



Screening of groundwater remedial alternatives for brownfield sites: a comprehensive method integrated MCDA with numerical simulation

Wei Li^{1,2} · Min Zhang^{2,3} · Mingyu Wang² · Zhantao Han³ · Jiankai Liu⁴ · Zhezhou Chen⁴ · Bo Liu⁵ · Yan Yan⁶ · Zhu Liu⁶

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Abstract

Brownfield sites pollution and remediation is an urgent environmental issue worldwide. The screening and assessment of remedial alternatives is especially complex owing to its multiple criteria that involves technique, economy, and policy. To help the decision-makers selecting the remedial alternatives efficiently, the criteria framework conducted by the U.S. EPA is improved and a comprehensive method that integrates multiple criteria decision analysis (MCDA) with numerical simulation is conducted in this paper. The criteria framework is modified and classified into three categories: qualitative, semi-quantitative, and quantitative criteria, MCDA method, AHP-PROMETHEE (analytical hierarchy process-preference ranking organization method for enrichment evaluation) is used to determine the priority ranking of the remedial alternatives and the solute transport simulation is conducted to assess the remedial efficiency. A case study was present to demonstrate the screening method in a brownfield site in Cangzhou, northern China. The results show that the systematic method provides a reliable way to quantify the priority of the remedial alternatives.

Keywords Brownfield sites · Groundwater remediation · AHP-PROMETHEE · Numerical simulation

Introduction

Brownfield sites are classified as land previously used for industrial or commercial purposes; are abandoned because of real or perceived contamination problems; and have the

potential to be reused once they are cleaned up (Hartmann et al. 2014; Rizzo et al. 2015; U.S. Environmental Protection Agency 2016a). Thus brownfield sites could be contaminated or suspected (potential) contaminated, and the authorities may probably take double attitude—love and hate—to them,

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- Highlights** • Human health risk assessment, MCDA method, and numerical simulation are integrated for groundwater remedial alternatives selection.
- An improved screening criteria framework is proposed and classified.
 - Criteria weights have strong impacts on ranking the priority scores of remedial alternatives.

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✉ Wei Li
liwei@szhec.gov.cn

¹ Shenzhen Academy of Environmental Sciences, Shenzhen, China

² College of Resources and Environment, University of Chinese Academy of Sciences, Beijing, China

³ Institute of Hydrogeology and Environmental Geology, Chinese Academy of Geological Sciences, Hebei, China

⁴ Beijing Institute of Hydrogeology and Engineering Geology, Beijing, China

⁵ School of Environment, Tsinghua University, Beijing, China

⁶ College of Environmental Science and Engineering, Liaoning Technical University, Liaoning, China

especially the contaminated sites. On one hand, contaminated sites have long been recognized as a potential health threat from a toxicological perspective (Bambra et al. 2014; Brombal et al. 2015), contaminants such as nitrogen, heavy metals, and volatile organic compounds (VOCs) are frequently detected in soil, surface water, and groundwater surrounding the sites which possibly pose negative impacts on local environmental system and the residents (Kaufman et al. 2005; Apostolidis and Hutton 2006; Cao and Guan 2007), and numerous studies have focused on the pollutants load in soil and groundwater of the old industrial zones (Krishna et al. 2009; Li et al. 2009; Li et al. 2013; Rahman et al. 2013; Ren et al. 2014), associated risk assessment for human health (Hough et al. 2004; Liu et al. 2010; Gong et al. 2012) and ecosystem (Cao et al. 2009; Ren et al. 2012; Wang et al. 2012; Cappuyens 2013); on the other hand, population growth and urbanization accelerate the multiple demands on land resources, redevelopment of brownfield sites is more encouraged by the professionals rather than using greenfields (Hartmann et al. 2014), and it is thus considered as an important means of pursuing sustainable urban development (Atkinson et al. 2014). However, inappropriate reuse of contaminated sites may lead to spread of contamination and impede future revitalization in urban areas (Swickard 2008; Ren et al. 2015). Consequently, remediation of contaminated sites is of great importance to the safety of environment and ecosystem as well as urban sustainable development, and it is challenging the world especially for developing countries like China (Güneralp and Seto 2008; Ren et al. 2014).

With rapid expansion of chemical production, industrial manufacture, and mining development in recent decades, the number of brownfield sites has significantly increased worldwide (Morio et al. 2013; Qian et al. 2014). In the whole European Union, it has been estimated that 3.5 million sites may be potentially contaminated and 1/7 of which were in urgent pollution (Steazar et al. 2013); in US cities, abandoned industrial sites were estimated to occupy 20,200 km² (Palta et al. 2014). Unlike the EU and the USA, the statement of “brownfield” is not used in China, and the contaminated sites in China are classified as “the suspected contaminated sites that exceeding the related soil quality standards assessed by the national technical guidelines of risk assessment” (the suspected contaminated sites are the sites that conducting (or conducted) industry production and management activities such as non-ferrous metal smelting, oil processing, chemical, coking, electroplating, tanning, and hazardous waste storage, utilization, disposal activities) (Brombal et al. 2015; MEP 2016). From 2001 to 2008 in China, the total amount of halted and relocated industrial factories and enterprises increased from 6.61×10^3 to 2.25×10^4 , leaving about 0.5 million contaminated sites (Liu et al. 2013; Brombal et al. 2015).

