Remediation of chlorinated solvents with Electrical Resistance Heating (ERH) at an active industrial site in Italy

Contaminazione da solventi clorurati. Bonifica mediante Electrical Resistance Heating (ERH) in un sito industriale attivo in Italia

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ARTICLE INFO
Ricevuto/Received: 10 May 2023
Accettato/Accepted: 25 July 2023
Pubblicato online/Published online: 30 September 2023
Handling Editor: Daniele Pedretti

Citation: Mori, P., Baldock, J., Gigliuto, A., Cappelletti Zaffaroni, M., Marino, C. (2023). Remediation of chlorinated solvents with Electrical Resistance Heating (ERH) at an active industrial site in Italy

Acque Sotterranee - Italian Journal of Groundwater, 12(3), 41- 50
https://doi.org/10.7343/as-2023-674

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Keywords: Italy, in situ thermal treatment, electrical resistance heating, chlorinated solvents remediation.


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Abstract
Italian legislation defines stringent groundwater chemical quality criteria, to be applied at a site’s downgradient property boundary, irrespective of whether the underlying aquifer is, or could be, used for water resource purposes. In some scenarios, the regulatory authorities may identify less stringent standards, but this rarely occurs. This means that many sites with groundwater contamination are managed using hydraulic barriers, as source zone remediation may not achieve the stringent groundwater standards required due to technology limits or time constraints; therefore, the parties responsible for contamination often decide to continue to operate these hydraulic barriers indefinitely.

This article describes the first application in Italy of source treatment using Electrical Resistance Heating (ERH), a remediation technology capable of removing a large percentage of contaminant mass, at a site where a hydraulic barrier is operating within a low yielding aquifer that is not used for water supply. The implementation of this technology was possible since the source zone was far from the downgradient site boundary, thus making achievement of the stringent quality standards at the boundary possible within a reasonable timeframe. The ERH system recovered of about 600 kg of contaminants within a timeframe of 90%. This article also emphasizes that, in similar low yielding aquifers, setting less stringent groundwater standards at the site boundary whilst still protecting downgradient receptors may promote more widespread implementation of source remediation activities in Italy.
Introduction

Groundwater is a major source of fresh water for population, used for several purposes, mainly for drinking water, but also for agricultural and industrial uses (Li et al., 2021; Preziosi et al., 2022). The importance of groundwater was celebrated on 22 March 2022, which marked the occasion of the World Water Day (United Nations, 2022), titled “Groundwater, making the invisible visible”. In Italy, about 26 billion cubic meters of water are consumed annually (roughly 55% used for agriculture, 27% for industrial purposes and 18% for civil purposes) and in 2018 more than 9.2 billion cubic meters of drinking water were used, of which about 85% were derived from groundwater (Legambiente, 2022). Since groundwater contamination is a growing global issue, it is necessary to identify major challenges associated with its remediation, critically considering existing methods and understanding current research trends.

The Italian legislation for contaminated sites, in particular for groundwater (Legislative Decree 152/06, IV, Title V, Annex V, Table 2) sets stringent quality standards at the downgradient boundary of a contaminated site. These limits are equivalent to or are sometimes lower than drinking water values (Legislative Decree 18/23, Annex I), regardless of the suitability of an aquifer to be used for water supply purposes, e.g. for the contaminants Tetrachloroethene (PCE) and Trichloroethene (TCE), groundwater quality standards are 1.1 µg/L and 1.5 µg/L respectively, while the drinking water standard is 10 µg/L and represents the sum of these two solvents. According to the same Legislative Decree 152/06, Public Authorities may identify less stringent standards (designed to achieve protection of the downgradient receptors), but to the best of the authors knowledge, this has occurred only to manage background contamination (as reported for example in Lombardy Region, 2017) and not to take into account site-specific aquifer characteristics. This is different to the approach of many other Countries, where quality standards for groundwater are identified on the basis of site-specific risk assessments and aquifer characteristics; for example in Germany, Belgium, The Netherlands, United Kingdom (Bundesministerium der Justiz, 1999; Gouvernement Wallon, 2018; Rijksinstituut voor Volksgezondheid en Milieu, 2009; United Kingdom Environmental Agency, 2017).

