

# Thermal conductive heating coupled with *in situ* chemical oxidation for soil and groundwater remediation: A quantitative assessment for sustainability

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## ABSTRACT

Thermal conductive heating (TCH), broadly applied to remove volatile and semi-volatile organic contaminants from subsurface, is often considered unsustainable due to its high carbon emissions, energy consumption and expensive costs. TCH coupled with *in situ* chemical oxidation (TCH-ISCO) has drawn much attention recently due to its ability to achieve cleanup goals of groundwater remediation in a timely manner as TCH does, but at much lower temperatures. However, there is a lack of quantitative assessment on the sustainability of TCH-ISCO based on field data. In this study, a quantitative life cycle assessment approach coupled with best management practices (LCA-BMPs) model was developed with data collected in the field to assess the sustainability of TCH-ISCO for chlorinated aliphatic hydrocarbons (CAHs) contaminated soil and groundwater remediation for the first time. The results revealed that the energy supply via electricity contributed to 68% and 93% of the adverse environmental impacts (e.g., global warming, mineral resource scarcity, and fine particulate matter formation), for TCH-ISCO and TCH, respectively. In addition, the carbon emissions and direct cost of TCH-ISCO were estimated to be 80% and 70% less than TCH, respectively. With regard to the social sustainability, TCH slightly outperformed TCH-ISCO. The overall sustainability scores of TCH-ISCO and TCH were 89.6 and 61.9, respectively, demonstrating that the sustainability performance of TCH-ISCO was significantly better than that of TCH. In view of the current demand for sustainable remediation technology, this study provides reliable data evaluation for the application of TCH-ISCO.

## 1. Introduction

*In situ* thermal treatment (ISTT), a technology by introducing energy into the subsurface to mobilize and recover volatile and semi-volatile organic contaminants (O'Brien et al., 2018; Vidonish et al., 2016b), is particularly applicable at sites where short cleanup timeframes are required (Zhao et al., 2019). Being less sensitive to subsurface heterogeneities than other technologies, ISTT can simultaneously treat many volatile organic contaminants (VOCs) and semi-volatile organic contaminants (SVOCs), hence, ISTT can be the only method that achieves the cleanup objectives in many cases (Cioni and Petarca, 2011). However, the cost of ISTT is typically higher than other remediation

technologies (Vidonish et al., 2016b). In addition, the heat utilization efficiency of high-temperature remediation (e.g., 300 °C–550 °C) is low, with up to 80% of the heat lost in the process of soil and groundwater heating (Vidonish et al., 2016b). Furthermore, soil minerals and organic matter can be decomposed and destroyed at high temperatures during ISTT (Bucala et al., 1996; Vidonish et al., 2016a). Therefore, ISTT is often considered unsustainable because of its high cost, high energy consumption, and limited ability to restore soils and ecosystems (Lemming et al., 2010b; O'Brien et al., 2018). A few researchers have observed that combined physical and chemical remediation methods are significantly superior to single remediation methods, with improved remediation efficiency, novelty, sustainability and reduced cost (Zheng

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et al., 2022).

Green and sustainable remediation (GSR) has gradually become an integral global approach for remediating contaminated sites, especially in reducing global carbon emissions and protecting the ecosystems. In recent years, researchers explored different options to improve the sustainability of ISTT and sought more sustainable ISTT (Ding et al., 2019), an example of which is the thermal conductive heating coupled with *in situ* chemical oxidation (TCH-ISCO). Previous studies have demonstrated that persulfate (PS) is more environmentally friendly than other reagents in ISCO (Ayoub et al., 2010). Persulfate is a robust and effective oxidant, and upon activation, strong and non-specific free radicals, such as the hydroxyl ( $E_h = 2.8$  V) and sulfate radicals ( $E_h = 2.6$  V), were generated for the purpose of destroying VOCs and SVOCs, despite their short persistence of these radicals under environmental conditions. Laboratory studies have shown that thermally activated persulfate can remediate contaminated soil and groundwater more efficiently, and remove refractory pollutants (Head et al., 2020; Ranc et al., 2017; Zhang et al., 2018). Therefore, it was believed that TCH-ISCO can be used to remove volatile or semi-volatile organic pollutants, and achieve cleanup goals of soil and groundwater remediation in a timely manner as TCH does, but at much lower temperature, energy and costs, fitting into the aim and concept of GSR. Since studies on TCH-ISCO evaluation are mostly conducted at the laboratory-scale, the potential environmental benefit of TCH-ISCO applications at the field-scale contaminated sites is unknown, and the sustainability of TCH-ISCO has not yet been evaluated adequately. There is a growing need to establish a framework for evaluating TCH-ISCO with sustainable objectives, such as energy saving, human health risk and ecosystem preservation (Vidonish et al., 2016b).

In the past few years, many countries have published GSR guidelines globally, which generally classify sustainability into the categories of environmental, economic, and social, considering these three pillars equally important (Rosen et al., 2015). GSR requires that the environmental impacts of remediation processes be reduced, while maximizing the net benefit between the society and the economy (Cundy et al., 2013; Rosen et al., 2015). There is a great leap in the development of methods for the quantitative evaluation of the environmental impacts of remedial technologies (Ding et al., 2022; Hou and Al-Tabbaa, 2014; Jin et al., 2021; Sparrevik et al., 2011), such as life cycle assessment (LCA). However, the majority of the quantitative assessments using LCA were conducted with data from various sources and assumptions (Ding et al., 2022; Lemming et al., 2010a; Liu et al., 2022), due to the difficulty in collecting field data. In addition, there are still challenges to quantify the social and economic impacts and net benefits (Bardos et al., 2015; Hou et al., 2014b). For example, it was concluded that the sustainability of the contaminated site remediation project depends on the opinions and preferences of various stakeholders (Harbottle et al., 2008). Hou et al. found that most decision-makers' perceptions of social and economic sustainability are subjective (Hou et al., 2016). Emerging economic and social assessment methods, such as the sustainability assessment framework proposed by the Sustainable Remediation Forum in the UK (SuRF-UK) (Bardos et al., 2016), have primarily adopted qualitative or semi-qualitative scorings, which are inevitably influenced by subjective factors, resulting in significant uncertainty in economic and social indicators, and making the sustainable remediation of contaminated site more complicated (Bueno et al., 2021). Therefore, it is important to establish an applicable quantitative assessment framework to make economic and social impacts more reliable while minimizing the subjective influence of decision-makers toward ensuring the credibility of sustainable assessment results.