According to the National Soil Pollution Survey Bulletin released in 2014 (MEP and MIR 2014), 36.3% of 5846 soil monitoring point (surrounding 690 heavy polluting enterprises sites) and 34.9% of 775 soil monitoring point (surrounding 81 abandoned industrial sites) exceeding the national soil quality standard in China, and cadmium, mercury, arsenic, copper, lead, DDT, and polycyclic aromatic hydrocarbons (PAHs) are the major contaminants.

To clean up the hazardous waste sites in the USA, *Superfund* program was established and conducted by the U.S. Environmental Protection Agency since 1980 (U.S. EPA 2016a, b). The superfund cleanup process mainly consists of eight steps (Fig. 1), remedial investigation and feasibility study (RI/FS), which serves to characterize the site conditions, determine the nature of the waste, assess risk to human and the environment, and conduct treatability to screen and evaluate the potential performance of considered remedial technologies (U.S. EPA 1988), is vital to the success of a remediation project. The human health risk assessment model recommended by the U.S. EPA (1989, 1991a, b, 2001, 2004, 2009) is an effective evaluating tool implemented during the RI process and which has been used by the researchers worldwide such as Lee et al. (2006) in Korea; Muhammad et al. (2011) in Kohistan, Pakistan; Krichna and Mohan (2014) in Hyderabad, India; and Gao et al. (2011), Cui et al. (2013), and Li et al. (2015) in China. However, selection of remedial technologies is generally conducted by experts' subjective conduct without multiple criteria decision analysis (Betrie et al. 2013), and which would lead to discard of information (McDaniels et al. 1999; Betrie et al. 2013); moreover, further evaluation (like bench scale or pilot scale treatability studies) are needed to make detailed assessment of the considered remedial alternatives but which is restricted by time and cost (U.S. EPA 1988), thus a more adequate and cost-effective evaluation method is necessary for screening the remedial technologies. In China, the Ministry of Environmental Protection of China (MEP) approved the “Soil Pollution Prevention and Remediation Action Plan” in March, 2014, and technical standards and guidelines for site investigation, environmental monitoring, risk assessment, and site soil remediation were released in 2014 as well, calling for the strengthening of environmental management of contaminated sites (Xinhua 2014; CWR 2014; Brombal et al. 2015). However, capacity available in China in dealing with soil contamination remains limited, due to legislative, administrative, financial, and technical bottlenecks (Brombal et al. 2015).

Being an efficient approach to handle conflicting and both qualitative and quantitative objectives (Cinelli et al. 2014; Dehe and Bamford 2015), multiple criteria decision analysis (abbreviated as MCDA, hereafter) deals with problems whose alternatives are predefined and decision-makers rank

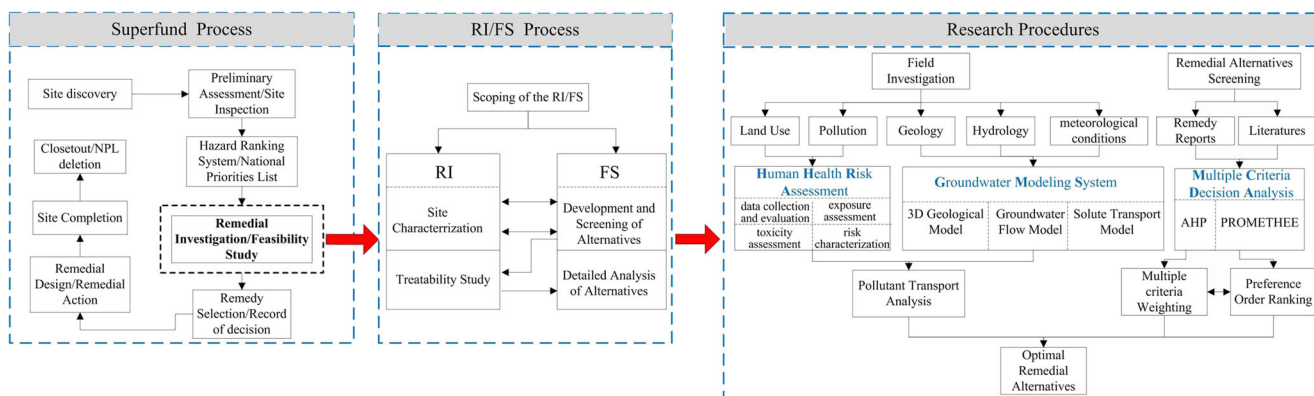


Fig. 1 Superfund and RI/FS process (U.S. EPA 1988) and the proposed remedial alternatives selection procedures in this study

