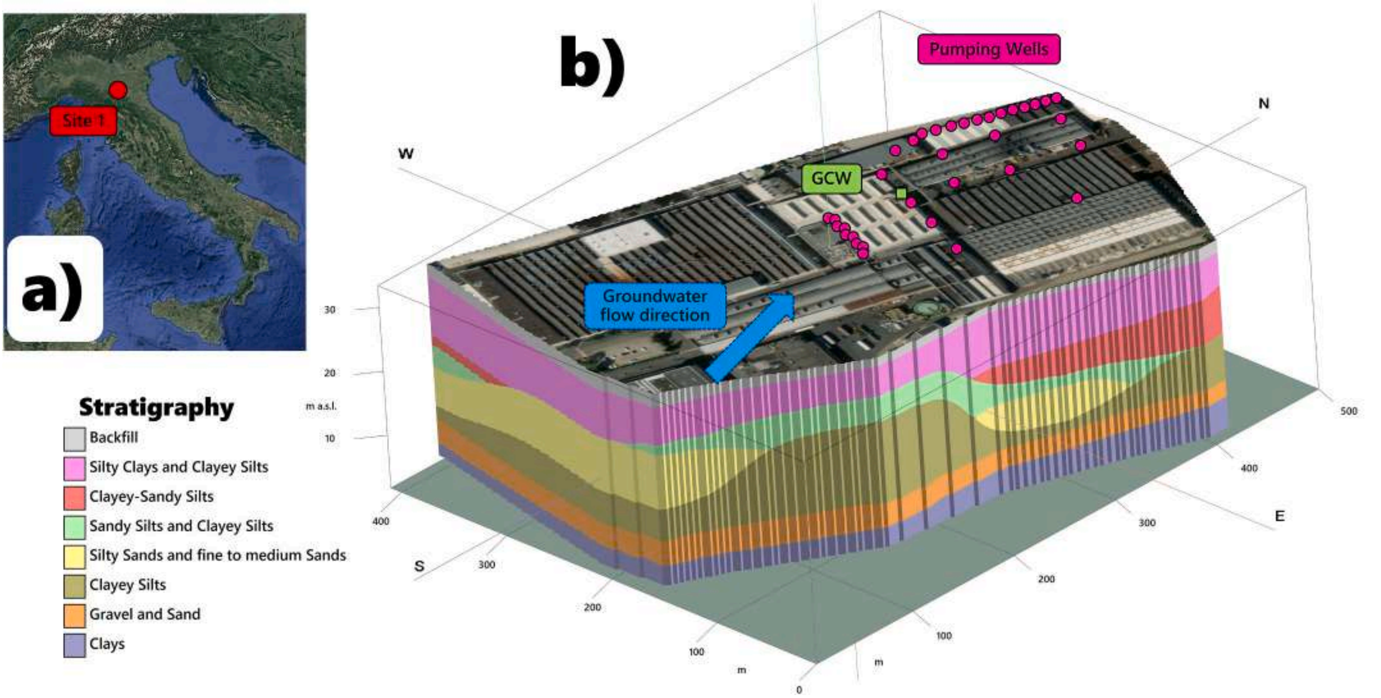

**Fig. 1.** Geographical location of Site 1 in Italy (a) and a 3D geological model of the site, including the position of pumping wells in the industrial plant and the preferential direction of groundwater flow (b).

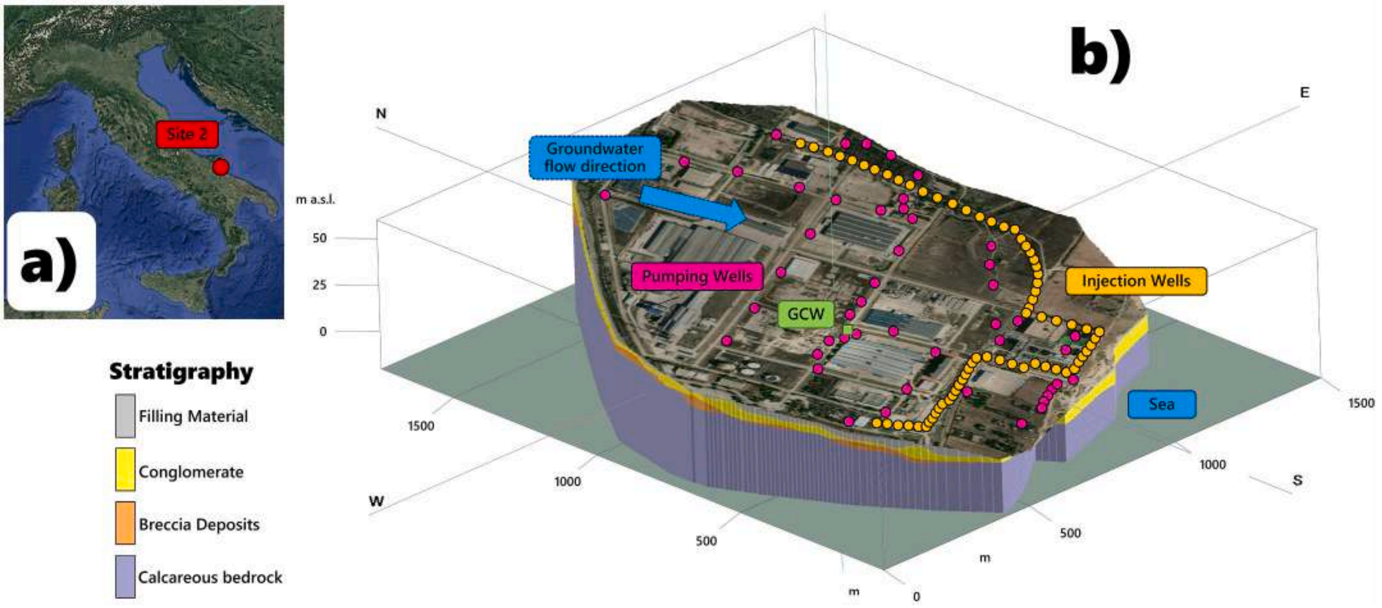

**Fig. 2.** Geographical location of Site 2 on the Italian peninsula (a) and a 3D stratigraphic model of the site indicating the position of the industrial plant's pumping wells and the direction of groundwater flow (b).

This sequence was deduced from 812 surveys completed in the area. Since 2006, 212 points of the piezometric network, which intercept the limestone aquifer, have been monitored. Under undisturbed conditions, the elevation of groundwater is close to sea level, and the flow direction is oriented from approximately north to south. Over the past 15 years, a P&T system has been gradually implemented to contain a long-lasting As plume within the plant and prevent dissolved contaminants from migrating toward the sea. The P&T system currently consists of 48 pumping wells and operates at a flow rate of 188 $m^3$ $h^{-1}$. The wells for extracting groundwater are equipped with screens that cover different depths, ranging from approximately 15 m to 71 m. Additionally, 68 wells inject clean water into the downstream area of the plant to recharge the aquifer. This recharge helps to reverse the groundwater flow near the coastline, creating a reverse hydraulic gradient that mitigates the intrusion of saltwater resulting from the plant's extraction activities. To expedite the removal of As in the secondary source zone pinpointed by the CSM of Ciampi et al. (2023), a GCW was installed in 2020 (Fig. 2b) and kept in operation for a single extended stage (phase I).

