Can electrochemistry help in the treatment of soils polluted with volatile organic compounds?

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Abstract

Treatment of polluted soil is one of the priorities in the search of a more sustainable planet. Electrochemically assisted soil remediation has been considered a good option for removing organic contaminants contained in soil, including the removal of volatile organic compounds, associated to gaseous streams produced during the treatment. Also, recently, electrochemical gas treatment technologies have been appointed as promising for the treatment of volatile organic compounds. In this work, we review the current opinion about the most recent studies in both areas. The first section focuses on the production of gaseous compounds during soil remediation by conventional and electrochemical systems. The second section describes the recent progress in the integration of adsorption and absorption with electrochemical processes. Finally, we discuss the holistic application of assisted electrochemical technologies in the soil remediation considering also emerging processes recently published in the literature.
Keywords: Soil remediation, Gas treatment, Electrochemically-assisted technology, electro-adsorption-desorption, adsorbent.

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**Introduction**

In the recent years, electrochemically assisted soil remediation (EASR) technology has become promising for removing contaminants from soil. In EASR technology, an electric field is applied across contaminated soil by using electrodes appropriately located in the subsurface, promoting different physical, chemical, and electrochemical phenomena. Some of the most relevant are related to the transport of species throughout the soil and they are included in the concept of electrokinetic (EK) treatments: electroosmotic (transport of water, organic compounds, inorganic compounds and oxidants species), electromigration (transport of ions), electrophoresis (transport of charged particles of colloidal size). Others can promote indirectly transport processes but are of different nature: electrolysis (electrochemical processes happening on the surfaces of the anodes and cathodes) and electro-heating (thermal processes associated to the electric and ionic resistance of the soil) [1–3]. Thus, the electrolysis promotes the generation of protons and hydroxyl ions (among other species) that, inside the electric field generated among the electrodes, are transported throughout the soil and lead to the appearance of acid and basic fronts (related to electromigration). On the other hand, the electro-heating is especially important because it leads to changes in soil temperature, which may have an impact on the transport of species by EK processes (e.g. temperature may affect to viscosity and solubility) and also in the generation of gases, with their corresponding enhanced transport throughout the soil (not really an EK process but rather important to be considered). This heating can also affect other processes happening in the soil, such as the biological processes, promoting or preventing their action and, because of the important number of potential interactions, it is important to consider their potentially huge input in the overall efficiency of the remediation process. The magnitude of the electric field applied in the EASR processes
is important and it is contributed by 1) the anodic and cathodic cell potentials and overpotentials, which are required to carry out the electrode reactions from the thermodynamic and kinetic viewpoints, respectively, and 2) the transport of charged species throughout the soil, which in turn is related to the resistance/conductivity of the soil and, because of the Joule–Thompson effect, to the heating of soil, which is more notably observed in the vicinity of the electrodes [4]. The resistance depends on many factors such as water content of the soil and composition of the soil and pore water, being the heating effect proportional to the resistance and to the square of the current intensity passed between anodes and cathodes. It is important to highlight that the rate of removal of contaminants from the soil also depends on this electrical current intensity which, in addition, also fix the rate of transport of species.

Additionally, the electrical heating can promote the generation of gaseous effluents, being in some cases the evaporation of contaminants the primary mechanism of removal, mainly in pilot and prototype systems.

The purpose of this review is to present the state of the art in the gaseous flows generated in electrochemically assisted soil thermal remediation, paying attention to the fundamentals of the technology together with the phenomena that occur in the soil, which are the basis of its effectiveness. Aspects related to the gas treatment by electrochemical technologies, as well as its applicability to volatile contaminants, drawn from contribution published in different level (lab-scale, pilot scale and prototype scale) will be also discussed.

**Production of gaseous effluent during soil remediation processes**

Over the past decades, many technologies have been applied to the recovery of soils contaminated with volatile and semi-volatile organic compounds (VOCs and SVOCs,
respectively) by heating the soil[5–9]. To heat up soil, different types of heating have been tested including conductive, convective, resistive, radio frequency or microwave heating. They differ in dominant heating interaction mechanisms.

Application of these technologies is known to be very important, because in case of a non-suitable management, these pollutants can easily be spread in the environment, far away from origin location, contributing to diffuse pollution and threaten not only water resources but also human health and viability of ecosystems. Among these treatment technologies, thermal desorption technologies are worth to mention, because the heating of the soil, and the subsequent elevated temperature reached, weaken sorption bonds and, consequently, improve contaminant mobility. In general, the Henry’s law is used in the case of very dilute systems, which define the maximum amount of the volatile species aqueous concentration that would produce the release of gas bubbles from the system. According the partial pressure is possible to obtain the equilibrium expression to the aqueous components[10,11].

Table 1 shows relevant works regarding soil remediation technologies, at lab scale and pre-pilot scale, in which gaseous flows are produced during the treatment under selected conditions. Among these technologies, one of the most promising is the microwave heating. Its volumetric character allows to directly heat different contaminated materials usually 150–600°C with significantly lower energy loss as compared with conductive heating techniques. A good example is shown in the work of Luo et al. [5] in which microwave-assisted low-temperature remediation of petroleum-contaminated soil was investigated for oil recovery. These authors obtained a high removal efficiency (99%) at 300 °C, and the oil was recovered with a dominant C11–C30 composition. They also pointed out the good performance of this technology regarding soil toxicity evaluating the germination of clover seeds in soil remediated at 250 °C. However, this technology
has limited applicability, associated with the length of the conductor rods, that may be insufficient in case of deeper water layers, which could absorb microwave energy, consequently needing for higher energy exposure[6].

On the other hand, and although the electric heating of soil is known for a long time, more recently, several studies have demonstrated that the application of the electric field during EASR processes also can favor the heating of the soil and, thus, the transport of pollutant to gaseous phase together with the mobilization of pollutant in liquid phase[7,12].