Best management practices (BMPs) in different remedial technologies have been developed extensively in the past few decades, and the U. S. Environmental Protection Agency (USEPA) suggested that BMPs can help balance environmental, economic, and social sustainability (Prior, 2016). SuRF-UK defines BMPs as a method to achieve the main sustainability goals and listed 182 potential BMPs that impact

environmental, economic, and social sub-indicators (Bardos et al., 2015). A clear mapping exists between the benefits of BMPs and the metrics criteria of GSR, indicating that BMPs can be a popular tool for assessing sustainability, especially for economic and social assessment (Petruzzi, 2011). Many BMPs have been applied throughout different remedial processes to enhance its sustainability by taking relatively common beneficial actions to maximize the conservation of natural resources, protect of human health, and lower economic costs (Chen et al., 2020b; Ye et al., 2017). Therefore, exploring a set of BMPs that applicable to the remedial activities in the application of TCH-ISCO can be of great value for the sustainability evaluation of the social and economic impacts. Furthermore, researchers are paying more attention to the tools that can achieve the overall sustainability assessment with the advancement of GSR, yet it is still challenging to integrate the results of various sustainability assessment sub-indicators into the overall sustainability (Cappuyns, 2016). The multi-criteria analysis (MCA) normalizes various quantitative and semi-quantitative sub-indicators into a comprehensive evaluation standard for the sustainability assessment of different remediation strategies (Albizzati et al., 2021; Gallagher et al., 2013), with the primary objective to define the extent to which the remediation technologies meet the established criteria.

In this study, we developed a quantitative life cycle assessment approach coupled with best management practices (LCA-BMPs) model, using an extensive dataset, including the consumption of energy and materials, economic and social input indicators, for a field-scale study on the application of the TCH-ISCO. A systematic evaluation on the sustainability of TCH-ISCO was conducted with a quantitative assessment method established for the environmental, social and economic impacts. The objectives of this study were to (1) quantitatively assess the environmental sustainability assessment of TCH-ISCO by LCA, (2) explore the feasibility of using BMPs as the basis for economic and social sustainability of TCH-ISCO, and (3) integrate the environmental, social and economic impacts into the overall sustainability of TCH-ISCO using MCA.

## 2. Materials and methods

### 2.1. Sustainability assessment framework and methodology

The sustainability assessment framework consists of environmental, social, and economic indicators, with a total 14 sub-indicators developed in the framework, as summarized in Table S1 and discussed briefly below (Bardos et al., 2016; Cappuyns, 2016; Hou et al., 2014b; Liu et al., 2022; Song et al., 2018). Environmental sub-indicators included human health, ecosystems, and resources (Liu et al., 2022), which were calculated quantitatively by LCA. Economic sub-indicators included cost (direct project cost, project risk, and duration) and benefit (land value increase and employment), among which the cost of remediation was one of the most significant barriers to sustainability (Hou et al., 2014a). Social sub-indicators included worker safety (worker injuries and health risk), public acceptance (community involvement and community satisfaction), and community impact (project transparency, disturbance of community due to dust, noise, and odor). Previous studies have shown that worker safety and community issues were the most critical factors in social sustainability assessment (Hou et al., 2014a). Moreover, public acceptance was necessary for measuring the public satisfaction with the remediation effort. In this study, direct project cost, remediation duration in economic sub-indicators, and worker injury in social sub-indicators were calculated quantitatively based on the actual data collected for the TCH-ISCO demonstration program. Other economic and social sub-indicators were obtained quantitatively by BMPs developed as described in Section 2.1.2.

2.1.1. LCA-based environmental impact assessment

2.1.1.1. *Goal, scope and functional unit.* The life cycle scope spanned from the start of the material acquisition to the end of remediation for both TCH-ISCO and TCH. We thoroughly evaluated the application processes for TCH-ISCO and TCH and divided the remediation life cycle into four phases: material acquisition, construction, operation, and end-of-life activities (Fig. 1) (Ni et al., 2020; Sanscartier et al., 2010). For the engineered application of TCH, the major remedial activities include the construction and operation of wells (heating wells, extraction wells and monitoring wells) and heating of soil and groundwater with significant energy input. In the application of TCH-ISCO, injection wells are needed for the injection of chemical oxidative reagent. The activities post remediation mainly consist well abandonment and waste disposal. It is worth noting that the evaluation excluded machinery and vehicle manufacturing, primarily due to the lack of reliable data, as discussed similarly in previous studies (Lemming et al., 2010b; Liu et al., 2022; Ni et al., 2020). In order to compare the LCA results of TCH-ISCO and TCH, a functional unit in this study was defined as “the remediation of 1000 m<sup>3</sup> of contaminated soil and groundwater”.

2.1.1.2. *Life cycle inventory.* The primary life cycle inventory (LCI) data, such as the electricity, water and the amounts of materials consumed and resource depletion, should be accurately recorded during the remedial process, and the Ecoinvent v3.5 database was used as the upstream data (Wernet et al., 2016). Specifically, the material acquisition inventory included the materials that make up the heating wells, injection wells and temperature wells, the on-site equipment, and the transportation (Fig. 1). The inventory of the construction phase included the energy used for drilling wells, the materials used for site leveling and the transportation. The operation phase inventory included the electricity consumption for wells working, the reagent dosage, and the transportation. The end-of-life activities consisted of wells filling and waste transportation. All basic materials/resources introduced into the inventory, calculation method and additional descriptions applied in the LCA for different phases are summarized in Support Information (Tables S2–S5).

2.1.1.3. *Life cycle impact assessment.* The SimaPro 9.0 software was used to evaluate the life cycle environmental impacts in this study. The ReCiPe 2016 method with midpoint and endpoint levels was selected

(Jin et al., 2021; Song et al., 2018; Sparrevik et al., 2011; Vigil et al., 2015), because it provides more intuitive environmental impact results (Song et al., 2018). The midpoint level is located along the cause-impact pathway to offer LCA results with higher certainty for the selected environmental categories. The midpoint categories evaluated and compared in this study included global warming (GW, kg CO<sub>2</sub> eq), fine particulate matter formation (FPMF, kg PM<sub>2.5</sub> eq), terrestrial ecotoxicity (TET, kg 1,4-DCB), freshwater ecotoxicity (FET, kg 1,4-DCB), human carcinogenic toxicity (HCT, kg 1,4-DCB), human non-carcinogenic toxicity (HNCT, kg 1,4-DCB), water consumption (WC, m<sup>3</sup>), mineral resource scarcity (MRS, kg Cu eq), fossil resource scarcity (FRS, kg oil eq), and terrestrial acidification (TA, kg SO<sub>2</sub> eq). Compared to the midpoint level, the endpoint level further reflects environmental impacts in three main areas: human health, ecosystems, and resources, and integrates them into a single final score allowing for comparative and management-oriented results to be obtained (Song et al., 2018).