It should be noted that the multi-source conceptual site models

(CSMs) presented for both Site 1 and Site 2 were derived from a comprehensive three-dimensional geomodeling exercise based on data stored in a georeferenced geodatabase.

### *2.2. Multi-source data mining from dynamic CSMs*

The information available for the two analyzed sites is the result of ongoing updates to the geodatabases and CSMs discussed in Ciampi et al. (2019a, 2021a, 2023).

The geodatabases for two sites also feature particle size analysis, permeability measurement in the triaxial cell, step-drawdown tests and slug tests, Lugeon and pumping tests, construction designs, and schemes of wells and piezometers (screened and blind parts).

Flow rates from active P&T systems on-site, pollutant concentration data from long-term P&T operations and hydrological monitoring, as well as groundwater extraction well and piezometer design schemes, enrich the big data package. Supplementary characterization investigations (i.e., geological surveys), hydrogeochemical monitoring campaigns, operational changes, and modifications to active remediation systems are stored and updated in near real-time in the data management models. Quasi-continuous reprocessing of information stored in multiple Excel worksheets constituting the relational geodatabases leads to dynamic redrafting and updating of the CSMs.

Geodatabase-driven geomodeling procedures dynamically follow the gathering and storage of new information to refine and enhance the conceptual hydrogeological model in a virtually continuous fashion. A multi-source, data-driven model is expected to be developed by spatially interpolating stratigraphic and hydrological parameters, such as stratigraphic deposit depths and groundwater elevations. Hydrogeological variables are interpolated with the inverse distance weighted (IDW) algorithm, employing a number of neighboring points ranging from 3 to 6 and an exponent generally equal to 2. A high-fidelity filter guarantees the exact interpolation of parameters at known points. Hydrogeomodeling aims to depict geological architecture, delineate saturated aquifer thicknesses, and highlight the dynamics induced by hydraulic interventions, gradually updating, validating, and refining the conceptual framework. This has the purpose of acting as a dynamic and interactive interface for the continuous extraction of multi-source, time-sensitive georeferenced attributes where remedial actions operate. The sampling and recovery of information from the geodatabase-driven dashboard aim to unveil hydraulic perturbations induced by the adopted remediation techniques by overlaying geological and hydrological information in space-time. This is intended to guide the remodeling and reconfiguration of flow rates for groundwater remediation interventions at the field scale, optimizing the performance of adopted measures.

Also, the coupled extraction of hydrochemical and hydraulic information from geodatabase-driven analyses for GCWs and conventional remedial systems has the goal of revealing the dynamics and decontamination mechanisms that characterize different remedial technologies. The examination of time trends in pollutant concentration measurements from samples collected in both conventional extraction well systems (during the initial and later phases of operation) and GCWs aims to provide evidence regarding contaminant transformations, as well as the mobilization and removal capacities of these two remediation systems. The concentration trends of contaminants at pumping wells were analyzed in two phases: the concentration reduction or shrinkage stage (phase I) and the plateau or asymptotic phase (phase II), as previously described in studies conducted by Brusseau and Guo (2014), Guo et al. (2019), Mackay and Cherry (1989), and Rivett et al. (2006). It is worth noting that at Site 1, the GCW was operated in two distinct remediation periods (phases I and II), which were detailed by Ciampi et al. (2021a). To assess the sustainability and performance of the two technologies for removing and treating secondary sources of contamination, a comparative evaluation is conducted using data on groundwater withdrawal rates from pumping wells, recirculation rates from GCWs, mobilized concentrations, and mass balances of pollutants removed by recirculation wells and the associated treatment plant of P&T systems over time. The findings aim to underscore the diverse pollutant removal behaviors characterizing two distinct remediation strategies in different geological environments. Additionally, the study seeks to bring attention to the limitations of traditional groundwater extraction and containment systems in effectively targeting aged pollution sources.

## 3. Results and discussion

### *3.1. Hydrostratigraphic framework around GCWs and pumping wells at sites 1 and 2*

The local stratigraphic sequence of Site 1, as described in Ciampi et al. (2019a, 2021a), depicts the rhythmically alternating and juxtaposition of fine and coarse deposits, identifying a single, heterogeneous, and variously communicating aquifer, characterized by uneven permeability. The vertical hydraulic conductivity of silty-clay deposits, obtained via triaxial cell, varies between 2.04 x $10^{-11}$ m $s^{-1}$ and 1.07 x $10^{-9}$ m $s^{-1}$. Step-drawdown tests return a hydraulic conductivity coefficient value for sandy-gravelly deposits between 9.1 x $10^{-5}$ m $s^{-1}$ and 1.2 x $10^{-4}$ m $s^{-1}$. Slug tests yield permeability between 5.4 x $10^{-8}$ m $s^{-1}$ and 1.3 x $10^{-7}$ m $s^{-1}$ for sandy silts. Several wells extracting contaminated groundwater for in-plant treatment and a GCW are installed in the DNAPL source area (Fig. 3a). The recirculation system develops stacked ellipsoidal recirculation cells by extracting groundwater from the filter sections between 8 and 12 m and 15 and 19 m and reinjecting it into the lower filter section between 22 and 26 m. The average discharge rates of pumping wells W1 and W2 in the observation period are 2.50 $m^3$ $h^{-1}$ and 3.48 $m^3$ $h^{-1}$ respectively, while the recirculation flow rate of the GCW is approximately 0.35 $m^3$ $h^{-1}$. The depth to groundwater in the treatment area is about 2 m. The discontinuous distribution of low-permeability deposits and fully screened wells contribute to the vertical hydraulic communication of the groundwater circulation hosted in the saturated complex (Fig. 3b).