To increase the soil temperature and promote the evaporation of VOCs, electrical resistance heating (ERH) has been studied as an in-situ thermal treatment for soil and groundwater remediation. Thus, ERH was investigated by Munholland et al. for the remediation of soils contaminated by a dense non-aqueous phase liquid (DNAPL) on lab scale [7]. The gas production and migration of DNAPL was investigated using titanium coated with a metal oxide mixture (IrO$_2$-Ta$_2$O$_5$), and two velocities of groundwater (1.9 and 10.9 m day$^{-1}$). More than 114 L of gas was produced, while no DNAPL was detected in the treatment zone. The transport of the produced gas was associated with the DNAPL-water co-boiling controlled by capillary barrier, where the gas outside the heated sites resulted in condensation of DNAPL. According to the authors, the main advantage of this technology is that, regardless of lithology, it is beneficial over conventional in-situ technologies that are dependent on advective flow.

The efficiency of electrical ERH was also studied under passive venting conditions in order to promote the removal of TCE at lab scale [12]. The gas produced during soil remediation was accumulated into a fine-grained capillary barrier that contributed to the removal aqueous and gas phase TCE, following co-boiling as a result of subsequent boiling of water.
More recently, it has been reported that the application of electrochemically assisted technologies to remediate soil can lead to significant increases in soil temperature and the subsequent release of volatile and semi-volatile pollutants that must be treated. Lopez-Vizcaíno et al. [13] investigated the applicability of EK technology for the removal of pesticides (2,4-dichlorophenoxyacetic acid and oxyfluorfen) from lab scale to prototype: 1 L (lab scale), 175 L (bench scale) and 32 m$^3$ (prototype) and demonstrated the role of the controlling mechanisms in each scale. Although the mechanisms of transport were the same in the different scales, the controlling mechanisms changed with the size of the testing setup. Thus, the higher was the size, the lower was the magnitude of the electrokinetic (EK) processes and the higher was the relevance of the volatilization. EK transport mechanisms (electroosmosis, electromigration and electrophoresis) were the main responsible for the removal of pesticide in lab-scale test, while volatilization was the main responsible of the removal of pesticide at prototype scale. This outcome was related to the higher temperature reached in the larger scale facilities, which in turn is related to the higher ohmic losses in energy that lead to the heating of the soil. Therefore, the results from small-scale tests cannot be extrapolated to large-scale applications because the control mechanisms can vary. Figure 1 shows, as an example the temperature profiles obtained during an EK test with a prototype of 32 m$^3$ during one month of operation, where it can be seen the relevant increase of temperature with time and the non-uniform distribution of temperatures. These huge variations of temperature are not observed in any case when carrying out test at lab or bench scales, where increases of temperature rarely overpass 5ºC during the complete remediation tests because of the much lower ohmic resistances, which in turn are associated to the higher proximity of the electrodes.
Fig. 1. Changes in the average temperature of soil and electrolyte wells during the EK tests and 2-D maps of temperature in soil for four representative times. Obtained with permission from Elsevier [13]

Significance of the volatilization during EK processes have been reported in many other studies in which EK processes are evaluated together with other technologies[14–20]. Thus, in a work focused on the removal of trichloroethene (TCE) from spiked soil (clay and sand) coupling EK with bioremediation (EK-bioremediation) at lab scale [14], it was stated the significance of the gaseous emissions during the treatment. This was also confirmed in a different work, focused on EK-bioremediation of pesticides [15]. Also in works in which EK is coupled with permeable reactive barriers consisting of ZVI or granular activated carbon (GAC) beds [16], in which it was also found the huge relevance of the volatilization processes. However, as already commented, the conclusions drawn in a laboratory or bench-scale (where the electrokinetic processes control the rate) are poorly reproduced or cannot be easily extrapolated to the full scale (where the ohmic and heating effects can dominate).
Furthermore, taking into account the edaphology (influence of soil on organisms) the increase in temperature results in lowering the oxidation-reduction potential in water, thereby affecting redox reactions. In general, EASR technology integrate, as bioremediation show decreases the water loses by evaporation to a significant extent [2].
## Table 1. Production of gaseous streams in soil remediation processes