2.1.1.4. *Sensitivity analysis.* The sensitivity analysis was conducted to examine the importance of the main input parameters to each environmental impact category for the comparison between TCH-ISCO and TCH. It was found in previous studies that transport distance was a key factor in environmental sustainability, especially for *ex situ* treatment (Liu et al., 2022). It was expected that the electricity consumption, as well as reagent consumption, would be key parameters affecting the sustainability of TCH-ISCO. Therefore, the specific parameters analyzed for sensitivity included electricity consumption, transport distance, and PS dosage, using values half and double of the LCI data collected from the demonstration program, respectively.

2.1.2. Assessment of economic and social sustainability based on BMPs

This study assessed the economic and social sustainability quantitatively by correlating the sub-indicators with BMPs (Tables S6–S8). The BMPs suitable for TCH-ISCO and TCH, including remediation workplan, material acquisition, site construction, site operation, monitoring, and management post-remediation, were obtained and screened. The corresponding relationship between BMPs and economic and social sustainability sub-indicators was explored by referring to relevant policies (Hou et al., 2014b; Jin et al., 2021; Song et al., 2018). To reduce the influence of human subjectivity, BMPs were applied to the scoring process based on the proportion distribution method (Chilingarian and Zazian, 1990). The score was calculated by the ratio between

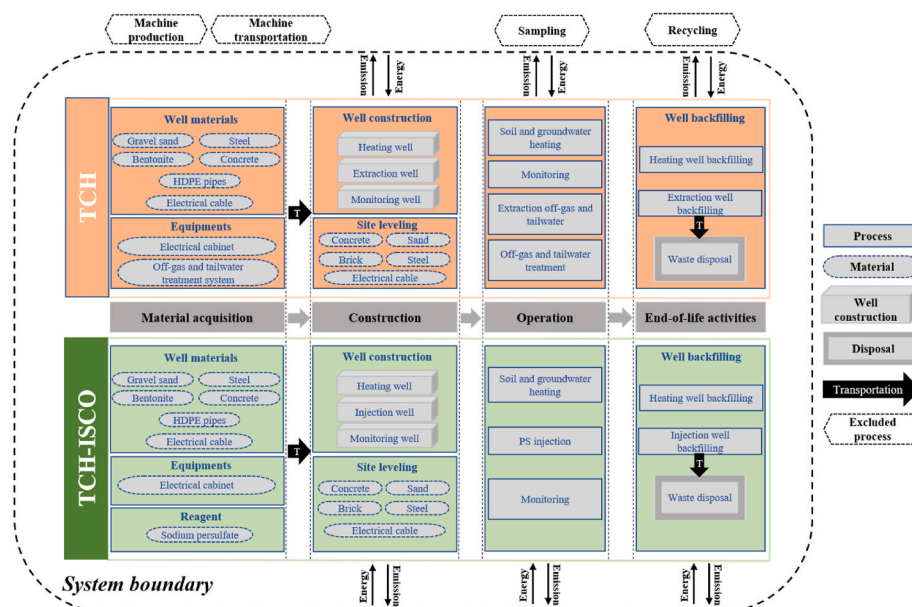


Fig. 1. System boundary of LCA for TCH-ISCO and TCH.

implementing BMPs related to each sub-indicator and each sub-indicator to the total number of BMPs.

However, there is still uncertainty in the evaluation results of some sub-indicators through BMP implementations only. In this study, we integrated the results of the expert evaluation into the calculation for controversial sub-indicators, and the weights of expert evaluation results and BMPs were both set at 50%. The specific calculation results are shown in Tables S6–S8. The formulas without/with consideration of the expert scores are shown in Equations (1) and (2), respectively:

$$S_{BMP} = \frac{S_a}{M_a} \times M \tag{1}$$

$$S_{BMP} = \left( \frac{S_a}{M_a} \times M \times 50\% \right) + 50\% \times S_b \tag{2}$$

where  $S_a$  is the total value of the quantitative assignment, and the value is 5;  $S_b$  is the expert scoring value, ranging from 1 to 5;  $M$  is the number of BMPs implemented;  $M_a$  is the number of BMPs among related sub-indicators;  $S_{BMP}$  is the final sub-indicator score, which ranges from 1 to 5. The higher the  $S_{BMP}$  score, the greater the sustainability.

### 2.2. Overall sustainability assessment

Several decision-making tools for contaminated sites in the US and Europe are based on the MCA analysis (Cappuyns, 2016). Given the current situation of remedial activities, the environmental impact of contaminated site remediation is more considered than the social and economic impacts. Applying the MCA to the sustainable assessment process can improve the balance of these three pillars (Colapinto et al., 2020; Harbottle et al., 2008). The MCA score calculation method is presented as follows:

$$S_{MCA} = \sum \left( \frac{V}{V_{max}} \times W \right) \times 100 \tag{3}$$

where  $V$  is the score of each sustainability sub-indicator (0–5);  $V_{max}$  is the maximum score of each sub-indicator;  $W$  is the weight of each sub-indicator, and the sum of sub-indicator weights of environmental, economic, and social is 1 (Song et al., 2018);  $S_{MCA}$  is the final sustainability score (0–100), and the higher the score, the more sustainable the technology.

The weights of each indicator and sub-indicator were considered, aiming to reduce the subjectivity of the weighting (Balasubramaniam et al., 2007). The three environmental sub-indicators, including human

health, resources, and ecosystems, were considered equally important and assigned 1/3 each. In the economic assessment, the project risk was considered the most weighted sub-indicator, assigned a value of 0.3, due to the high cost of additional remediation activities associated with failing to meet the remedial objectives in the first place. Worker injuries and health risks in social sub-indicators had a higher weighting (0.2) because it was found that the most significant concern during remediation was workers' safety and health risk to the community (Hou et al., 2014a). Detailed weight setting is provided in Table S9 in the Supplementary Material.