At Site 2, the geological setting comprises a bedrock of calcareous origin, overlain by deposits of breccia, conglomerates, and other filling materials. The limestones underlying the stratigraphic sequence constitute an aquifer characterized by intense cracking and karstification. A range of hydraulic conductivity values between 1.1 x $10^{-8}$ m $s^{-1}$ and 8.9 x $10^{-5}$ m $s^{-1}$ are provided by 50 Lugeon tests for the carbonate aquifer. Hydraulic conductivity values from pumping testing range from 1.1 x $10^{-5}$ m $s^{-1}$ and 1.9 x $10^{-4}$ m $s^{-1}$. Ciampi et al. (2023) reconstructed a conceptual site model that identifies a source of historical arsenic contamination in the saturated zone of a calcareous aquifer. The site has been impacted by long-term pumping and groundwater treatment measures to reduce the residual arsenic masses. The pilot-scale start-up of a GCW in 2020 is accelerating the depletion processes of the aged contamination source (Ciampi et al., 2023). Focusing on the secondary source area, pumping wells W1, W2, and W3 operate massive and long-term groundwater withdrawals, with average flow rates of about 4.0 $m^3$ $h^{-1}$, 3.94 $m^3$ $h^{-1}$, and 1.04 $m^3$ $h^{-1}$ respectively. On the other hand, the GCW recirculates groundwater at a rate of about 1.9 $m^3$ $h^{-1}$ (Fig. 4a). Groundwater extraction wells are screened at depths between 6 and 69 m. The GCW is equipped with four screened sections located at depths ranging from 15 to 20 m, 22–27 m, 30–35 m, and 38–43 m. Groundwater is extracted at the middle screens and re-injected into the upper and lower filters, generating stacked circulation and superimposed cells in the fractured rock. The groundwater table stands at an elevation close to 0 m a.s.l. and is perturbed by groundwater withdrawals operated by the active pumping systems. A comparison of the piezometric level recorded when the pumps are active (dynamic condition) and shut off (static condition) reveals a hydraulic head loss in extraction wells W1, W2, and W3 of roughly 3.1, 16.9, and 13.5 m respectively (Fig. 4b). These findings suggest that deactivating the pumps at groundwater withdrawal wells can help recover hydraulic

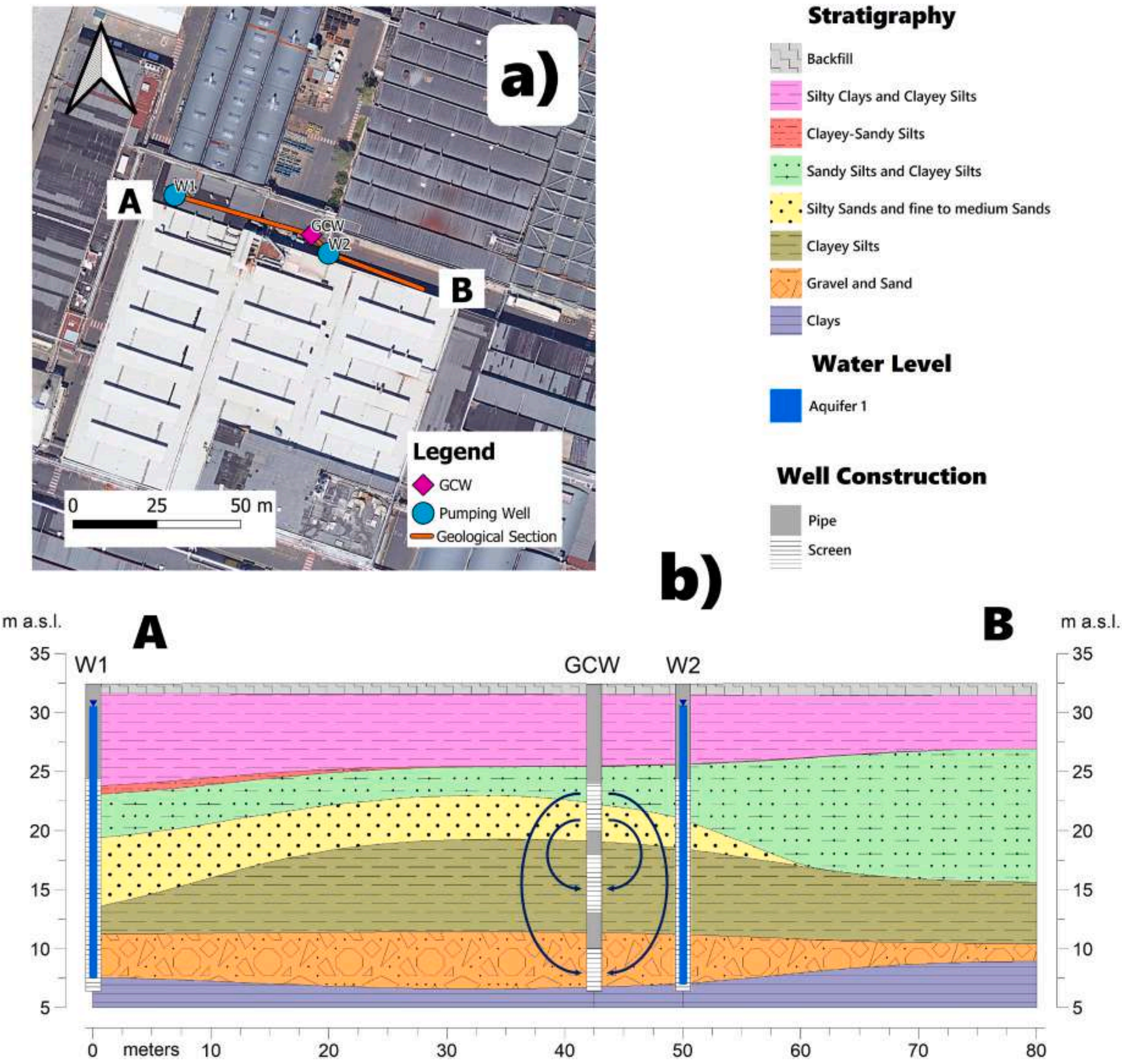

**Fig. 3.** Geological profile map illustrating the location of the groundwater extraction wells W1 and W2 and GCW at Site 1 (a). Stratigraphic section with the piezometric level, the screening sections of the pumping wells and the GCW with schematic circulation direction (b).

head, prevent aquifer desaturation at the GCW, supply flow to recirculation system cells, and improve access to residual arsenic that may be inaccessible to conventional groundwater withdrawal systems due to pumping-induced unsaturation.

### 3.2. Decontamination dynamics induced by remediation efforts at the two sites

Fig. 5a and b depict the analysis of historical concentrations of 1,2-DCE and VC collected at Site 1 for pumping wells W1 and W2. These exhibit a progressive decline in measured concentrations and thus masses discharged at groundwater withdrawal points over time. In particular, the graphs reveal rather steady-state concentrations in the later phase of operation (II), following a more rapid decline in the initial period (I).