<table>
<thead>
<tr>
<th>Different level</th>
<th>Pollutant / type of soil / concentration / Technology</th>
<th>Technology Readiness Level / work objective</th>
<th>Operation conditions</th>
<th>Maximum efficiency reported/related remarks</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lab Scale</td>
<td>Petroleum / 95300 mg kg⁻¹ / real soil / Low-temperature Microwave</td>
<td>4 / investigate the low-temperature microwave-assisted remediation of petroleum-contaminated soil with simultaneous oil recovery at bench scale.</td>
<td>Temperature: 80–300 °C; time: 20 min; moisture content: 6%.</td>
<td>99% of mass removal; temperature: 300 °C / The germination rate of clover seeds reached the maximum using 250 °C.</td>
<td>[5]</td>
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<td></td>
<td>¹(DNAPL) / 126–1122 mg kg⁻¹ / Sand / Electrical resistance heating</td>
<td>4 / investigate gas production and migration during for the removal of DNAPL</td>
<td>Temperature: ≤ 90 °C; time = 35 h; Flow cell; electrodes: (IrO₂-Ta₂O₅); groundwater velocities: 1.9 or 10.9 m day⁻¹; potential: 200 V; power density: 6–13 kW m⁻²; DNAPL volume: 270 mL; indicator: 40 mg L⁻¹ Oil-Red-O.</td>
<td>No DNAPL was present within the treatment zone; volume of gas produced: 114–131 L / high groundwater velocities can limit subsurface heating rates.</td>
<td>[7]</td>
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<td></td>
<td>²TCE / not reported / Graded sand / Electrical resistance heating</td>
<td>4 / Investigate the temperature for the removal of TCE and validation of numerical models on gas accumulation under passive venting conditions.</td>
<td>Temperature: ≤ 100 °C; time = 72 h; DNAPL pool volume: 293 mL stainless steel plate electrodes (30.48 x 10.16 x 0.3175 cm); spacing between electrodes: 35 cm. TCE was pigmented with dye (Oil-Red-O 144 mg L⁻¹) to allow visibility</td>
<td>Post-treatment soil concentrations of TCE ranged from: 6–136 mg kg⁻¹</td>
<td>[12]</td>
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<td>¹⁰PCP / 90±5.7 mg kg⁻¹ / sandy /</td>
<td>4 / investigates the behavior of ¹⁰PCDD/Fs formation, dichlorination and destruction in PCP-contaminated sandy soil.</td>
<td>Temperature: 200–400 °C; time: 30 min; Heating tape was wrapped around the connection tubes and the temperature was controlled at 150 °C to minimize pollutant accumulation on the tube walls.</td>
<td>70% of PCP removal / Desorption from soil was supposed as a main mechanism for the distribution of PCDD/Fs in the gaseous phase</td>
<td>[8]</td>
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<td>Tetrachloroethylene / 0.96 μg L⁻¹ / Sand sediment / EK</td>
<td>4 / Influence of field-extracted groundwater, sandy sediment and groundwater aquifer temperatures on the removal of contaminant</td>
<td>Temperature: 8–22 °C; time: 20 h; an undivided horizontal flow-through acrylic column reactor was used; i = 120 mA; constant seepage velocity: 150 m yr⁻¹ (Darcy velocities of 0.01 cm min⁻¹); groundwater: synthetic and field-extracted</td>
<td>80 ± 7% of tetrachloroethylene removal in the sandy sediment / VOCs in the gas collected accounted for 0.10 ± 0.05% of the chlorinated ethene removals observed in the porewater</td>
<td>[18]</td>
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<td><strong>Contaminated groundwater</strong> / <strong>not reported</strong> / <strong>EK</strong></td>
<td>Evaluate the transport of chlorinated solvents by electro-osmotic in clayey</td>
<td>Temperature: not reported; time: 31 days; pressure: 30 psi; electric field: 3.0~6.57 V cm⁻¹</td>
<td>It was observed chloroform production in the anode reservoir, resulting from electrochemical production of chlorine gas reacting with trace organics [19]</td>
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<td>**Trichloroethene / 2 mM / Clay topsoil and sand / <strong>EK- bioremediation</strong></td>
<td>Determine the influence of high level of acceptor sulfate, compare <strong>EK-Bio</strong> to a traditional bioremediation application without <strong>EK</strong>.</td>
<td>Temperature: not reported; time: 11 weeks; <strong>bioaugmentation culture</strong>: <strong>ZARA-10</strong>; the electrolyte was synthetic groundwater (10 mM sodium bicarbonate, 11.45 mM sulfate, and 2 mM TCE) at a pH of 8.5</td>
<td>Complete transformation of TCE to ethene and acetylene by <strong>EK-Bio</strong>, while only 15% of TCE was transformed to cis-dichloroethene and vinyl chloride via traditional <strong>bioaugmentation</strong>. In the <strong>EK-Bio</strong> reactor, the majority of the TCE was converted to acetylene, likely due to its electrochemical reduction at the cathode [14]</td>
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<td><strong>Pilot scale</strong></td>
<td>**Lindane / 10 mg kg⁻¹ / silty loam / <strong>EKSF with three types of PRB</strong></td>
<td>Evaluate <strong>EKSF</strong> couple with n-ZVI, m-ZVI or GAC*</td>
<td>Volatilized or transformed (removal efficiency): <strong>EKnZVIB</strong>: 50%; <strong>EKmZVIB</strong>: 65%; <strong>EKAB</strong>: 70%; no-<strong>EK</strong>: 2%; <strong>EKSF</strong>: 70%; <strong>REKSF</strong>: 30% [16]</td>
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<td>**Heavy oil, POPs, 3 PAH / 51200 mg kg⁻¹ / real soil / <strong>Microwave</strong></td>
<td>Contribute to the field of microwave remediation in empirical way.</td>
<td><strong>Removal efficiency</strong>: 99%; maximum temperature: 400 °C. [6]</td>
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<td><strong>Prototype scale</strong></td>
<td>**Benzene, chlorobenzene, and petroleum / 1560 mg kg⁻¹ / real soil / <strong>Thermal desorption steam extraction</strong></td>
<td>The performance of gas thermal remediation system in removing organics from site soil and groundwater.</td>
<td>Removal efficiency: benzene: 99%, chlorobenzene: 99% and petroleum: 98% / The removal rates in groundwater is: benzene: 98%, chlorobenzene: 97% and petroleum: 99%. [9]</td>
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<td>6ESTERON 60 and FLUOXIL / 20 mg kg⁻¹ / silty loam / EK</td>
<td>7 / The influence of size matters in the mechanisms for the removal of herbicides.</td>
<td>Maximum temperature: 55 °C; time: 35 days; dimensions of prototype: 2 m of height and a square plant of 16 m²; capacity: 32 m³; 6 electrodes: cylinders of graphite (15 x 100 cm); electric field: 1.0 V cm⁻¹; the microbial consortium was obtained from an activated sludge; Lab-scale soil: 1 L Pilot-scale mockup: 175 L Prototype: 32 m³ Removal efficiency: oxyfluorfen: 85.20% and 2,4-D: 87.26% were volatilized.</td>