### 2.3. Demonstration program and data collection

#### 2.3.1. Site description

The demonstration program was carried out at a contaminated site located at a chemical reagent production factory in Tianjin, China (Fig. 2a). The factory was in operations from 1956 to 2007, and the demolition began at the end of 2009. Improper storage and disposals of chemicals during the operation resulted in severe soil and groundwater contamination. The contaminants of concern are volatile organic compounds, including chloroform, vinyl chloride, cis-1,2-dichloroethylene, 1,2-dichloroethane, 1,1,2-trichloroethane, trichloroethylene, tetrachloroethylene, 1,1,2,2-tetrachloroethane, and hexachlorobutadiene. There are residential areas east of the factory, a small park to the south, and a recently commissioned large hospital to the north. The site is planned for re-development as a residential area.

#### 2.3.2. Demonstration program for TCH-ISCO

The TCH-ISCO demonstration program covers an area of 70 m<sup>2</sup>, and the well layout consisted of seven heating wells, five pairs of injection wells (shallow and deep), and four temperature monitoring wells (Fig. 2b). The heating wells and injection wells were arranged both in two rows in an equilateral triangle with a distance of 3 m, the temperature monitor wells were arranged in a single row between the heating wells. The stratigraphic profile of the study area is shown in Fig. 2c, and the site is underlain by different layers of fills, which overlay on some clayey/sand silt, and the groundwater table was at approximately 0.9 m below ground surface (mbgs). The shallow injection wells were screened between groundwater table and 6 mbgs, and the deep injection wells were screened at 6–13.5 mbgs. The target temperature was set at 55 °C, and the oxidative reagent, sodium persulfate (PS), was injected once the target temperature was achieved. The operational period was three months, and a total of ~1000 m<sup>3</sup> of chlorinated aliphatic hydrocarbons (CAHs) contaminated soil and groundwater was remediated.

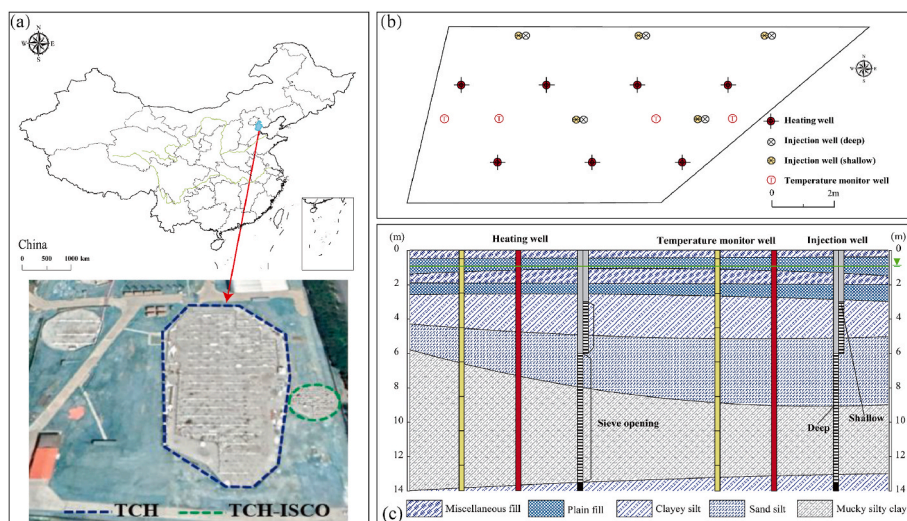


Fig. 2. Site information: (a) geographical location of the contaminated site; (b) layout of wells in the study area, and (c) stratigraphic profile of the study area.

In addition, a TCH remediation area was chosen to be used as a control, where the target temperature in the subsurface was 120 °C for thermal treatment. The TCH system included a heating control unit, an extraction system, an off-gas and tailwater treatment system, and a temperature monitoring system. The wells layout of TCH was consistent with that of TCH-ISCO, except that the injection wells was replaced by the extraction wells. The operational period was five months, and a total of ~44,800 m<sup>3</sup> of CAHs contaminated soil and groundwater was remediated.

It was assumed that both TCH-ISCO and TCH achieved the same degree of cleanup goals with the above-mentioned settings. In addition, it is worth noting that the current study focused on the sustainability of the TCH-ISCO, in comparison with that of TCH. The underlying remedial mechanisms, different factors affecting the effectiveness, and the impacts on soil and native microbial communities by TCH-ISCO are being evaluated in other on-going studies.

### 2.3.3. Data collection for TCH-ISCO

In this study, all the primary LCI data were collected from the TCH-ISCO and TCH demonstration program conducted in the field, and this is different from previous LCA studies in which the evaluation was carried out primarily based on various assumptions (Ding et al., 2022; Lemming et al., 2010a; Liu et al., 2022). Specifically, energy consumption was recorded by electricity meters, and the actual consumptions of steel, reagents, water and other construction materials and resources were recorded in detail in the construction log. Since most materials were sourced from the vicinity of the remediation site, the transportation distance of all materials was uniformly set at 50 km (Hu et al., 2011). Furthermore, large pieces of equipment used for remediation were transported directly from the site nearby, and as a result, their transportation was not considered in the LCI data compiling.

## 3. Results and discussion

### 3.1. Environmental impacts of TCH-ISCO and TCH

#### 3.1.1. LCA midpoint results of TCH-ISCO and TCH

For comparison purpose, the quantitative assessment on the environmental impacts was conducted for the application of both TCH-ISCO and TCH. The contributions of four LCA phases to each environmental midpoint indicator for TCH-ISCO and TCH are illustrated in Fig. 3a and b, and the values of the environmental impacts of each phase are listed in Tables S10 and S11, respectively. The operational phase of TCH-ISCO, the primary activity in the life cycle, contributed considerably to adverse

environmental impacts, including global warming (77.4%), fine particulate matter formation (75.3%), fossil resource scarcity (82.5%), water consumption (71.5%), and terrestrial acidification (76.2%). In comparison, the material acquisition and construction phase of TCH-ISCO dominated in human carcinogenic toxicity (61.6%), mineral resource scarcity (52.7%), terrestrial ecotoxicity (45.3%), and freshwater ecotoxicity (43.7%). Nevertheless, the environmental impacts of end-of-life activities, which only involved filling wells and transporting wastes, were rather limited. The general trends of these phases' contributions to the environmental categories for TCH is similar to those for TCH-ISCO. The major difference is that impact of remedial operation increased significantly and prevailed in all impact categories.