This behavior is consistent with evidence derived from studies of P&T systems. Initially, concentration declines relatively rapidly. Then the rate of concentration reduction is slowed, showing asymptotic conditions (Brusseau and Guo, 2014; Ciampi et al., 2021b; Guo et al., 2019). Although groundwater withdrawal efforts reduce contamination appreciably, large amounts of organochlorine chemicals persist in the asymptotic stage. The average concentrations of 1,2-DCE in groundwater influent to the treatment plant in the prolonged stationary stage are 645.7 μg L$^{-1}$ (W1) and 494.0 μg L$^{-1}$ (W2). Back-diffusion phenomena from secondary pollution sources, adsorbed to low-permeability media pose a contributing factor to the persistence of the detected plume (Tatti et al., 2019). This statement is supported by the outcomes of previous studies dealing with the MCS reconstruction and the pilot-scale testing of a bioremediation technology for Site 1 (Ciampi et al., 2019a, 2021a; Petrangeli Papini et al., 2016; Pierro et al., 2017). Besides, the above works detail the combined application of GCW with the synthesis of electron donors to enhance BRD in the aquifer. The increase in VC concentrations in the steady-state phase of Fig. 5a and b match the stimulation of BRD and/or the acceleration of pollutant desorption mechanisms. These changes followed the pilot testing of the GCW in 2014, the scale-up of remediation efforts in 2019, and the reduction in withdrawal rates from extraction wells, which previously exerted hydraulic disturbances on the recirculation system in 2020.

Historical data from long-term pumping and treatment operations at Site 2 also exhibit a rapid drop in As concentration during the initial operational phase (I) and an asymptotic-stationary condition of pollutant contents throughout the subsequent stage (II) (Fig. 6a, c, d). In the period I, As concentrations in the influent to the treatment plant reach maximums of 39.2 mg L$^{-1}$ (W1), 219.0 mg L$^{-1}$ (W2), and 68.4 mg L$^{-}$

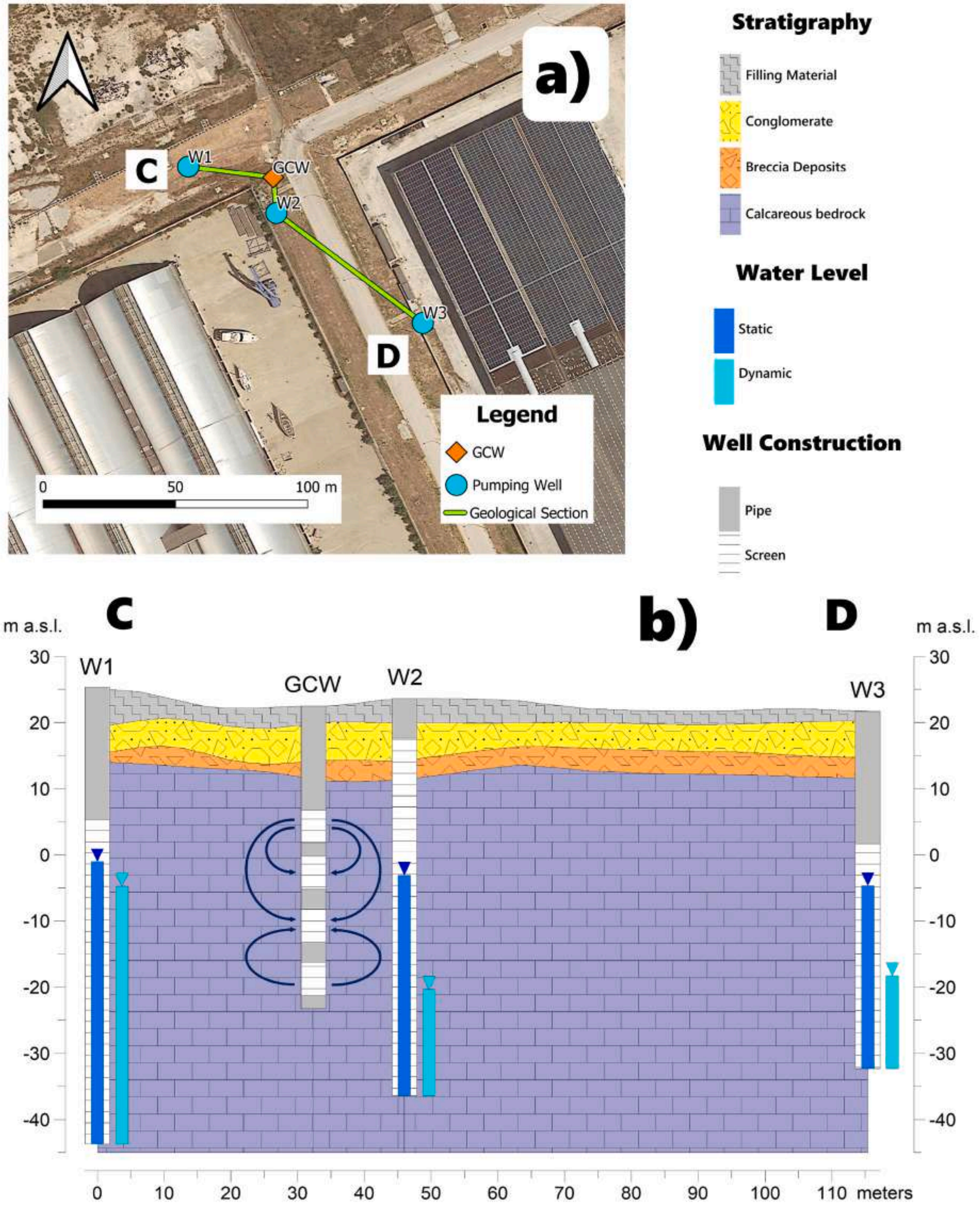

**Fig. 4.** Geological profile map showing the position of the groundwater extraction wells (W1, W2, and W3) and GCW at Site 2 (a). Stratigraphic section illustrating the piezometric level of the pumping wells under static and dynamic conditions and the schematic layout of the GCW-induced recirculation cells with vertical positioning of the screens (b).

$^{1}$ (W3). During phase II, As concentrations consolidate around average values of 2818.8 µg $L^{-1}$ (W1), 6813.6 µg $L^{-1}$ (W2), and 3429.4 µg $L^{-1}$ (W3). These findings suggest the accumulation of aqueous solutions with high concentrations of As inside the cracks and pores of the calcareous aquifer, which slowly but significantly release As into the groundwater as a result of the source mass discharge process. These hypotheses on the architecture of the pollutant are consistent with the removal behavior of withdrawal wells, the properties of the subsoil, and the results of Brusseau and Guo (2014) which investigated the mobility of a poorly accessible mass of contaminant in a bedrock. The response of the aquifer to withdrawal operations and dynamic drawdown of the groundwater table (Fig. 4b) explain why conventional extraction wells are not impacting the residual masses of As potentially stored in a pumping-induced unsaturated and fractured zone. Activation of the GCW at the pilot scale in September corresponds to a sudden increase in As concentrations in pumping well W1 (Fig. 6b). Pumping operations interfere with recirculation cells inside the GCW's radius of influence (Miller and Elmore, 2005). According to field data, the hydrodynamic