<td>[13]</td>
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<td>4,2,4-D and 4OXY / 5.991 mg kg⁻¹ and 4.742 kg⁻¹ / silty loam / Electrobioremediation</td>
<td>7 / Perform a large-scale electrobiremediation test on a prototype composed of a soil remediation electrokinetic reactor with a soil treatment capacity of 32 m³.</td>
<td>Maximum temperature: 60 °C; time: 35 days; dimensions of prototype: 2 m of height and a square plant of 16 m²; capacity: 32 m³; 6 electrodes: cylinders of graphite (15 x 100 cm); electric field: 1.0 V cm⁻¹; the microbial consortium was obtained from an activated sludge. 7% of the 2,4-D and 9% of the oxyfluorfen remained in the soil after the treatment / volatilization was found to be the key mechanisms to explain the removal of both herbicides.</td>
<td>[15]</td>
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<td>CVOCs / 30 mg kg⁻¹ / Real soil / EK</td>
<td>7 / Investigated electrokinetics to enhance the delivery of persulfate and ERH to thermally activate the delivered persulfate.</td>
<td>Temperature: ~40 °C; time: 14 months; real applications in southwestern Ontario, Canada; all electrodes were composed of a titanium tube and IrO₂ coating; 925 L of 40 g L⁻¹ sodium persulfate was injected. Removal efficiency: 93% / The decrease in total CVOCs in groundwater can be associated with chemical oxidation.</td>
<td>[17]</td>
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</tbody>
</table>
1 Dense non-aqueous phase liquids
2 Trichloroethylene
3 Pentachlorophenol
4 Polychlorinated dibenzo-p-dioxins and furans
5 Ca (88 mg L⁻¹), Mg (38 mg L⁻¹), Na (124 mg L⁻¹), K (1.2 mg L⁻¹), PO₄³⁻ (0.2 mg L⁻¹), Cl⁻ (137 mg L⁻¹), SO₄²⁻ (46 mg L⁻¹), NO₃⁻ (5.4 mg L⁻¹), SiO₂ (30 mg L⁻¹)
6 ZVI (n-ZVI), granular particles of ZVI (m-ZVI), or granular activated carbon (GAC).
7 Polycyclic Aromatic Hydrocarbons
8 ESTERON 60: composed of 2,4-dichlorophenoxyacetic acid (2,4-D) with a composition of 60% (v/v), and FLUOXIL 24: composed of 24% (v/v) Oxyfluorfen
9 2,4-dichlorophenoxyacetic acid
10 Electro-kinetic soil flushing (EKSF) with three types of permeable reactive barriers (PRB)
<table>
<thead>
<tr>
<th>Type of soil / soil organic carbon</th>
<th>Technology</th>
<th>Technology Readiness Level / work objective</th>
<th>Operation conditions</th>
<th>Maximum efficiency reported/related remarks</th>
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<td>100 mg kg⁻¹ / soil</td>
<td>Low-temperature microwave</td>
<td>4 / investigate the low-temperature microwave-assisted remediation of petroleum-contaminated soil with simultaneous oil recovery at bench scale.</td>
<td>Moisture content: 6%; temperature: 80–300 °C; time: 20 min.</td>
<td>99% of mass removal; time: 20 min; temperature: 300 °C. The germination rate of clover seeds reached the maximum using 250 °C.</td>
<td>[5]</td>
</tr>
<tr>
<td>P₄₀ / PAH / T₇₅ / real soil</td>
<td>Microwave</td>
<td>7 / contribute to the field of microwave remediation in empirical way.</td>
<td>Ex-situ; power: 6 kW; frequency: 2.45 GHz; reactor volume: 250 L; sample mass: 70–95 kg; heating time: 260–400 min; pressure: 0–30 kPa; maximum temperature: 330–380 °C.</td>
<td>Removal efficiency: 99%; maximum temperature: 400 °C.</td>
<td>[12]</td>
</tr>
<tr>
<td>5 mg kg⁻¹ / Sand</td>
<td>Electrical resistance heating</td>
<td>4 / investigate gas production and migration during the removal of DNAPL.</td>
<td>Flow cell; electrodes: (Ir₂O₃-Ta₂O₅); groundwater velocities: 1.9 or 10.9 m day⁻¹; potential: 200 V; power density: 6–13 kW m⁻²; DNAPL volume: 270 mL; indicator: 40 mg L⁻¹ Oil-Red-O.</td>
<td>No DNAPL was present within the treatment zone; volume of gas produced: 114–131 L / high groundwater velocities can limit subsurface heating rates.</td>
<td>[6]</td>
</tr>
<tr>
<td>mg kg⁻¹ / sand</td>
<td>Electrical resistance heating</td>
<td>4 / Investigate the temperature for the removal of TCE and validation of numerical models on gas accumulation under passive venting conditions.</td>
<td>DNAPL pool volume: 293 mL stainless steel plate electrodes (30.48 x 10.16 x 0.3175 cm); spacing between electrodes: 35 cm; temperature: ≤100 °C. TCE was pigmented with dye (Oil-Red-O 144 mg L⁻¹) to allow visibility</td>
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<tr>
<td>benzene, and toluene / real soil</td>
<td>Thermal desorption steam extraction</td>
<td>1 / The performance of gas thermal remediation system in removing organics from site soil and groundwater.</td>
<td>Moisture content: 26–40 %; temperature: 100–200 °C; pressure: 0.5–2.0 kPa; experimental time: 34 days; the pilot trail is 10 m x 10 m with a depth of 18 m.</td>
<td>Benzene: 99%, chlorobenzene: 99% and petroleum: 98% / The removal rates in groundwater is: benzene: 98%; chlorobenzene: 97% and petroleum: 99%.</td>
<td>[7]</td>
</tr>
<tr>
<td>3 – 10 mg kg⁻¹ / sandy</td>
<td>Low-temperature pyrolysis</td>
<td>4 / investigates the behavior of PCDD/Fs formation, dichlorination and destruction in PCP-contaminated sandy soil.</td>
<td>Temperature: 200–400 °C; soil mass: 2 g; Heating tape was wrapped around the connection tubes and the temperature was controlled at 150 °C to minimize pollutant accumulation on the tube walls.</td>
<td>Most PCDD/F removal from the soil was observed at 350 °C; The main byproduct of PCP during pyrolysis was 2,3,4,5-tetra chlorophenol</td>
<td>[13]</td>
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<tr>
<td>5.3 kg / silty loam</td>
<td>EK</td>
<td>7 / The influence of size matters in the mechanisms for the removal of herbicides.</td>
<td>Lab-scale soil: 1 L Pilot-scale mockup: 175 L Prototype: 32 m³</td>
<td>Volatilization is the main mechanisms that explain the removal of 2,4-D and oxyfluorfen.</td>
<td>[8]</td>
</tr>
<tr>
<td>1–3 kg / Real soil</td>
<td>EK</td>
<td>7 / Investigated electrokinetics to enhance the delivery of persulfate and ERH to thermally activate the delivered persulfate.</td>
<td>Real applications in southwestern Ontario, Canada. All electrodes were composed of a titanium tube and IrO₂ coating</td>
<td>In situ transformation of CVOCs was assessed by compound-specific isotope analysis (CSIA) of 1,2-dichloroethane (1,2-DCA) samples collected after ERH</td>
<td>[15]</td>
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<tr>
<td>lene / kg</td>
<td>in soil/ sediment</td>
<td>Trichloroethene kg</td>
<td>Production L/ 45</td>
<td>Tetrachloroethylene kg</td>
<td>Production L/ 45</td>
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<td>Production L/ 45</td>
<td>Tetrachloroethylene kg</td>
<td>Production L/ 45</td>
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</table>