available alternatives based on the evaluation of multiple criteria (Tefamariam and Sadiq 2006; Sadiq and Tefamariam 2009; Betrie et al. 2013). Among the most theoretical and empirically sound MCDA methods, there are AHP (analytical hierarchy process, Saaty 1987; Saaty and Vargas 2001), ELECTRE (elimination et choix traduisant la realite, Figueira et al. 2010), TOPSIS (technique for order preference by similarity to an ideal solution, Hwang and Yoon 1981), UTA (utilites additives, Jacquet-Lagrèze and Siskos 1982), DESYRE (DEcision Support sYstem for the REqualification of contaminated sites, Carlon et al. 2004), and PROMETHEE (preference ranking organization method for enrichment evaluation, Brans and Vincke 1985), and these decision technologies are widely used in multiple fields such as economics, construction, public administration, ecological, and environmental management (Chen 2006; Karasakal et al. 2009; De Moraes et al. 2010; Gorsevski et al. 2012; Hsu et al. 2015). For instance, Macary et al. (2014) used MCDA model based on ELECTRE for erosion risk assessment in an agriculture area; study conducted by Rahman et al. (2014) was attempted to analyze the regional environmental quality with spatial MCDA and remote sensing/geographical information system (RS/GIS); Deng et al. (2014) proposed AHP extended method for supplier selection; Tony et al. (2011) applied MCDA for health technology assessment; and Carlon et al. (2004, 2007), Critto et al. (2006), Critto and Agostini (2009), and Stezar et al. (2013) used DESYER for remedial alternative selection in Venice and Milan, Italy, and Romania. Groundwater modeling is a scientific tool for practical engineering, water resources protection, and decision-making process involved in groundwater management (Singhal and Goyal 2014; Sharma et al. 2015); with the help of powerful models like MODFLOW (USGS 2016), MT3DMS (Zheng and Wang 1999), and RT3D (Clement 1997), researchers around the world have studied and solved a great deal of environmental issues related to groundwater, such as surface water-groundwater interactions (Yihdego and Becht 2013; Roy et al. 2015),

evolution of groundwater depression cone (Zhang and Li 2013), groundwater quality deterioration and storage change affected by aquifer exploitation (Don et al. 2005; Zammouri et al. 2007) and climate/land use changes (Xu et al. 2011; Rasmussen et al. 2013; Yakirevich et al. 2013; Han et al. 2014), solute transport in the unsaturated zone, porous medium aquifer and karst aquifer (Reimann et al. 2011; Morway et al. 2013; Xu et al. 2015), groundwater pollution hazard assessment (Lima et al. 2015), groundwater resource sustainability (Eissa et al. 2013; Shao et al. 2013), groundwater pollution sources classification (Pizzol et al. 2015), and groundwater quality management strategy determination (European Union 2006).

In this present study, the RI/FS process (one step of conducting the U.S. EPA Superfund cleanup program) for assessment and selection of remedial alternatives is improved by introducing MCDA and numerical simulation methods. Based on the field investigation and human health risk assessment, AHP-PROMETHEE is used to screen and rank property of the suitable remedial alternatives according to multiple criteria (maturity of the alternatives, removal rate of contaminants and cost of construction and operation of remedial facilities, etc.). Meanwhile, the groundwater modeling system (GMS) is applied to build three-dimensional geological structure model to simulate and analyze the groundwater flow system by MODFLOW-2000 and predict the migration and transformation of contaminants by MT3DMS to evaluate the effectiveness and implementation of the remedial alternatives in the field quantitatively, and which is different from other decision tools such as DEYSER that assesses the residual risk by the percentage of reduction of the contaminant concentration, and the performance rates are typical of the specific remediation technology and the contaminant category (Carlon et al. 2004; Critto et al. 2006; Critto and Agostini 2009). Finally, the integrated method is used as a case study in the field of an abandoned chemical plant in Cangzhou, northern China. The research procedures are shown in Fig. 1.

Methodology

Human health risk assessment

The human health evaluation process developed by the U.S. EPA is generally used to assess the potential risk caused by the contaminants in the sites, and which consists of four main steps: (1) data collection and evaluation, (2) exposure assessment, (3) toxicity assessment, and (4) risk characterization (U.S. EPA 1989). Briefly, field investigation and monitoring analysis are both conducted firstly to identify the substances related to risk assessment and determine the magnitude, frequent, and duration of the exposures as well as the actual/potential exposed pathway; toxicity assessment is then implemented to confirm whether there exists the increase of adverse health risk caused by being exposed to the contaminants previously been identified, and quantitatively evaluate the relationship between the dose of the contaminants and the incidence of adverse health effects in the exposed population.