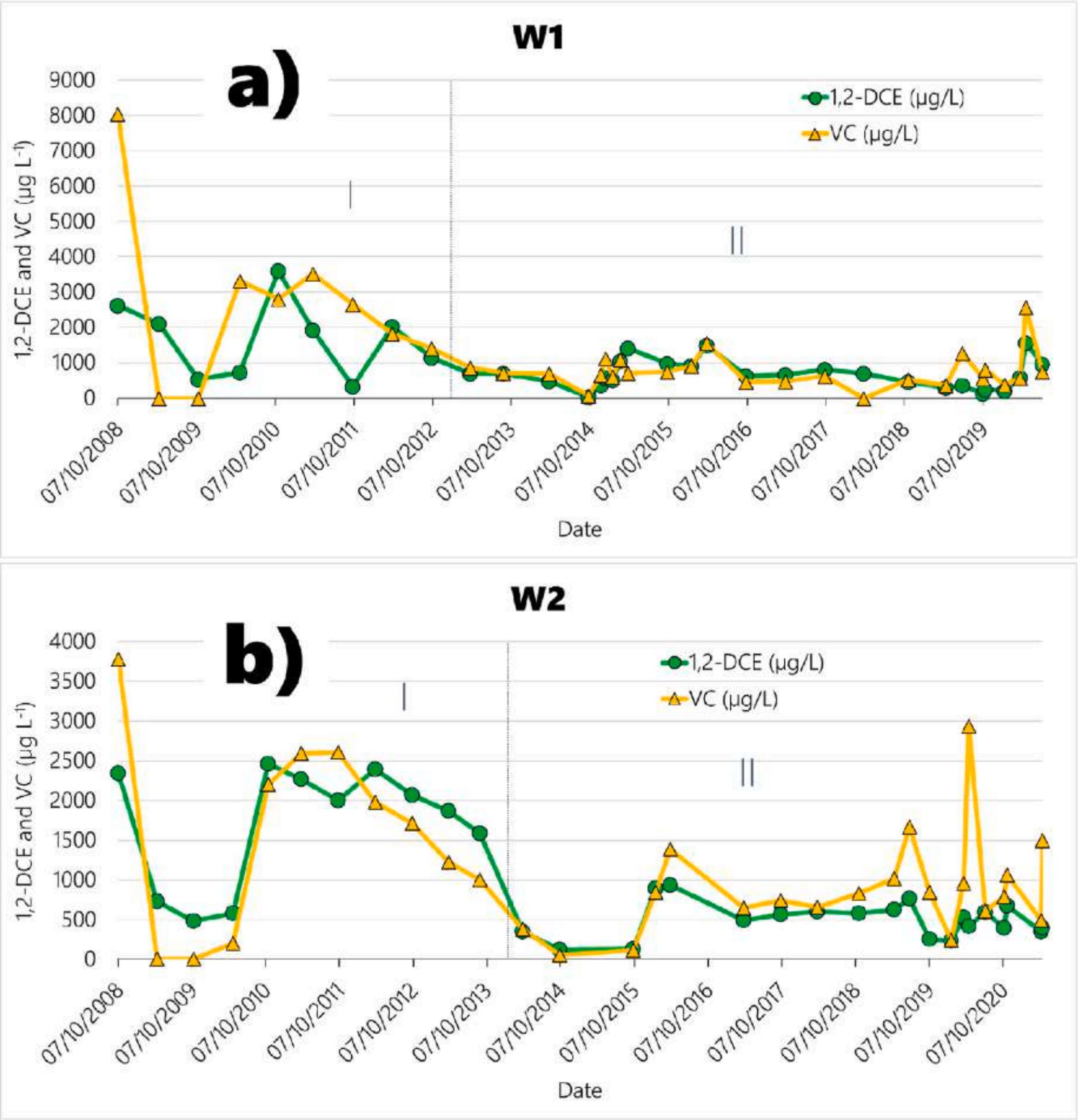

**Fig. 5.** Time trends of 1,2-DCE and VC concentrations in pumping wells W1 (a) and W2 (b) at Site 1, during the initial (I) and later stages of P&T operation (II).

overlap of pumping-induced drawdown and the development of recirculation cells may enhance the mobilization of pollutants. The recirculating system promotes the flushing of secondary source masses, mobilization of pollutants, and increases As removal by the pump-and-treat system. The GCW acts as a physical enhancer for groundwater extraction wells, attacking masses of contaminant that is poorly accessible to groundwater flushing associated with traditional withdrawal well.

Data gathered from the two sites' GCW installations emphasize the efficacy of recirculation systems over traditional pumping wells for remedying secondary sources of pollution in complex hydrogeological contexts. The influent concentration of low-chlorinated compounds (i.e., 1,2-DCE and VC) at the recirculation system reveals the marked ability to mobilize the residual fraction of pollutants adsorbed to low-permeability layers at Site 1 than conventional groundwater withdrawal systems. The average concentrations of 1,2-DCE in the GCW discharge flow in the first (I) and second (II) operational phases of the recirculation system are 9028.6 μg $L^{-1}$ and 2602.2 μg $L^{-1}$, values significantly higher than the concentration data from long-term pump-and-treat operations (Fig. 7a). The application of the recirculation system at Site 2 points to a significant acceleration of source depletion processes from As in the fractured aquifer. The GCW startup results in the mobilization of average As concentrations of 14312.5 μg $L^{-1}$ (Fig. 7b), although the recirculation rates are significantly lower than the extraction rates of the pump-and-treat system.

The mean As content mobilized by the GCW significantly exceeds the average concentrations of traditional groundwater withdrawal wells in the asymptotic-stationary stage. Such evidence suggests that forced groundwater flow induced by recirculation cell development can impact As pools in the fractured medium that happen to be excluded from 1-dimensional flushing activity generated by the P&T wells. Secondary sources are found to be otherwise inaccessible to flushing associated with extraction wells due to the water table drawdown as a result of pumping activities.