Table: Trichloroethene, Tetrachloroethylene, and Lindane Production in kg.

- **Time:** 20 h; An undivided horizontal flow-through acrylic column reactor was used; \( i = 120 \text{ mA} \); constant seepage velocity: 150 m yr\(^{-1}\); (Darcy velocities of 0.01 cm min\(^{-1}\)); temperature: 8–22 °C; groundwater: synthetic and field-extracted.

- **Capacity:** 175 dm\(^3\); time: 720 h; electric field: 1.0 V cm\(^{-1}\); tests: 1) a reference test (no-EK), 2) an electrokinetic soil-flushing test (EKSF), 3) reversible electrokinetic soil-flushing test (REKSF), 4) electrokinetic adsorption barrier (EKAB), 5) electrokinetic nano-ZVI barrier (EKmZVIB), and 6) electrokinetic multi-ZVI barrier (EKnZVIB).

- **Volatilized or transformed,** EKnZVIB: 50% EKmZVIB: 65% EKAB: 70% no-EK: 2% EKS: 70% REKS: 30%

- **Dimensions of prototype:** 2 m of height and a square plant of 16 m\(^2\); capacity: 32 m\(^3\); 6 electrodes: cylinders of graphite (15 x 100 cm); electric field: 1.0 V cm\(^{-1}\); the microbial consortium was obtained from an activated sludge.

- **Volatilization of gaseous streams in soil remediation processes**

- **PCE concentrations of 7 ± 2 mg L\(^{-1}\) were reached with TCE and cis-DCE being completely removed. / VOCs in the gas collected accounted for 0.10 ± 0.05% of the chlorinated ethene removals observed in the porewater.**

- **It was observed chloroform production in the anode reservoir, resulting from electrochemical production of chlorine gas reacting with trace organics.** Complete transformation of TCE to ethene and acetylene by EK-Bio, while only 15% of TCE was transformed to cis-DCE and VC via traditional bioaugmentation. In the EK-Bio reactor, the majority of the TCE was converted to acetylene, likely due to its electrochemical reduction at the cathode.
**Electrochemically-assisted treatments of gaseous effluents**

Regardless of the soil remediation technology implemented, in the search of a holistic treatment, both liquid and gaseous effluents produced during soil remediation must be treated. The treatment of wastewater has been extensively studied in the recent literature and, because of that, it is out of the scope of this manuscript [1].

However, the treatment of gaseous effluent generated in soil remediation processes is still a field with important gaps that must be covered. Thus, the gaseous effluent generated by soil remediation can be captured and treated using different technologies, among which, thermal and catalytic oxidation are the most important processes.

Direct application of electrochemical technologies is possible with the use of cells equipped with solid electrolytes (either membranes or ceramic oxides), such as those typically used in fuel cells, although almost no work can be found in the recent literature regarding the application of this technology to the removal of COVs. However, recently, it has been proposed the combination with electrochemical technologies with two well-known gas treatment technologies: adsorption and absorption. Both are not finalistic technologies, that is, they only separate the pollutant from the gaseous stream and transport it to a different support (solid or liquid) that should be further treated.

Adsorption is a surface process, where the pollutant molecules contained in a gaseous stream (adsorbates) adhere onto the surface of a solid (adsorbent). The mechanism can be chemical (chemisorption) or physical (physisorption) [21] and it is used in many industrial applications to fractionate air mixtures or to depurate gas [22,23], being actually at a very high state of
maturity, with a technology readiness level (TRL) range of 8–9. Activated carbon is the most frequently used adsorbent for gas treatment, but other adsorbents, such as biochar, graphene, carbon nanotubes [24], zeolites, molecular sieves, and synthetic polymers, are also gaining attention. The various advantages of adsorption include high efficiency, easy operability, economy feasibility, and simple equipment. Currently, there is a huge effort in the development of new adsorbents, because this is the key in reaching highly efficient processes. As an example of this significance, it has been recently reported that synthetic zeolite produced from coal fly ash and Na-zeolite (coal fly ash (CFA)-based zeolite Na, demonstrated a much higher capacity (383–582 µg g⁻¹) for the removal of benzene, toluene, and xylene in flux gas as compared to conventional Na-zeolite (zeolite without CFA) (1.44–20.50 µg g⁻¹), natural clinoptilolite (0.68–2.84 µg g⁻¹), and diatomite (0.20–0.96 µg g⁻¹) [25]. Disadvantages of adsorption include large and complicated equipment and complex procedures that are necessary for the desorption and regeneration of saturated adsorbents. In this context, recently, adsorption with GAC particles has also been integrated with electrochemical technology, trying to improve the removal of volatile compounds and the regeneration of the adsorbent. Thus, it has been proposed the adsorption of gases such as perchloroethylene onto GAC and the regeneration of GAC with methanol, which is then treated in an electrochemical cell equipped with diamond electrodes [26]. With this technology (Figure 2) a very successful removal of the gaseous pollutant can be reached because the electrolytic removal of the pollutants in methanol media is very efficient as methanol is almost not oxidized during the electrochemical process. The advantage of integrating adsorption-electrochemical technology is associated with the production of active species, such as chlorine that contribute towards eliminating the pollutant, as well as all
intermediates produced. Regeneration of GAC with water is also possible but the desorption equilibrium is more favorable in using methanol [26–29].

Figure 2. (a) The ADE (adsorption + electrolysis) concept for the treatment of gaseous streams polluted with organics and (b) The ABE (absorption + electrolysis) concept for the treatment of gaseous streams polluted with organics.