At several Italian sites, contaminated groundwater is managed through hydraulic barriers (Pump and Treat systems). These consist of abstraction wells that remove impacted groundwater to prevent off-site migration; abstracted groundwater is then treated and often discharged into the sewer network. These systems are usually installed at the downgradient site boundary (Friuli Venezia Giulia Region, 2018; Sustainable Remediation Forum Italy, 2015; Eupolis Lombardia, 2015; Petrangeli Papini M. et al., 2013; Province of Milan, 2014). Hydraulic barriers can be effective to avoid contaminated groundwater migration but cannot remediate source zones (i.e., subsoil areas with high concentrations of adsorbed contaminants or those present as Non-Aqueous Phase Liquid), since the dissolution rate of such contaminants is generally slow, which can result in hydraulic barriers operating for decades (USEPA, 2021; Antelmi et al., 2020; Cohen et al., 1997). This is why international best practices on remediation suggest to integrate hydraulic barriers with more aggressive source treatment techniques, that can accelerate the extraction or degradation of contaminants, reducing the remediation timeframe, restoring the sites for beneficial reuse and increasing remediation resilience (USEPA, 2021; Horst et al., 2021; Eupolis Lombardia, 2015; Voudrias E.A., 2001; Federal Remediation Technologies Roundtable, 1995; USEPA, 1994). However, it must be considered that the achievement of stringent groundwater quality standards (e.g., drinking water values) at a downgradient boundary within a reasonable timeframe may not be technically achievable, despite the use of the most aggressive remedial technologies (e.g., Excavation and Disposal, In Situ Thermal Treatment (ISTT) or In Situ Chemical Oxidation/Reduction). This is particularly true at sites impacted by persistent contaminants (e.g., chlorinated solvents), those underlain by difficult-to-treat lithologies (e.g., low permeability layers), or where the contamination is present at significant depth (USEPA, 2019; Stroo et al., 2012). The risk of not achieving stringent quality standards increases when the source zone is located close to the downgradient site boundary. This may not be the case at all sites, and the implementation of remediation activities within the source may result in the achievement of remediation goals in a few years (Antelmi et al., 2021); however, according to the experience to the authors, in Italy the parties responsible for the contamination often decide to continue to operate hydraulic barriers indefinitely without implementing source treatment, given the uncertainty of regulatory acceptance.

ISTT systems achieve a high degree of remediation performance (Horst et al., 2021; USEPA 2019; USEPA 2014), using a variety of heating methods that are selected mainly based on subsurface permeability and treatment temperature required. ERH is a heating methodology that involves passing electrical current through unsaturated or saturated soil, resulting in increased subsurface temperatures of up to 110°C. The soil is heated by the passage of current through it, as induced by the electrodes. ERH is usually designed to increase the subsurface temperatures beyond the boiling point of the contaminants causing them to transition into the vapour phase and be removed under negative differential pressure through recovery wells (Gavaskar A. et al., 2007). ERH performance is not significantly affected by the presence of low-permeability layers. During heating, pore water increases in volume 1,700-fold as it is converted to steam; this has the potential to create fissures in clayey and silty soils, allowing capture of the vaporized contaminants and steam from nearby extraction wells (U.S. Army Corps of Engineers, 2014). Volatilization of contaminants from the subsoil is generally slow, which can result in hydraulic barriers operating for decades (USEPA, 2021; Antelmi et al., 2020; Cohen et al., 1997).
is 88°C, while boiling points of PCE and water are 121°C and 100°C, respectively (USEPA, 2016). The ERH technology is paired with extraction systems that consist of wells removing vapours, and liquids if needed, from the subsurface. The recovered vapour and liquid steams would typically be treated to remove contaminants prior to discharge (USEPA, 1999).

This article describes the first (to the best of the authors knowledge) application in Italy of ERH, at a site where a hydraulic barrier is operating within a low yielding aquifer. The implementation of this technology was possible since the contamination source (consisting of approximately 5,000 m³ of subsoil impacted by chlorinated solvents) was far from the downgradient site boundary, thus making the achievement of quality standards possible within a reasonable timeframe. Due to the legislation described previously, if the contaminated area had been closer to the Site boundary, source remediation activities may have not been implemented and the benefits in terms of rapid and efficient removal of the contaminant mass from subsoil using ERH would not have been achieved.

This article also emphasizes that setting site-specific and less stringent groundwater quality standards at the downgradient site boundary, when technically possible, would increase the possibility to achieve such standards and therefore increase the likelihood of using ISTT or other source remediation approaches at additional sites in Italy.