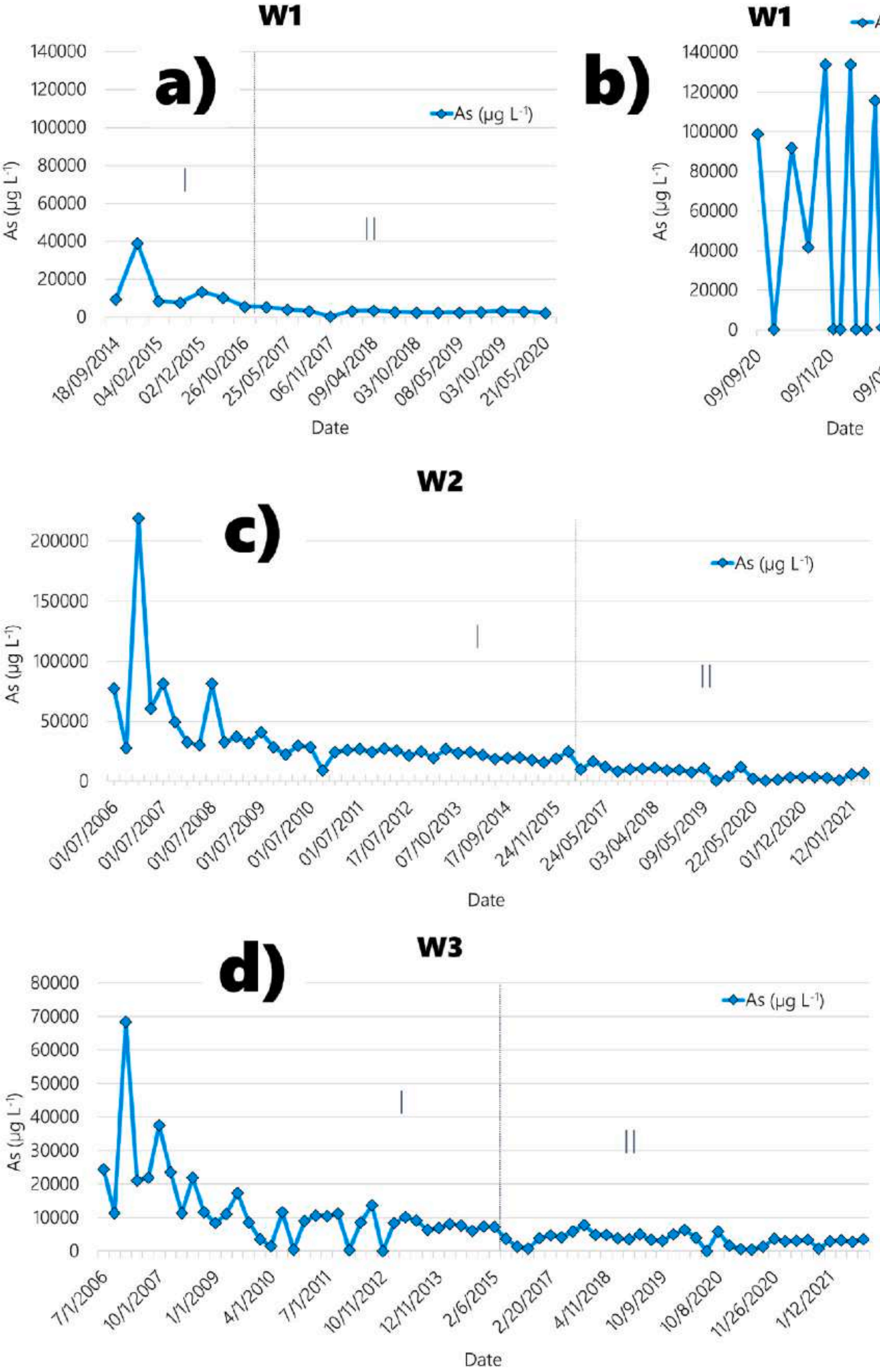

**Fig. 6.** As concentrations in the groundwater extraction wells W1 (a), W2 (c), and W3 (d) at Site 2, during the initial (I) and latter stages of activity (II). Development of the As concentration in W1 (b) during the GCW operation phase.

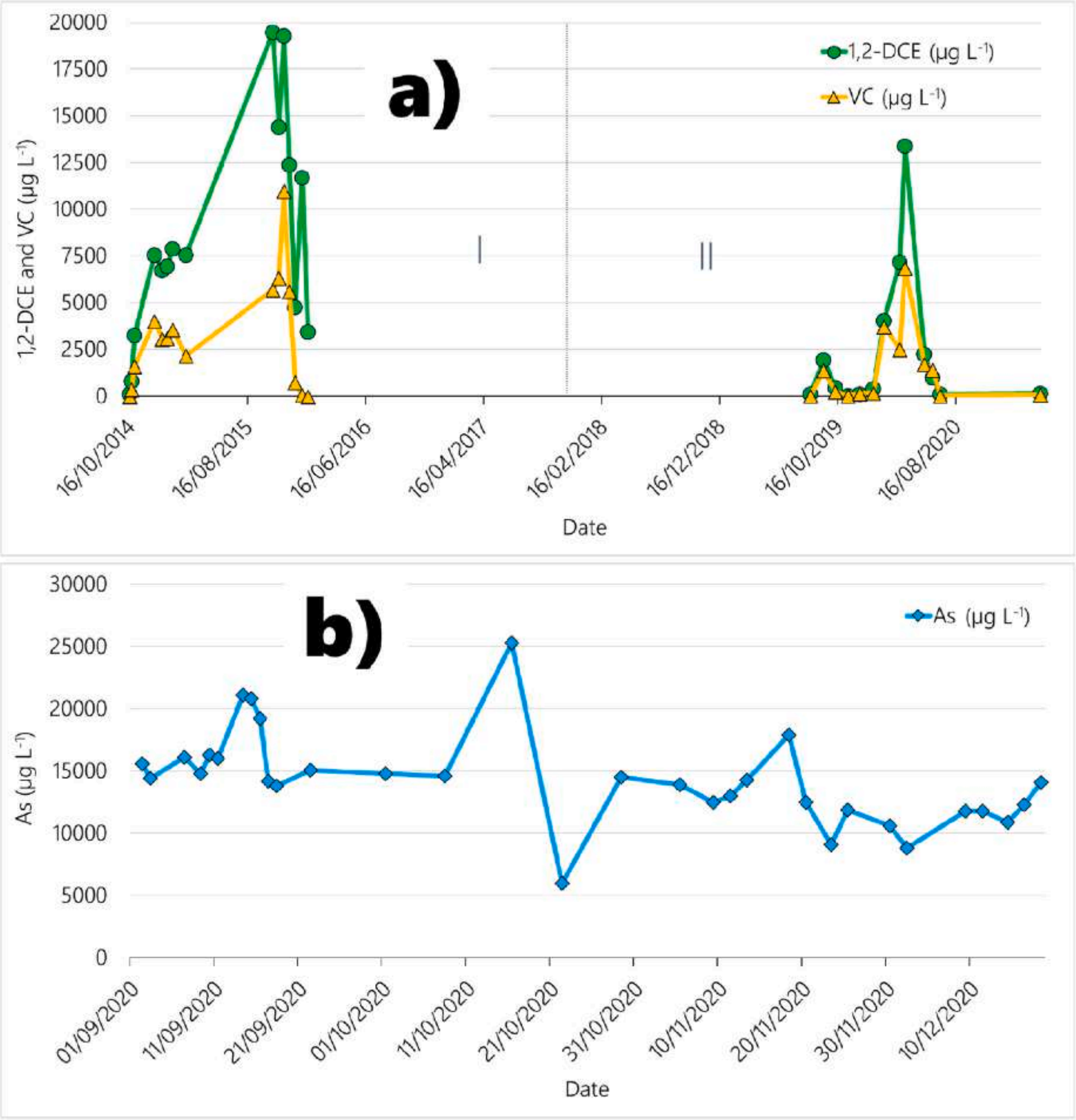

**Fig. 7.** GCW influent concentrations of 1,2-DCE and VC in the pilot (I) and full-scale (II) operational phases at Site 1 (a). Concentrations of As mobilized by the GCW at Site 2 (b).