Absorption is a completely different treatment, where there is a transfer of one or more species (called solutes or absorbates) from the gaseous polluted stream into a liquid solvent. It is used widely to separate gas mixtures, remove impurities, or recover valuable chemicals. It is also at a very high state of maturity with a TRL range of 8–9. The selection of an appropriate solvent is crucial because the solubility of the solute in the solvent may become the bottleneck of the separation process, as recently pointed out by Sui et al. [30] in a study focused on the use of triethylene glycol (TGE) to treat an off-gas stream containing 16 VOCs. The recovery and recycling of solvent is also important and, depending on the
pollutant, it is generally done by stripping or distillation (in case of using water as solvent) or by destruction technologies. Thus, in the previously referenced work, authors reported a high recovery of VOCs (~97%) associated to the coupling of thermal processes during the stages of absorption and desorption. Absorbers are also coupled with destructive technologies and among them, it is worth to highlight electrochemical technologies. Figure 3 shows a scheme of an absorption column coupled with electrolysis.

Regarding this combination of absorption technologies with electrochemical systems, Huang et al. [31] studied the removal of styrene from an air stream in an aqueous absorption solution, which enhanced the mass transfer when combined with electrochemical oxidation using Ti-based anode with a RuO₂ layer. They reported complete removal of the initial styrene content in the presence of chloride precursors, (HCl and 1% NaCl), and partial removal in the presence of Na₂SO₄. The process efficiency increased when the pH of the solution decreased (pH value of solution adjusted to 1). The major liquid-phase products, obtained from styrene oxidation, were 1-phenyl-1,2-ethanediol and benzaldehyde. This indicates that the absorption-electrochemical approach not only helps in the removal of styrene, but it also may promote the conversion of pollutants into high-value-added products.

Yang et al. [32] studied the effect of different factors, such as a cell voltage, electrolyte and temperature, on the electrochemical removal of gaseous elemental mercury in the liquid phase using a Ti/Sn-SbO₂/PbO₂ electrode. More than 90% of mercury was removed under the conditions of 3 V, gas flow of 50 mL/min, electrolyte concentration of 0.5 M Na₂SO₄, pH 3, and temperature of 40 °C. It was observed that Ti/Sn-SbO₂/PbO₂ was more suitable for use in acidic conditions, while Ti electrodes are more suitable in alkaline conditions. González-Pérez et al. [23] proposed the use of a novel jet electro-absorber that combines absorption based on the Venturi effect and electro-oxidation for the treatment of gaseous organochlorine
Results confirmed that the target compound can be absorbed into the electrolytic solutions proposed and that the efficiency depends on the solubility of pollutant in the selected electrolytic media. The size of the bubbles generated by the jet-absorber had a paramount influence on the mass-transport and on the removal pathway. Castañeda-Juárez et al. [33] proposed the development of electro-absorption and photoelectro-absorption technologies to treat gases produced by a synthetic waste containing the highly volatile perchloroethylene (PCE). To do this, a packed absorption column coupled with a UV lamp and an undivided electrooxidation cell was used. Firstly, it was confirmed that the absorption in a packed column is a viable method to achieve retention of PCE into an absorbent-electrolyte liquid. Electro-absorption using a non-divided electrolytic cell is capable to promote hard oxidation conditions leading to different reaction pathways. Mineralization obtained was very low and treatment costs increased seriously with the total power applied. Ramu et al. [34] studied the electrochemical reductive degradation of gaseous chlorobenzene using electrogenerated homogenous low-valent [Ni(I)(CN)₄]³⁻. An electro-scrubbing column at ambient temperature associated with the cathodic half-cell with a membrane-divided electrolytic cell was employed. They reported that the presence of Ni(I) contributed to the removal of chlorobenzene by a homogenous electron mediator [Ni(I)(CN)₄]³⁻ (Ni(I) low-valent). In addition, FTIR gas analyses confirmed the presence of chlorine, and CO₂ was the only gaseous product. Furthermore, phenoxide anion, which is an important material in the pharmaceutical industry was also detected. Hence, electrochemical technology, coupled with other processes, contributes to the removal of contaminants present in gaseous flow, as well as it may generate important compounds during the regeneration of the absorbent [33]. It is important to highlight that estimated energy requirements and costs of the adsorption-absorption-electrolysis, it is not easy to obtain for all information to do a complete techno-
economic because depend mainly on electrode materials, materials absorption or adsorption, reactor configuration, operational parameters, as well as the electricity cost varies for different gaseous effluents.

**Integration of technologies: soil remediation and gaseous flow treatment.**

Very often, the performance of soil decontamination techniques has been evaluated separately and very few works have addressed a holistic treatment of the processes, focusing not only on the mobility and elimination of the pollutant from the soil but also on the treatment of the liquid and/or gaseous phases generated [4,6,16,35,36]. In fact, the treatment of liquids generated is found occasionally but it is rare to observe the treatment of gases produced (except for studied focused on technologies which exclusively aims to produce gases) [1]. Table 2 exhibits data regarding different approaches that integrate soil remediation and treatment of gaseous effluent.

As seen, electrochemical technologies are not yet fully integrated in this treatment, despite the good prospects shown in the previous sections. Thus, among the works included in the Table 2, it is worth to highlight several which are non-based on the use of electrochemistry. Thus, in order to achieve a high efficiency in the removal of toluene in the gas stream generated from the steam extraction process, Fenelon et al. [37] proposed a photocatalytic module under visible light and an adsorption module to eliminate residual contaminants. Titanium dioxide doped with lanthanum was used as a photocatalyst and activated carbon as an adsorbent. The removal efficiency was stable for more than 72 hours and exceed values above 95% in one hour. On the other hand, to assess the effectiveness of heat treatment of a soil contaminated with hexachlorocyclohexane (HCH), Araújo et al. [38] subjected soil samples to different temperatures and heating times. The gaseous emissions generated during the process were subjected to a purifier containing NaOH and adsorption columns with activated carbon. The
gas emissions detected were HCH isomers, chlorine derivatives and benzene compounds. Almost 100% of the HCH isomers were volatilized.