**Materials and methods**

**Site Description**

In the early 2000’s, an environmental investigation carried out at an active industrial site in Northern Italy by the property owner detected the presence of chlorinated solvents in soil and groundwater. The main contaminants were PCE and its daughter products (TCE, 1,2-dichloroethene and vinyl chloride) with concentrations of up to circa 120 mg/kg of PCE in soils and 20,000 µg/L in groundwater detected. As a first management measure, a Hydraulic Containment System comprising 6 pumping wells was installed downgradient of the most impacted area to prevent off site contaminant migration. Given the low yield of the aquifer, pneumatic pumps were installed. In the years following installation, additional characterization activities were carried out in order to further delineate in 3D the impacted contamination sources, using specific High Resolution Site Characterization techniques that included the Membrane Interface Probe technology (CLU-IN, 2021; Geoprobe systems, 2021; Dijkshoorn et al., 2014; Heron et al., 2009; Sale et al., 2008; Griffin et al., 2007; USEPA, 2004 & 2005). The underlying lithology can be summarized as follow:

- 0.0 - 0.2/0.4 m below ground level (bgl): concrete slab, present beneath most of the site;
- 0.2/0.4 – 1.0/1.5 m bgl: unsaturated fill material (sand and gravel or silt/silty sand);
- 1.0/1.5 – 2.0/2.5 m bgl: unsaturated silt;
- 2.0/2.5 – 3.0/4.0 m bgl: fine sand (confined aquifer);
- 3.0/4.0 – 13 m bgl (maximum investigated depth): clay, with layers of sandy clay.

These data confirmed that the aquifer thickness is very limited, in the order of about 1 m, correlating with limited abstraction rates due to low transmissivity (maximum abstraction rates were in the order of 0.1 m³/hour at each well). For comparison purposes, the Italian Ministerial Decree 260/2010 (Technical criteria for the classification of the status of groundwater bodies) sets the minimum average abstraction rate to define an “aquifer”, as 10 m³/d (0.416 m³/hour).

Overlapping the characterization data with the reconstructed lithologic cross-sections allowed the identification of the most...
impacted lithologies, as shown in Figure 1. Soil and ‘grab’ groundwater samples were also collected and analysed for Chlorinated Hydrocarbons (CHC). Analysis of these samples detected concentrations of up to 570 mg/kg in soils (as a sum of PCE, TCE, 1,2-dichloroethene and vinyl chloride) and 180,000 µg/L in groundwater, to a maximum depth of 7 m bgl, as shown in Table 1. Most of the contamination was located within low-permeability layers (clays), acting as continuing sources of contaminants for the aquifer through back-diffusion (Chapman et al., 2005).

<table>
<thead>
<tr>
<th>Contaminant</th>
<th>Maximum concentration in the most impacted pre-remediation groundwater sample (µg/L)</th>
<th>Maximum concentration in the most impacted pre-remediation soil sample - vadose zone (mg/kg)</th>
<th>Maximum concentration in the most impacted pre-remediation soil sample - clays below the aquifer (mg/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tetrachloroethene</td>
<td>73,000</td>
<td>125</td>
<td>560</td>
</tr>
<tr>
<td>Trichloroethene</td>
<td>17,200</td>
<td>1</td>
<td>10</td>
</tr>
<tr>
<td>1,2-dichloroethene</td>
<td>79,370</td>
<td>9</td>
<td>3</td>
</tr>
<tr>
<td>Vinyl chloride</td>
<td>7,500</td>
<td>1</td>
<td>not detected</td>
</tr>
<tr>
<td>Total</td>
<td>177,070</td>
<td>137</td>
<td>573</td>
</tr>
</tbody>
</table>

Figure 2 shows the trend in concentrations over time of the total chlorinated compounds in groundwater, at a pumping well in the vicinity of the most contaminated area, where data were available from 2006 until the period before the remediation. A decrease in concentration over time can be observed, but high dissolved concentrations were still present in 2019.