The contributions of various subordinate activities to each environmental midpoint category for TCH-ISCO are described in Fig. 4a. Specifically, electricity consumption for the well operation (40,698 kWh) accounted for 68.8% of CO<sub>2</sub> emission (50,700 kg CO<sub>2</sub> eq), 60.8% of fossil resources consumed (9740 kg oil eq), 60.2% of PM<sub>2.5</sub> produced (73 kg PM<sub>2.5</sub> eq), and 59.4% of SO<sub>2</sub> emission (160 kg SO<sub>2</sub> eq). This observation agreed with the experience and perceptions reported in previous studies (Ni et al., 2020). Based on the investigation locally, 80% of Tianjin's electricity in 2021 was produced from coal or oil-based power plants (Zheng et al., 2022). Therefore, the total amount of electricity consumption contributed considerably to greenhouse gas emissions and particulate matter formation. The well materials were the main sources of human carcinogenic toxicity (60.2%, 7290 kg 1,4-DCB) and mineral resource scarcity (51.2%, 325 kg Cu eq), due to the large amount of steel used in the construction of heating wells. Site construction influenced the freshwater ecotoxicity (37.1%, 1450 kg 1,4-DCB), terrestrial ecotoxicity (38.4%, 133,000 kg 1,4-DCB), and human non-carcinogenic toxicity (26.5%, 21,700 kg 1,4-DCB) significantly. These deductions due to the construction materials were ascribed to upstream traceability, and among which the electrical cable exhibited remarkably high ecological and human non-carcinogenic toxicities due to the rubber, asphalt, and insulation paint used. As an essential part of TCH-ISCO, the electrical cable used in TCH-ISCO was identified as a significant factor affecting the sustainability for the first time. The impacts of PS on global warming (8.1%, 5980 kg CO<sub>2</sub> eq), fine particulate matter formation (14.6%, 17 kg PM<sub>2.5</sub> eq), terrestrial acidification (16.2%, 43 kg SO<sub>2</sub> eq), and fossil resource scarcity (10.9%, 1750 kg oil eq) were much lower than those of electricity impact. However, the effects on freshwater ecotoxicity (25.8%, 1010 kg 1,4-DCB) and terrestrial ecotoxicity (30.5%, 105,000 kg 1,4-DCB) were higher than those of electricity consumption because PS production involved the emission of toxic by-products. Surprisingly, the result indicated that electricity

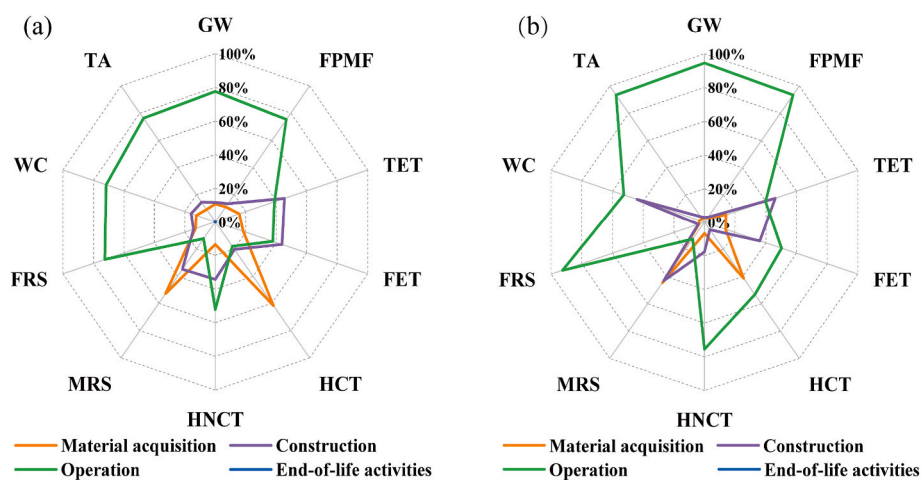


Fig. 3. Contributions of various LCA phases for (a) TCH-ISCO and (b) TCH. The acronyms are as follows: global warming (GW), fine particulate matter formation (FPMF), terrestrial acidification (TA), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), human carcinogenic toxicity (HCT), human non-carcinogenic toxicity (HNCT), mineral resource scarcity (MRS), fossil resource scarcity (FRS), and water consumption (WC).

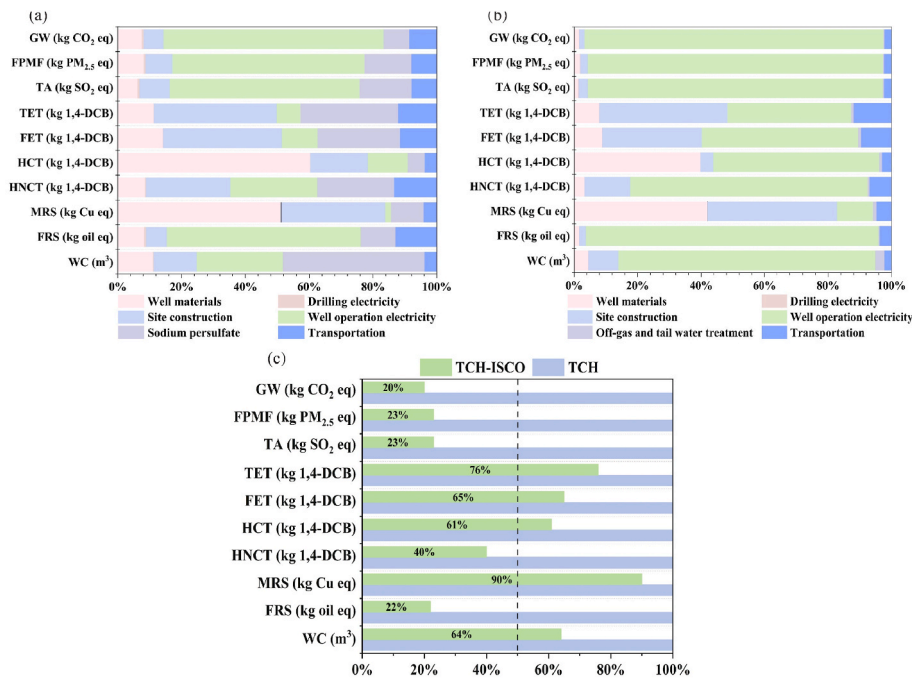


Fig. 4. Contributions of subordinate activities to each category for (a) TCH-ISCO; (b) TCH, and (c) comparison of the environmental impacts between TCH-ISCO and TCH.

consumption exhibited the main environmental impact for TCH-ISCO. In addition to the water required for well production and site construction materials (24.4%, 111 m<sup>3</sup>) and operation electricity (27.1%, 123 m<sup>3</sup>), PS production (44.3%, 202 m<sup>3</sup>) also significantly affected water consumption. In the life cycle of TCH-ISCO, the effect of transportation for materials was negligible.