### 3.3. *A comparative analysis of the performance and sustainability of GCWs versus conventional pumping wells (P&T)*

The findings of the remediation technique comparison analysis for Site 1 are discussed in light of the two GCW running periods (I and II), as well as the concentration decrease (I) and plateau (II) stages for groundwater extraction wells. Comparing data on the average concentration of 1,2-DCE clearly shows the capability of the recirculation systems to mobilize trapped and residual contaminants residing within the low-permeability zones. 1,2-DCE concentrations mobilized by the GCW are always significantly higher than those in the influent to the pump-and-treat system and appear even an order of magnitude greater when considering the steady-state period of conventional wells (Fig. 8a). Since groundwater extraction wells operate for a significantly longer time frame than GCW, the computation of 1,2-DCE masses removed by the P&T wells exceeds the amount mobilized by the GCW (Fig. 8b). However, the mass-based attenuation rate looks comparable for the two groundwater treatment technologies (Fig. 8c). On the other hand, the volume of water extracted from conventional withdrawal wells to flush the contaminated zone highlights the significant consumption of groundwater resource compared to recirculation systems. The GCW recirculates a total groundwater volume of 8845.2 $m^3$, while the conventional wells pump and treat water volumes of 275559.0 $m^3$ (W1) and 382354.5 $m^3$ (W2) throughout the monitored time frame (Fig. 8d). The comparison of masses extracted/treated per unit volume by a GCW and P&T system wells exemplifies the performance as well as the sustainability of recirculation systems versus traditional groundwater pumping wells in the specific hydrostratigraphic and physicochemical context. In the first phase of running, the groundwater circulation well mobilizes 10 times more pollutant mass per unit volume than standard withdrawal wells. In the second operation phase, the GCW removes a higher mass of 1.2-DCE (1.5 g $m^{-3}$) than the W1 (0.7 g $m^{-3}$) and W2 (0.5 g $m^{-3}$) extraction wells in the concentration plateau stage (8e).

The recirculation well installed in Site 2's source area mobilizes average As concentrations that are often greater than those of the pumping wells. This behavior is perhaps most evident during the steady-

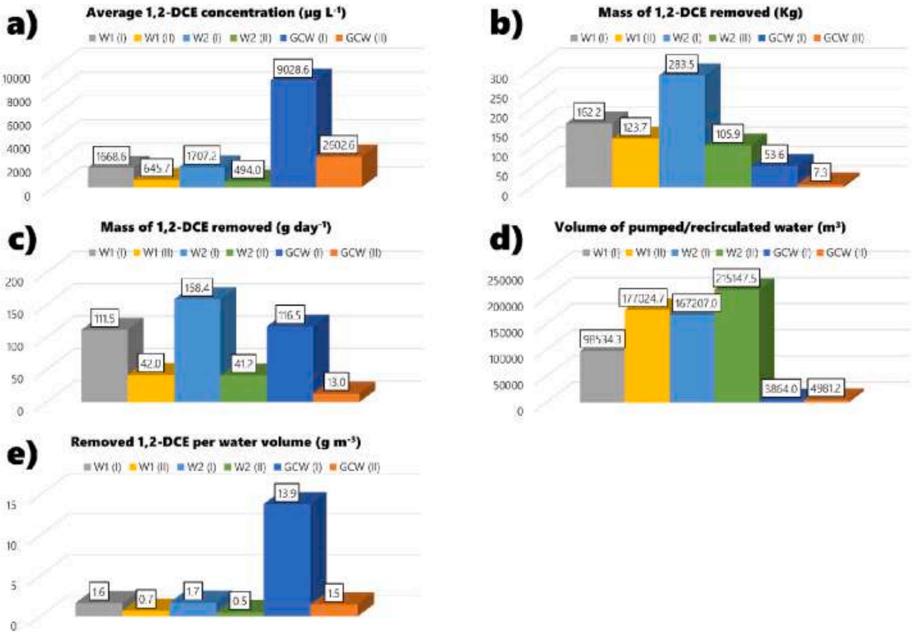

**Fig. 8.** Comparison of GCW vs pumping wells in the initial (I) and later operational phases (II) at Site 1, in terms of mobilized 1,2-DCE concentrations (a), physically removed 1,2-DCE mass discharge (b, c, e), and treated/recirculated groundwater volumes (d).

state concentration phase of extended pump-and-treat efforts. The average concentration of As removed from well W2 in the initial phase of P&T operation (36.0 mg $L^{-1}$) deviates from this general trend and exceeds the value estimated for GCW (14.3 mg $L^{-1}$) (Fig. 9a). The computation of masses removed emphasizes that the W2 pumping well removes 12345.7 kg of total As in the first stage that characterizes the trend curve of pollutant-concentration data gathered from 15 years of P&T operations (Fig. 9b). The mass-based attenuation rate stresses high As mobilization during the initial stage of P&T operation for well W2 (3.2 Kg $day^{-1}$). However, the mass of As mobilized daily by GCW is comparable to the removal rate of W2 during the plateau phase of concentrations as well as to the quantity removed daily by W1 and W3 groundwater extraction wells (Fig. 9c). The analysis of groundwater amounts drawn from pumping wells and circulated by the GCW reveals the standard P&T's significant water resource consumption. In particular, well W2 withdraws a volume that is 74 times greater than the amount of groundwater that the GCW recirculates (Fig. 9d). A comparative evaluation of masses mobilized per unit volume by the two technologies emphasizes the added value and advantages of recirculation systems over typical P&T in targeting aged As pools in the fractured aquifer. The recirculating well mobilizes a mass of As per unit volume generally higher than the wells associated with the P&T during the initial and later stages of operation. In contrast to this behavior, the computation for the concentration reduction stage of the standard W2 pumping well reveals the removal of 34.6 g As per $m^3$ of extracted water, a value higher than the mass mobilized per unit volume from the