Selection of the remediation technology

Traditional remediation techniques were assessed as being unable to remove/degrade the contaminant mass effectively, given the limited radius of influence of injection or extraction-based technologies at ambient temperatures that could be achieved in low permeability formations (USEPA, 2019; Newell et al., 2012). Excavation and soil mixing were also excluded since part of the source zone was located beneath an operational production building and associated exterior Loading Bay area. Therefore, ISTT was considered the best remedial technology to achieve a significant reduction of the contaminant mass in the short-medium term, minimizing the impacts on the site production activities. ERH was selected as the heating methodology at this site, due to the presence of low permeability deposits. A target treatment temperature of 88°C, corresponding to the boiling point of a PCE/water mixture (USEPA, 2016; U.S. Army Corps of Engineers, 2014), was set as the minimum target temperature for the volatilization of contaminants from the subsoil.

Geotechnical evaluations

When the temperature of soil changes by 80-100 ºC, significant thermo-mechanical interactions occur. These interactions can result in thermally reversible or irreversible strains in soil (especially if they are fine-grained), which may lead to foundation settlements (USEPA, 2016; Towhata et al., 1993).

Oedometric tests using undisturbed fine-grained soil samples collected from the Site were conducted during the remediation design phase at ambient temperatures and at 80°C, in order to verify that heating would not result in significant changes in the geotechnical properties of fine-grained material. The results showed a limited increase in deformation (up to about 0.1 mm) following the application of different loads to the samples, as shown in Figure 3.

Overall the risk of subsidence was considered low, since: 1) the observed increase in deformation at 80°C was limited; 2) the presence of a sandy layer overlying the saturated clays was expected to mitigate any thermo-induced volumetric variations; 3) only one case of significant subsidence at sites remediated using ISTT was reported by the USEPA (USEPA 2016), when clays had been removed and then used again for
backfilling, which was not the case at the site described in this article. However, the installation of a monitoring system on the building above the treatment area was planned, in order to confirm the absence of deformation in the building structure during soil heating. The monitoring plan included the continuous measurement of the inclination of selected pillars and the relative displacement between pillars and external panelling, by means of 6 biaxial clinometers and 5 pairs of crack gauges communicating via wireless connection to a data acquisition unit. The results of the geotechnical tests were included in the Remediation Plan, that was approved by the Authorities.

**Remediation system layout**

The installed ERH remediation system consisted of the following below and above ground infrastructure:

- 38 co-located heating/vapour extraction points, within an area of 800m\(^2\). The maximum heating depth ranged from 4 to 7 m bgl;
- 19 horizontal screened wells, for a total length of about 95 m. These were installed within trenches at about 0.5 m bgl, to increase the recovery of vapours;
- 5 Multi-Phase Extraction (MPE) wells screened from 1 to 7 m bgl, mainly aimed at extracting liquids, in order to maintain hydraulic control within the treated area;
- 6 thermocouples to collect subsurface temperature data, at multiple depths, during the heating process;
- An MPE plant for the extraction and treatment of hot fluids and vapours;
- Electrical cabins and dedicated equipment for voltage transformations and power delivery to the MPE system and the electrodes;
- A network of underground and above ground pipelines to connect the different elements of the remediation system.

Figure 4 shows the layout of the ERH remediation system. Data from the well field were monitored remotely in real time using a dedicated webpage to tabulate or visualise the information obtained.

**Operation & Maintenance of the remediation system**

Heating started on October 12\(^{th}\), 2020, and ended on May 6\(^{th}\), 2021 (206 days). MPE recovery was continued until June 2021 to recover residual contaminated fluids from the subsurface.

During the remedial activities, many parameters were periodically measured to monitor the progress of the remediation, the most important being 1) temperature of the heated subsoil, 2) extracted liquid and vapour flow rates, 3) concentration of contaminants in the extracted liquids and vapours (before and after treatment), 4) negative differential
pressure and flow rates at each extraction well, 5) power and potable water quantities delivered to each electrode, and 6) the abstracted groundwater flow rate.

After 6 months of continuous soil heating, in April 2021 an interim sampling and analysis campaign was performed for initial validation assessment of soil and groundwater concentrations, to determine remediation performance within the heated area. In the same area, in July 2021 (following shutdown of the remediation system) a regulatory approved validation campaign was also completed and comprised additional soil (6 samples at 4 locations) and groundwater (8 samples at 6 locations) collection with the local Environmental Protection Agency.