Regarding electrochemically-based process, Wieczorek et al. [35] studied on lab-scale and pilot-scale remediation of soil contaminated with heavy metals (Zn, Ni, Cu, Cr). It was reported the formation of chlorine gas near the anodic region associated with the chloride present in the soil or electrolyte. To prevent atmospheric contamination, a gas absorber unit was developed using an activated carbon filter loaded with potassium carbonate. Furthermore, they observed that on the lab-scale, the EK phenomenon is predominant in the transport of pollutants, while on the pilot scale, volatilization is more dominant. This behavior is due to the higher temperature observed in pilot-scale facilities, which is associated to the higher ohmic losses because of the larger interelectrode distance, as discussed before. Also pointing out the necessity of gas control scrubbers to prevents atmospheric contamination in full-scale plants, Iannelli et al. [39] reported the formation of gaseous chlorine due to the oxidation of chlorides associated with the HCl electrolyte in low pH conditions. On the other hand, during a study about the EK transport of ionic species and the removal of pesticides, Vizcaíno et al. [13,36] observed that the amount of gases produced could be successfully treated by absorption in the presence of 10 g L\(^{-1}\) of sodium dodecyl sulfate (SDS), which becomes a very relevant input because to the knowledge of authors, in the recent literature, there are very few works in which this type of treatments are associated to EASR.

Most recent works have been carried out toward of the integration between electrokinetic remediation and production of hydrogen, which despite it is not a pollutant it may have important implications in the sustainability of the processes because of its role as energy carrier. Hydrogen production is an eco-friendly approach that has been investigated for water splitting or simultaneous degradation of organic pollutants in divided or undivided
electrochemical cells [40,41]. Nevertheless, the reactive and thermal processes that naturally occur during EK soil remediation can consequently contribute to the production of gases [4]. Most recently, electrokinetics have been coupled with electrodialytic technology for H$_2$ production during the removal of pollutants. The cathode compartment can be used in proton-exchange membrane fuel cells, which can produce electrical energy and reduce the energy costs of treatment [42,43]. The application of electrochemically assisted technologies to remediate soil and produce H$_2$ can also be applied to solid-liquid matrices. The energy requirements for this system are considerably smaller owing to the higher conductivity of the liquid. New ideas on the topic have been explored to save energy during electrochemically assisted soil remediation and H$_2$ production for in electrochemical technology.
<table>
<thead>
<tr>
<th>Pollutant / concentration / soil</th>
<th>Gas treatment technology</th>
<th>work objective</th>
<th>Primary operation conditions</th>
<th>Maximum efficiency reported/related remarks</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Toluene / 15 mg L(^{-1}) / solid vapor extraction</td>
<td>Photocatalytic oxidation and adsorption</td>
<td>Develop a commercial module that can be directly connected to exhaust the SVE method by using lanthanum-doped titanium dioxide (La-TiO(_2)); Irradiation sources: Visible light; 5 lamps holders inside the module adjusted in different position; the adsorption module used 5 layers of carrier and 30 g of activated carbon in each carrier.</td>
<td>Photocatalyst: glass fiber cloth coated with lanthanum-doped titanium dioxide (La-TiO(_2));</td>
<td>Removal efficiencies: 95 %; time: 1 h.</td>
<td>[37]</td>
</tr>
<tr>
<td>Lindane / 10 mg kg(^{-1}) / silty loam</td>
<td>Adsorption</td>
<td>Evaluate EKSF couple with n-ZVI, m-ZVI or GAC.</td>
<td>Cylindrical adsorption column: 1 cm in diameter and 20 cm in length filled with activated carbon.</td>
<td>3 mg were retained (and quantified) in the adsorption column through gas phase passes</td>
<td>[16]</td>
</tr>
<tr>
<td>2,4-D and oxyfluorfen / 20 mg kg(^{-1}) / real soil</td>
<td>Absorption</td>
<td>Evaluate the transport of ionic species during to the removal of pesticides</td>
<td>Gases produced in the electrolyte wells to an absorber tank, which contains with 10 g L(^{-1})of SDS</td>
<td>-</td>
<td>[13]</td>
</tr>
<tr>
<td>Zn, Ni, Cu, Cr / 308, 1251, 860 and 890 mg kg(^{-1}) / real soil</td>
<td>Absorption</td>
<td>Study in scale-up to the removal heavy metals, process water and gas treatment.</td>
<td>Lab: A series of electrode arrangements, 30V, 5000 h; Scale-up: A series of electrode arrangements, 30V, 10 months</td>
<td>Chlorine gas was treated by absorber unit using activated carbon filter.</td>
<td>[35]</td>
</tr>
<tr>
<td>2,4-D and oxyfluorfen / 20 mg kg(^{-1}) / real soil</td>
<td>Absorption</td>
<td>Focusing on the evaluation of the mechanisms by lab scale and pilot prototype system</td>
<td>The gases produced in the electrolyte wells to an absorber tank, which contains with 10 g L(^{-1})of SDS</td>
<td>-</td>
<td>[36]</td>
</tr>
<tr>
<td>Cd, Cr, Cu, Ni, Pb and Zn / 1.8, 86, 393, 54, 38 mg kg(^{-1}) / sediment</td>
<td>-</td>
<td>Evaluate the efficiency of the removal different electrolyte in lab scale and full-scale</td>
<td>Evaluate the effect of conditioning agents and dosages on the remediation yield using graphite electrodes</td>
<td>Chlorine gas was produced due to the very low pH condition and high chloride content in the electrolytes. Complete vaporization was achieved the cell</td>
<td>[39]</td>
</tr>
<tr>
<td>Trichloroethene / silica sand</td>
<td>Condensation</td>
<td>Understanding how gas production, transport changes electric current and temperature measurements</td>
<td>The graphite rod electrodes were used in presence of NaCl (4.6 g L(^{-1}))</td>
<td></td>
<td>[44]</td>
</tr>
</tbody>
</table>
Hexachlorocyclohexane / 96.4 μg g⁻¹ heating electrical resistance

Condensation, absorption, adsorption. 4 / Get thermogravimetric data, which involves HCH volatilization and its HCH derivatives from reference sample materials (RM) and from anthropogenic contaminated soil samples.