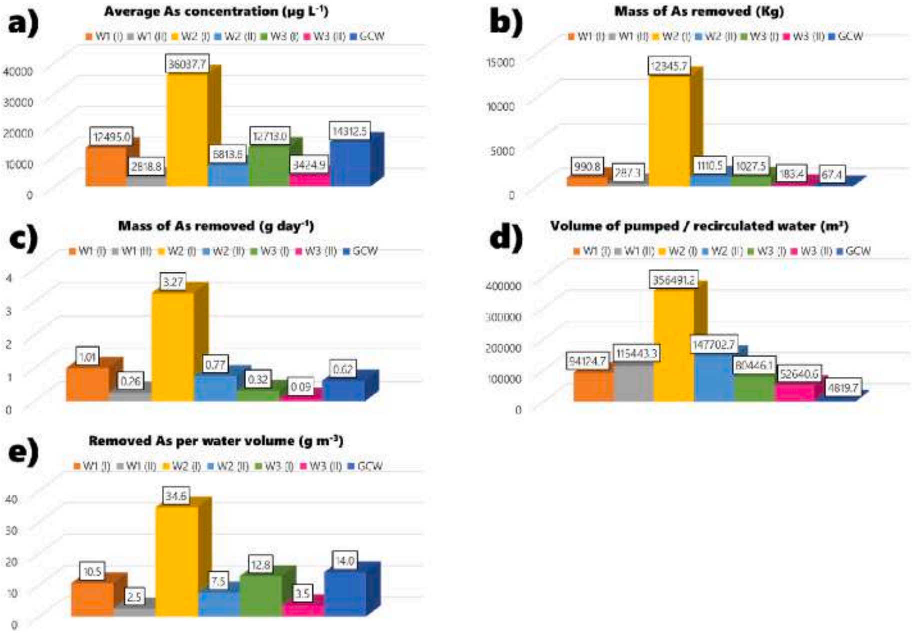

**Fig. 9.** Comparison of mobilized As concentrations (a), physically removed As mass discharge (b, c, e), and treated/recirculated groundwater volumes (d) at Site 2 by the GCW vs groundwater extraction wells during the first (I) and the latter period of activity (II).

recirculation well (14.0 g $m^{-3}$) (Fig. 9e).

High contaminant concentrations in the initial stage of W2 well P&T activities reflect the significant impact of highly contaminated and more readily accessible As pools to flushing groundwater and the induced gradient conditions associated with the initial operation of P&T systems (Brusseau and Guo, 2014; Guo et al., 2019; Truex et al., 2017). The successive drop in pollutant removal rate in the plateau stage and the aptitude in mobilizing long-term steady-state-asymptotic concentrations evince the limitations of classic groundwater extraction and containment systems to address aged sources of pollution in complex geological contexts (Rivett et al., 2006). Recirculating wells enhance mass transfer, mobilization of DNAPL and As concentrations, and removal of pollutants in both granular and fractured rock deposits by impacting secondary sources or contaminant pools accessible to conventional pump-and-treatment only via limited back-diffusion. Also, the development of the drawdown cone under dynamic conditions may make sources of contamination in a pumping-induced unsaturated zone unassailable to withdrawal wells. In this regard, evidence from hydrogeochemical modeling suggests that recirculation cells may impact contaminant pools in the geologic medium, prompting the shutdown of P&T and a revision of the remediation system. Similar to Brusseau and Guo (2014) and Truex et al. (2017), continuous CSM refinement assists in decision-making for remedy modification and evaluates P&T optimization alternatives as part of the decision logic for P&T performance evaluation. Furthermore, the long-term pump-and-treat hydrochemical data validate Langwaldt and Puhakka's (2000) hypothesis that pump-and-treat procedures can be improved by enhanced flushing induced by groundwater circulation wells. The above comparative performance and sustainability analysis reveals the diverse pollutant removal behavior that characterizes two distinct remediation approaches, highlighting the key benefit of applying GCW over more conventional remedial solutions. In contrast to pump-and-treat technologies, recirculation systems feature an inherently conservative nature, resulting in no net removal or discharge of groundwater. Such a treatment philosophy embraces the current attractive dogma of green technologies, limiting the net loss of groundwater and addressing the potential problems associated with discharging large amounts of treated groundwater to the surface (Elmore and Graff, 2002; Elmore and De Angelis, 2004).

## 4. Conclusions

Field evidence from two sites contaminated with DNAPL and As respectively indicate that GCW are a powerful alternative to reduce groundwater remediation times in porous media and in fractured rock and pave the way for more sustainable groundwater remediation in diverse geological settings. At Site 1, the increase in CAHs in the GCW influent shows a mobilization of residual contaminant levels that are not captured by conventional groundwater abstraction systems. The GCW mobilizes 13.9 g $m^{-3}$ of 1,2-DCE per unit volume of treated water during the pilot test, a mass which is 10 times higher than standard withdrawal wells in the shrinkage phase. In the full-scale implementation, the GCW removes a higher mass of 1.2-DCE (1.5 g $m^{-3}$) than the W1 (0.7 g $m^{-3}$) and W2 (0.5 g $m^{-3}$) pumping wells in the concentration plateau stage. The average concentrations of 1,2-DCE in the GCW discharge flow during the pilot and full-scale configuration of the recirculation system are 9028.6 μg $L^{-1}$ and 2602.2 μg $L^{-1}$ respectively, values significantly higher than the concentration data from long-term P&T operations. Also, the GCW recirculates a total groundwater volume of 8845.2 $m^3$, while the conventional wells pump and treat water volumes of 275559.0 $m^3$ (W1) and 382354.5 $m^3$ (W2) throughout the monitored time frame. The results also reveal that GCW application stimulates microbiological reductive dichlorination. The GCW installed at Site 2 tends to mobilize higher average concentrations and remove higher mass of As than the pumping wells, particularly during the steady-state concentration phase of extended P&T efforts. In contrast, during the initial phase of P&T operation, the average concentration of As removed from well W2 (36.0 mg $L^{-1}$) exceeds the estimated value for the GCW (14.3 mg $L^{-1}$). Also, the concentration reduction stage of the standard W2 pumping well shows the removal of 34.6 g As per $m^3$ of extracted water, which is higher than the mass mobilized per unit volume from the recirculation well (14.0 g $m^{-3}$). The mobilization of significant masses of As by P&T reflects the significant impact of easily accessible As pools in the early operational stages. The decline in the rate of pollutant removal during the steady-state concentration phase, which is a result of long-term P&T operations, highlights the limitations of traditional groundwater extraction and containment technologies in addressing aged sources of contamination. Moreover, well W2 withdraws a volume that is 74 times greater than the amount of groundwater recirculated by the GCW. The quantitative analysis, which is conducted over differing time frames for the pump-and-treat and conventional wells, reveals a significant waste of water resources in the P&T process. In conclusion, GCWs are a more sustainable and performing technology for groundwater remediation than traditional P&T methods in challenging geological environments. It's time to embrace this innovative solution and make groundwater remediation a more environmentally-friendly and high-performing process.

## Credit author statement

Paolo Ciampi: Conceptualization, Formal analysis, Software, Writing – original draft preparation. Carlo Esposito: Supervision, Visualization. Ernst Bartsch: Data curation, Investigation, Writing - Reviewing and Editing. Eduard J. Alesi: Resources, Writing - Reviewing and Editing. Marco Petrangeli Papini: Project administration, Supervision, Validation.

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## Data statement

Due to the sensitive nature of the topics discussed in this work, some limits apply to the availability of data that supports the findings. As a result, the data used for this work are not publicly accessible, however, the authors can still provide them upon reasonable request.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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