**Results**

During the treatment period, the target temperature was reached and exceeded in almost all parts of the treatment zone (temperatures up to 105°C were achieved in the most impacted area), with the only exception being a limited area below the building (due to an installed electrode configuration being restricted for site operational reasons). An example of the modelled temperature distribution after heating interruption, at a depth of 2.5 m bgl, is shown in Figure 5. The average subsoil temperature within the heated area increased until March 2021 and then stabilized, as shown in Figure 6.

![Fig. 5 - Temperature distribution at 2.5 m bgl (May 2021).](image)

**Fig. 5 - Distribuzione della temperatura a 2,5 m da p.c. (maggio 2021).**

![Fig. 6 - Average subsoil temperature.](image)

**Fig. 6 - Temperatura media del sottosuolo.**
Figure 7 shows the daily contaminant removal rates and the cumulative mass removal trend. These values were calculated based on the concentrations of contaminants and flow rates of the extracted process streams (vapours and liquids). Daily contaminant recovery rates increased until March 2021 (when the peak of the average subsoil temperature was reached). Contaminant recovery then decreased despite stable temperature values, indicating that contaminant removal from the subsurface was complete to the extent practical. The heating system was shut-down once the daily extraction rate declined to about 10% of the maximum historical values.

Approximately 600 kg of contaminant mass was recovered between October 2020 and June 2021.

The post-ERH treatment results (spring-summer 2021) showed a significant reduction in chlorinated solvent concentrations in both soils and groundwater, where the target temperature had been achieved, as follows:

- **Unsaturated soils**: Maximum pre-remediation concentrations of 137 mg/kg had been detected. Post ERH a maximum concentration of approximately 4 mg/kg was recorded, representing a two order of magnitude reduction following thermal application;

- **Saturated Zone (Soil)**: Two pre-remediation saturated clay samples detected solvent concentrations of approximately 100 and 570 mg/kg. Post ERH application solvent concentrations of <1 mg/kg were recorded in samples collected nearby. During the 2021 sampling rounds, 9 clay samples were collected and analyzed. Eight out of nine of these samples confirmed compliance with the contamination threshold values for industrial use of the site;

- **Saturated Zone (Groundwater)**: Maximum pre-remediation concentrations of approximately 180,000 µg/L had been detected. Post ERH, maximum concentrations of approximately 11,500 µg/L were identified.

In the limited part of the treatment area where the target temperature was not achieved by some 10-20%, residual groundwater concentrations of approximately 6,000 µg/L were detected.

Soil and groundwater samples were collected during April to July 2021 at 7 newly installed performance monitoring wells (IS1 to IS7 – see Figure 8). Data recorded from samples collected from these locations were compared to baseline data obtained from the nearest monitoring points that were present prior to remediation. A decrease in contaminant concentrations of by 87 to 94% was measured in the most impacted area (exterior loading bay area), where the highest temperatures were reached in the subsoil. The contaminant removal efficiency was lower in samples collected beneath the operational building (16 to 49%), where lower temperatures were achieved in subsoil. These data confirm that the efficiency of the remediation was heavily influenced by the subsoil temperature achieved.
Discussion

The results obtained confirm the capability of ERH to significantly reduce the concentrations of chlorinated solvents in soil and groundwater, including contamination residing in low-permeability layers. The observed reduction of solvents in clays is expected to reduce the potential for back-diffusion of contaminants into the overlying aquifer (Horst et al., 2021; U.S. Army Corps of Engineers, 2014; Gavaskar et al., 2007). An additional benefit of ERH is that it is a rapid technique, meaning source treatment can lead to the achievement of remedial targets within a short timeframe (Federal Remediation Technologies Roundtable, 2022; Horst et al., 2021); as confirmed by the described application of ERH, which allowed extraction of roughly 600 kg of chlorinated solvents in 8 months. Where these remain, further reduction of dissolved phase contaminant concentrations in the aquifer is expected in the future, due to natural attenuation, which will likely have been enhanced by the thermal treatment process (Nelson, et. al., 2019; Baldock, et. al., 2015). It is noted that ERH costs per unit volume are higher than those associated with more traditional remedial technologies; which is one reason why ERH is mostly applied at sites such as this one, where the contamination source was well delineated, high in magnitude and/or other techniques would have been ineffective at removing contamination within low permeability geology (USEPA, 2016; Stroo et al., 2012).