The contributions of various subordinate activities to each environmental midpoint category for TCH are described in Fig. 4b. The impact of electricity consumption in the well operation (286,858 kWh) of TCH was higher in all categories than for TCH-ISCO because the heating temperature in TCH operation increased from 55 °C to 120 °C. Specifically, for a functional unit of “the remediation of 1000 m<sup>3</sup> of CAHs contaminated soil and groundwater”, the effects on global warming (94.1%, 356,495 kg CO<sub>2</sub> eq), fine particulate matter formation (92.9%, 520 kg PM<sub>2.5</sub> eq), terrestrial acidification (92.9%, 1130 kg SO<sub>2</sub> eq), human non-carcinogenic toxicity (74.7%, 159,000 kg 1,4-DCB), water consumption (80.9%, 890 m<sup>3</sup>), and fossil resource scarcity (92.1%, 68,800 kg oil eq) were significantly increased. Electrical cable and steel consumption of heating wells were the same as TCH-ISCO, therefore, there was no significant difference among the impact on terrestrial ecotoxicity (35.1%, 185,000 kg 1,4-DCB), freshwater ecotoxicity (31.4%, 3120 kg 1,4-DCB), human carcinogenic toxicity (39.7%, 10,800 kg 1,4-DCB), and mineral resource scarcity (42.1%, 89.9 kg Cu eq).

Fig. 4c reveals the overall comparison of different environmental impacts between TCH-ISCO and TCH. TCH-ISCO had a lower environmental impact on all categories than TCH. More specifically, TCH-ISCO used 85% less energy (40,698 kWh versus 286,858 kWh) than TCH, and released approximately 80% less CO<sub>2</sub>, SO<sub>2</sub>, and PM<sub>2.5</sub>, which reduced 60% of human non-carcinogenic toxicity. Furthermore, the impacts of freshwater ecotoxicity, terrestrial ecotoxicity, human carcinogenic toxicity, and water consumption were reduced by about 25%–35%, and the electrical cable consumption was the same for TCH-ISCO and TCH, while TCH-ISCO had extra reagent consumption. The impact of mineral resource scarcity was similar between TCH-ISCO and TCH, because both technologies consumed the same amount of steel, which was a primary factor in mineral resource.

### 3.1.2. Carbon emissions and flows of TCH-ISCO and TCH

In particular, reduction of carbon emissions is a priority for sustainable remediation (Hou et al., 2016), and it is important to assess the carbon footprint of major remediation activities so as to implement the carbon reduction plans (Liang et al., 2021). In this study, the overall comparison of carbon emissions between TCH-ISCO and TCH was quantitatively calculated for different phases and subordinate activities (Fig. 5a and b). The total carbon emissions for TCH-ISCO and TCH are 73,771 kg and 378,848 kg, respectively. The operational phase and heating well electricity consumptions were the main phase and activity of carbon emissions for both technologies. Specifically, the carbon emissions for the operational phase dominated the total emission values, accounting for 76% (56,065 kg) and 94% (356.117 kg), respectively. The carbon emissions from subordinate activity of well electricity consumption for TCH-ISCO accounted for 68% (50,164 kg), while in TCH it accounted for 93% (352.328 kg) of the total carbon emissions. The carbon emission for all other activities other than heating wells in TCH was less than 7%. However, for TCH-ISCO, the reduction in electricity consumption increased the proportions of carbon emission for other activities, such as transportation (11%, 8114 kg), well materials (7%, 5163 kg), and site construction (6%, 4426 kg).

### 3.1.3. LCA endpoint results of TCH-ISCO and TCH

The LCA endpoint impact results of TCH-ISCO and TCH is presented in Fig. S1. The ReCiPe endpoint impact was quantified using the kilopoint (kpt), a normalized indicator that can reveal the overall environmental impacts, which primarily consist of impacts on human health, ecosystem and resources. In general, a higher kpt value indicates a lower environmental impact (Zhang et al., 2022). Overall, the kpt score of the environmental impact for TCH was 3.4 kpt, one-quarter of 13.7 kpt for TCH-ISCO (Fig. S1a). TCH had a higher adverse effect on human health, ecosystems, and resources (Fig. S1b), among which human health was the main impact sub-indicator (Liu et al., 2022; Song et al., 2018).

### 3.1.4. Sensitivity analysis results

The influence trend of various scenarios on these two technologies was similar (Fig. 6a and b). Electricity consumption was the most influential factor, and the environmental impact would reduce by 25.4%

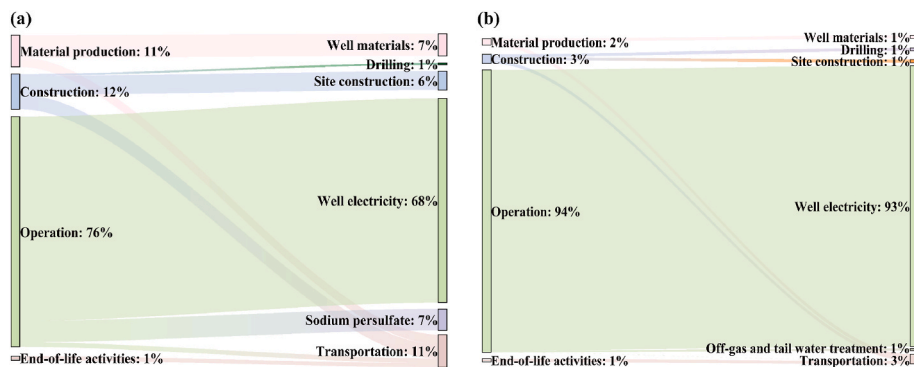


Fig. 5. Carbon flows between the four LCA phases and subordinate activities of (a) TCH-ISCO and (b) TCH.