Treatment system: vacuum pump, condenser, scrubber (0.25 mol m⁻³ of NaOH), zeolite and activated carbon filters.

The gas and vapors samples collected from output of the system revealed the absence of HCH isomers. The use of an alkaline solution of NaOH to assess the dechlorination of HCH showed that the NaOH solution was an environment useful to retain the isomers.
Mathematical modelling of electrochemical remediation

Mathematical models are necessary to gain a better understanding of the processes that occur in the EASR [11,45–47]. Many types of models are proposed in the literature and, considering the spatial distribution that they try to represent, they can be classified into:

- 1-D models, frequently used for preliminary understanding the decontamination of soils in low TRL works [46,48],
- 2-D models, which help to obtain a higher understanding of the mobility of pollutants within an electric field, being widely applied in intermediate TRL assessments to describe not only the electrokinetic removal of metals [49,50] but also of organic contaminants [51], and
- 3-D models, which allow to evaluate full-scale treatment in advanced TRL works by describing not only unsaturated flow but also multispecies solute transport, geochemical reactions, solute transport & reactions, electric gradient, electric current lines and gas phase exchange[52].

Complexity of the models and computational work increases with the dimensions of the model used and with the number of processes represented. Nonetheless, they are becoming key tools for the evaluation of full-scale remediation actions and, nowadays, more and more exigent models are being developed. A very interesting point worth to be highlighted is the modeling of the gaseous flows generated during the electrochemically assisted treatment, because these gaseous streams need to be properly collected and treated to reach an efficient and holistic treatment in the remediation of soils polluted with VOCs[53]. Many processes should be considered, including the formation and release of gas bubbles, the interaction of these bubbles with the soil and
the potential occurrence of heterogeneous chemical reactions [10]. Henry’s law states have demonstrated to be an important tool to simulate the production of gaseous waste and several works focused on considering multi-species chemical equilibrium have been recently reported[11], pointing out that there are a plethora of inputs to be considered to reach a proper interpretation of the results obtained in laboratory tests for attain an effective real application of the technology.

**Perspectives and recommendations**

Electrochemical processes can be used to recover and treat pollutants contained in soil. Despite there is a lot of information about the use of electrokinetic technology to remove organic pollutants from soil, very little attention has been paid to the electric heating of the soil during these processes and to the gaseous flows generated. Many works have highlighted that in full scale applications the controlling mechanism in the transport of species under application of electric fields to soil is not electrokinetic but the gas transport, because of the important heating of soil, which opposes to the evidences of the studies in lab-scale and points out the relevance of the topic discussed in this work.

Another important feature is that the energy consumption and associated costs are the major challenges for the application of these technologies at the full scale. Powering with renewable energies arises as a promising alternative to grid electricity, not only to save money but also to make these processes more sustainable, eco-friendly and autonomous, helping to reduce the environmental problems associated with the use of fossil energy[54–56].
Regarding the treatment of gases using electrochemical technologies, despite it is possible the use of cells equipped with solid electrolytes to directly treat gases, the most promising technologies found recently in the literature combine adsorption and absorption of gases with electrolytic treatment of the liquid used to regenerate the adsorbent in the first case or with the absorbent in the second. The very interesting results reported highlight the relevance of the topic and the necessities of further studies. Finally, despite holistic solutions for the removal of pollutants from soil are needed to propose the most sustainable treatment technologies, very few works can be found in the literature facing the treatment not only of polluted soil but also of the liquid and gases produced simultaneously. This situation is even worse in the case of the electrochemically-based processes for which only the recovery of gases generated in the electrode (chlorine or hydrogen) and, very rarely, the pollutants volatilized has been faced. The successful results reported suggest that this is a worthy topic of research for the next future.

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References

Papers of particular interest, published within the period of review, have been highlighted as:

*Paper of special interest.

**Paper of outstanding interest.


Relevant paper for understanding the combination of computing multi-species chemical equilibrium with algorithm to discuss about different species from soil.


The authors compare different scale EK technology for the removal of pesticides (2,4-dichlorophenoxyacetic acid and oxyfluorfen) demonstrated the role of the controlling mechanisms in each scale. In prototype scale the volatilization was the main mechanisms.


The combination of electrokinetically-delivered thermally activated persulfate for the remediation of organic contaminants, allows to demonstrate that, improve the removal of total volatile compounds due to both chemical oxidation.


The authors used for the first time Venturi effect as news system used as electro-absorber that combines absorption and electro-oxidation for treatment of perchloroethylene. The results indicates that the combination is a promising alternative to increase the efficiency, and consequently, its applicability is feasible.

This manuscript shows how adsorption-desorption-electrolysis can contribute for the removal of organochlorinated compounds (clopyralid, lindane and perchloroethylene).

The authors demonstrated that combination of absorption and electrochemical technology can be achieved using granular activated carbon (CAG) particles. Both the utilization of absorption and electrochemical contributed to the removal of perchloroethylene and the regeneration of the adsorbent can be successfully used.

This interesting study demonstrated the removal of styrene from an air stream in an aqueous absorption solution combined with electrochemical oxidation using Ti-based anode with a RuO2 layer. It concludes that total removal of the styrene in the presence of chloride precursors was associate to the active chlorine species.


This work describes how the presence of Ni(I) can be contributed to the removal of chlorobenzene and generation of value-added intermediates using electroscrubbing column associated with the cathodic half-cell and membrane-divided electrolytic cell.


This paper elucidates as scale-up of the electrokinetic fence technology influence the transport of pesticides and inorganic compounds. The results indicates an increase in the temperature caused by the ohmic resistance of soil, consequently contributes to volatilization of pesticides.


This paper investigate electrokinetic remediation for the removal Mn in soil, sediments, sludges and others solid porous matrices. The results demonstrate that the use of organic acids as facilitating agents improved the removal of Mn.


Example of the use optical fiber as sensor for monitoring volatile hydrocarbon.


Example of the use optical fiber as sensor for monitoring volatile hydrocarbon.