This work confirms that ERH represents a viable technology to remediate sites where low-permeability soils are impacted by contaminants and where the effective implementation of traditional in-situ technologies is challenging. However, according to experience of the authors, the Italian generic groundwater standards at the downgradient site boundary limit the implementation of ERH (or other aggressive remediation technologies), given the considerable investments needed and the high uncertainty in achieving these standards in the short/medium term, even with aggressive source remediation technologies that are able to reduce contaminant concentrations by several orders of magnitude. Instead, operating hydraulic barriers for an indefinite period of time still represents a more common choice for the management of groundwater contamination plumes (Friuli Venezia Giulia Region, 2018; Sustainable Remediation Forum Italy, 2015; Eupolis Lombardia, 2015; Petrangeli Papini M. et al., 2013; Province of Milan, 2004). According to the Legislative Decree 152/06, Italian Public Authorities could set site-specific and less stringent groundwater quality standards, but this is rarely done. For example, this may be the case when an “aquifer” is characterized by low abstraction rates due to its hydrogeological properties, preventing its use for drinking water purposes independent of its chemical status, and acceptable concentrations for (existing or future) downgradient receptors could be calculated by means of standard risk assessment procedures. This would make the achievement of remediation goals more certain and still protect downgradient receptors, thus promoting source remediation interventions, with the related benefits in terms of accelerated extraction/degradation of contaminants from subsoil, reduced remediation timeframe, site restoration to beneficial reuse, and increased remediation resiliency (Horst et al., 2021).

Following an ISTT source zone remediation, monitored natural attenuation can be applied to further reduce contaminant concentrations and biodegradation can be fostered by the moderate/high temperature remaining in subsoil after the completion of the remediation activities (Federal Remediation Technologies Roundtable, 2022). At this site, groundwater temperatures between 30 and 40 °C were recorded 1 year after the completion of the remediation within the treated area (background results showed groundwater temperatures in the order of 20 °C), creating favourable conditions for the growth of microbial populations and accelerating the biodegradation process.

Conclusions

Application of ERH resulted in a significant decrease in chlorinated solvent mass in both low permeability soils and within groundwater, with approximately 600 kg of contaminants recovered in 8 months. Given the reduction of impacts in the clays and the presence of natural attenuation processes, a further decline in contaminant concentrations in the aquifer is expected in the future, both within the treated and downgradient areas. The thermal remediation activities will also result in cessation of the hydraulic barrier operations much earlier than could be expected without source treatment.

From an Italian legislation perspective, the implementation of ERH was possible at this site since the source zone was far from the downgradient site boundary, thus making the achievement of stringent groundwater quality standards possible within a reasonable timeframe. In similar low yielding aquifers, setting less stringent groundwater quality objectives at the site boundary whilst still protecting downgradient receptors may promote the more widespread implementation of source zone remediation activities at other sites in Italy. Also, minimizing the operational period of the hydraulic barriers may result in an increase of the lifecycle sustainability of the remediation process, reducing the overall use of resources and costs.
Acknowledgment:
The work completed relates to a remediation project procured by ERM's client (identity needs to remain confidential). The authors acknowledge the project equipment suppliers and operators McMillan-McGee (McMillan-McGee Corporation, 4895-35B Street, SE Calgary, Alberta, T2B 3M9, Canada, https://www.mcmillan-mcgee.com/) and Geostream Group (Via Zire, Magnano in Riviera (UD), 33010, Italy, https://www.geostreamgroup.com/).

Funding source
This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors. The authors declare that data, associated metadata, and calculation tools are available through an online repository or supplemental files, available from the authors.

Competing interest
All authors, the corresponding author states that there is no conflict of interest.

Author contributions
Piero Mori: Conceived and designed the experiments; Analysed and interpreted the data. James Baldock: Conceived and designed the experiments; Analysed and interpreted the data. Andrea Gigliuto: Conceived and designed the experiments; Performed the experiments; Analysed and interpreted the data. Mattia Cappelletti Zaffaroni: Conceived and designed the experiments; Performed the experiments; Analysed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper. Cecilia Marino: Analysed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper. Andrea Gigliuto: Performed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper. Antonio Cappelletti Zaffaroni: Performed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper. All authors have read and agreed to the final version of the manuscript.

Additional information
Supplementary information is available for this paper at https://doi.org/10.7343/as-2023-674
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Associazione Acque Sotterranee remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

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