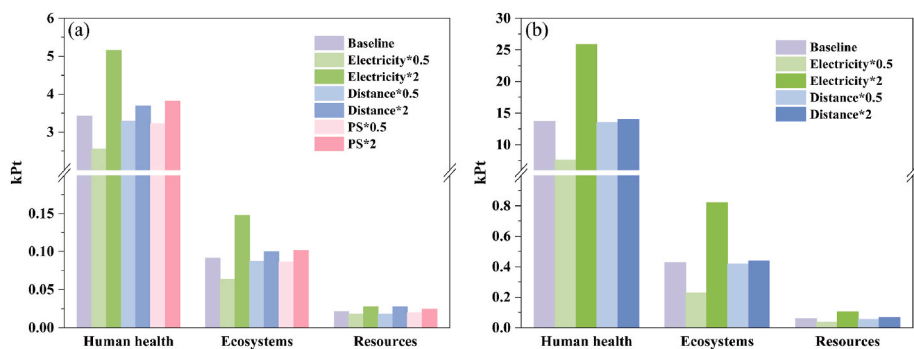


Fig. 6. Sensitivity analysis of (a) TCH-ISCO, and (b) TCH. Note that the baseline scenario refers to the evaluation using field data, and \*0.5 and \*2 scenarios refer to the evaluation using field data multiplied by 0.5 and 2, respectively.

and 44.6%, respectively for TCH-ISCO and TCH, when the electricity consumption was halved. In this study, the transport distance showed a minimal effect on the results since it was uniformly set at 50 km in LCI. The changes in carbon emissions in the various scenarios are illustrated in Fig. S2. When the electricity consumption was doubled, the carbon emissions of TCH-ISCO and TCH increased by about 68.8% and 94.3%, respectively. In contrast, other scenarios showed no significant impact on carbon emissions.

### 3.2. Economic and social impacts TCH-ISCO and TCH

The quantitative economic sustainability assessment, including sub-indicators for direct project cost, project risk for not being able to meet the remedial goals, remedial duration, the increase of land value and

employment opportunities, was conducted based on BMPs (Li et al., 2021b). The results for both TCH-ISCO and TCH are shown in Fig. 7a, with the specific scoring results listed in Tables S6–S8. It was demonstrated that the direct cost of TCH-ISCO was 562 ¥/m<sup>3</sup>, less than 1/3 of TCH (1874 ¥/m<sup>3</sup>), resulting in a 70% reduction of total remedial cost, which was primarily due to the lower target temperature of TCH-ISCO (55 °C) than that of TCH (120 °C). However, it is worth noting that the risk for not being able to meet the remedial goal was relatively higher for TCH-ISCO, because most studies on TCH-ISCO have been conducted only at the laboratory scale. On the other hand, TCH has been applied in the field at multiple contaminated sites for remediation, and a recent study reported that TCH has accounted for 30% of the ISTT remediation cases in the US (Horst et al., 2021). With respect to the remedial duration, the TCH-ISCO remedial operational duration was

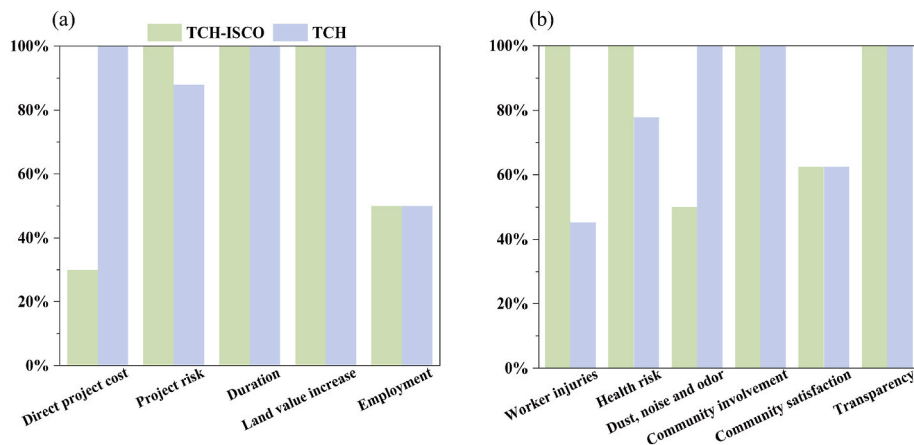


Fig. 7. Comparison results of (a) economic, and (b) social sub-indicators for TCH-ISCO and TCH.

three months, two months less than the operation period of 5 months in TCH because of the much higher target temperature. However, considering the potential rebound during long-term monitoring program associated with reagents in groundwater remediation (Liang et al., 2014; Sra et al., 2010; West and Kueper, 2012), the difference in remedial duration was considered insignificant between TCH-ISCO and TCH. Both technologies offered similar values for land value and employment sub-indicators, assuming that the requirements for re-development are met once the cleanup goal is achieved.

The social impact assessment, consisting of 6 sub-indicators of worker injuries, health risk for workers due to exposure to contaminants, disturbance of community due to dust, noise and odor, community involvement, community satisfaction, and project transparency, was carried out. The results are shown in Fig. 7b. The scores for health risk were higher in TCH-ISCO, mainly due to the application of corrosive PS, posing a potential risk to workers and ecosystem. However, it is believed that the safe use of PS can be achieved in the field with BMPs, and an example of such BMPs is that the safety awareness for workers should be adequately informed prior to their involvement in the remediation process, which is one of the key measures and management practices to ensure their safety (Hou et al., 2016; Song et al., 2018). On the other hand, TCH-ISCO outperformed TCH at reducing the impact of dust, noise, and odor to the community. Two factors contributed to the better performance of TCH-ISCO: (1) vapor extraction or multiphase extraction, a major source of dust and noise during remediation, was conducted, and (2) no off-gas was generated, thereby significantly reducing the impact on the air quality. The scoring results associated with community involvement and transparency were similar because all the relevant BMPs were implemented during the remediation of TCH-ISCO and TCH (Table S7), focusing on the number of people involved in the remediation (Liu et al., 2022) and the early organization of stakeholder meetings in the process (Cappuyens, 2016), respectively. However, it is worth noting that the health risk in the social impact and project risk in the economic impact can not be represented with the BMPs identified, suggesting that future effort be warranted for the development of BMPs to reduce the health risk and project risk activities.

### 3.3. Overall sustainability of TCH-ISCO and TCH

The results of environmental, social, and economic sustainability were normalized into the overall sustainability assessment scores through MCA (Jin et al., 2021). Table S9 lists the specific weight settings, calculation process and scores. Consistent with previous discussions, it was shown that the environmental sustainability performance of TCH-ISCO was much better than of TCH, however, TCH slightly outperformed TCH-ISCO with regard to the social aspect, whereas slightly underperformed regarding economic sustainability (Fig. 8a). The overall

sustainability scores of TCH-ISCO and TCH were 89.6 and 61.9, respectively (Fig. 8b), demonstrating that the sustainability performance of TCH-ISCO was significantly better than that of TCH.

## 4. Implications

Heating contaminated soils and groundwater to high-temperatures is energy intensive and, thus, a relatively expensive practice (Vidonish et al., 2016b), which can increase the environmental burden (Ding et al., 2019). The results of this study illustrated the feasibility of low-temperature TCH-ISCO to reduce energy consumption and improve sustainability while meeting the environmental cleanup goals. In addition, the LCA results showed that electricity consumption had more impact than reagent consumption in TCH-ISCO, being the key factor exerting the most impact on the environment. The sensitivity analysis results demonstrated that reducing electricity consumption by half would decrease the overall environmental impact of TCH-ISCO and TCH by 25.4% and 44.6%, respectively. Therefore, consistent with previous studies, reducing electricity consumption and changing the electricity source, can be the most effective ways to reduce the environmental burden (Chen et al., 2020a).

There are several ways to reduce the electricity consumption in the field implementations. First, the efficiency of heat utilization can be improved by optimizing the space among heating rods and their depths. Second, It was demonstrated that the optimal activation temperature of PS in organic contaminated sites was 50 °C (Huang et al., 2022), whereas the optimal activation temperature of ozone was 90 °C (Li et al., 2021a). Therefore, the selection of reagents and their corresponding optimal activation temperature is another important way to reduce the energy consumption. And third, it is important to develop more pioneering heating technologies, such as microwave and ultrasonic heating (Zheng et al., 2022), which can heat soil and groundwater more efficiently and uniformly. Peng et al. studied that when the temperature was 80 °C, the reaction time was 5 min, and 96% of phenanthrene was degraded by microwave heat-activated PS, contrary to 38% removal by conventional heat-activated PS (Peng et al., 2016). Microwave does show some levels of specific effects (non-thermal effect) on the decomposition of PS. Correspondingly, microwave can accelerate the decomposition of PS and generates higher levels of sulfate radical anions (Ergan and Bayramoglu, 2011; Peng et al., 2016), which can improve the removal efficiency. On the other hand, compared with thermal desorption alone, microwave coupled with thermal desorption can not only save energy but also reduce heating costs by 81% (Ding et al., 2019).

In addition, changing the electricity source to an alternative green energy source can reduce the cost and the carbon footprint (Zheng et al., 2022). Ni et al. evaluated that using aquifer thermal energy storage instead of electric heating for enhanced bioremediation and found that

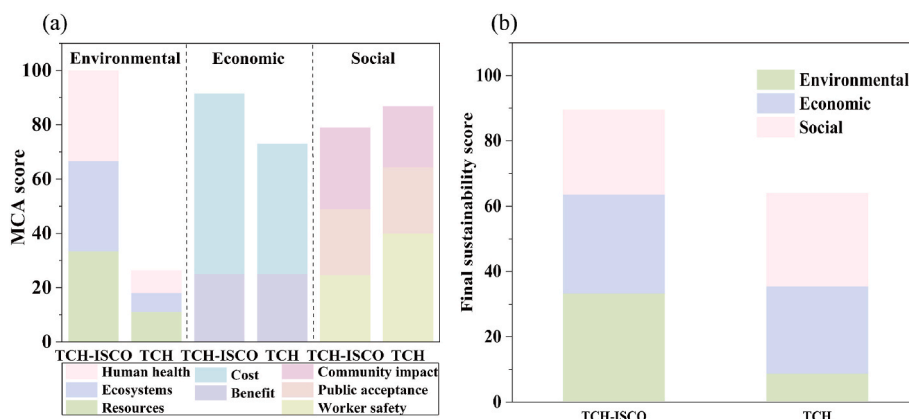


Fig. 8. Overall sustainability assessment results: (a) environmental, social, and economic MCA scores, and (b) final sustainability score results of TCH-ISCO and TCH.



the environmental impact was significantly reduced (Ni et al., 2020). Ding et al. and Visentin et al. found that Brazil would have a 30% lower environmental impact than China under the same scenario because Brazil derives most of its electricity from hydropower rather than coal-fired power (Ding et al., 2022; Visentin et al., 2019). There is a great potential for China to promote clean energy as it was reported that 69.7% of the electricity in China was produced from coal, and 27.4% from hydropower, wind power, and nuclear power in 2019 (Costa et al., 2019). Therefore, changing the power structure would be the one of the most efficient methods in optimization of TCH-ISCO and TCH.

## 5. Conclusions

In this study, a quantitative LCA-BMPs model was developed to assess the environmental, economic and social sustainability of TCH-ISCO and TCH. To the best of our knowledge, this is the first sustainability assessment for the research and development of an emerging technology using a set of comprehensive yet specific dataset collected in the field. The results showed that TCH-ISCO had a much lower environmental impact than TCH, and the energy supply via electricity contributed to 68% and 93% of the adverse environmental impacts, for TCH-ISCO and TCH, respectively. In addition, the carbon emissions and direct cost of TCH-ISCO were estimated to be 80% and 70% less than TCH. Furthermore, there are many challenges in incorporating GSR into practical site management, especially in developing countries where the remediation industry lacks experiences in GSR practices. This paper explored the relationship between BMPs and sustainability indicators in TCH-ISCO and TCH and could serve as a reference for developing countries to implement GSR strategies. The overall sustainability assessment score for TCH-ISCO was 89.6, outperforming that of TCH (61.9), demonstrating that the sustainability performance of TCH-ISCO was significantly better than that of TCH. However, it is worth noting that there are major application limitations for TCH-ISCO, including its difficulty to be applied in tight formations consisting of clay or silt, which is a niche for TCH applications. In addition, due to the usage of chemical reagent (e.g., persulfate), much attention is needed to monitor the potential secondary contamination issues, such as the increase of sulfate ions in groundwater, the decrease of pH, and the production of byproducts (e.g., vinyl chloride) of CAHs.

## CRedit authorship contribution statement

**Zongshuai Yang:** Conceptualization, Investigation, Methodology, Visualization, Writing – original draft. **Changlong Wei:** Investigation, Visualization, Supervision, Writing – review & editing. **Xin Song:** Resources, Conceptualization, Methodology, Supervision, Writing – review & editing. **Xin Liu:** Investigation, Writing – review & editing. **Zhiwen Tang:** Visualization. **Peng Liu:** Investigation. **Yunxiao Wei:** Investigation.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Xin Song republished by the National Key Research and Development Program of China.

## Data availability

Data will be made available on request.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2023.138732>.

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