The adverse health effects are generally classified as chronic (non-carcinogenic) and carcinogenic (Krishna and Mohan 2014) and the substances are divided into groups according to the toxicological classification (IARC 2016). There are various pathways through which human are exposed to contaminants, such as ingestion, dermal adsorption, and inhalation (U.S. EPA 2001; Li et al. 2015); different calculating models are developed to conduct risk assessment through multiple pathways especially for the brownfield sites. For instance, a technical guideline for risk assessment in contaminated sites was established by the Ministry of Environmental Protection of the People’s Republic of China (abbreviated as MEP, hereafter) which included nine exposure pathways: ingestion of soil, skin-contact of soil, inhalation of soil particles, inhalation of gaseous pollutants of outdoor air from topsoil, inhalation of gaseous pollutants of outdoor air from subsoil, inhalation of gaseous pollutants of indoor air from subsoil, inhalation of gaseous pollutants of outdoor air from groundwater, inhalation of gaseous pollutants of indoor air from groundwater, and ingestion of groundwater (MEP 2014). In this study, the human health evaluation is used to assess the groundwater pollution status and determine the priority remedial contaminant.

Groundwater modeling system

Groundwater numerical modeling and simulation is generally developed to solve the issues related to groundwater resource management and protection (Yao et al. 2015). In this study, GMS, a well-known modeling software package developed by the environmental modeling research laboratory of Brigham Young University, is used to conduct groundwater flow and solute transport simulation to display and compare the implement effectiveness of the remedial alternatives implemented in the brownfield site, and

the main process can be divided into three steps. Firstly, regional three-dimensional geological model is constructed to recognize the stratum and aquifer structure of the brown-field site based on the field boreholes data. Secondly, a groundwater flow field model is developed according to the field investigation and geological model mentioned above by MODFLOW-2000 (Harbaugh et al. 2000), and the simulation model is adjusted till the simulated groundwater level results are in accordance with the monitoring results, which demonstrates the truthfulness and accuracy of the constructed simulating model. And finally, solute transport model is conducted to simulate the migration and degradation of contaminants under different scenarios (treated by variable remedial alternatives) by MT3DMS (Zheng and Wang 1999), and the variation of contaminants are comparable indicators for remedial alternatives screening and selection.

MCDA method and remedial alternative screening criteria

Preference ranking organization method for enrichment evaluation

Introduced and developed by J.P.Brans (Brans 1982), PROMETHEE is one of the most prevalent outranking techniques for MCDA, which encompasses of Promethee I~VI and focuses on not only ranking of alternatives, but also continuous cases, problems with segmentation constraints (Brans and Vincke 1985) and human brain representation (Brans and Mareschal 1992, 1995), etc. (Chen 2014). Among the Promethee family, Promethee I and Promethee II are well-known and widely used for partial ranking and complete ranking of alternatives, respectively (Corrente et al. 2014; Kilic et al. 2015).

To obtain the priority ranking of alternatives by Promethee method, preference functions, criteria, and the weight of each criterion are the main factors which should be determined by the decision-maker. Considering the case that a finite set of alternatives $A = \{a_1, a_2, \dots, a_m\}$ is evaluated by a set of criteria $C = \{c_1, c_2, \dots, c_n\}$. The degree of preference for alternative a_i over alternative a_j is calculated by the preference function $p_k(a_i, a_j)$ and represented as $p_k[c_i(a_i) - c_i(a_j)]$, $0 < p_k(a_i, a_j) < 1$, and six preference functions are used in Promethee method (Kilic et al. 2015). There is no preference of a_i over a_j when $p_k(a_i, a_j)$ equals to 0 and a strict preference when $p_k(a_i, a_j)$ equals to 1. The multiple criteria preference index $q(a_i, a_j)$ can then be calculated by

$$q(a_i, a_j) = \sum_{k=1}^n w_l \times p_k(a_i, a_j) \tag{1}$$

where w_l is the weight of criteria l previously determined. The outgoing and incoming flow (represented as $\Phi^+(a_i)$ and

$\Phi^-(a_i)$), indicating alternative a_i is outranking or outranked by other alternatives, are defined by

$$\phi^+(a_i) = \frac{1}{n-1} \sum_{x \in A} (a_i, x) \quad (2)$$

$$\phi^-(a_i) = \frac{1}{n-1} \sum_{x \in A} (x, a_i) \quad (3)$$

where $n = |A|$. In Promethee II, complete ranking of alternatives is determined by the net flow between the outgoing and incoming flows and calculated by

$$\phi(a_i) = \phi^+(a_i) - \phi^-(a_i) \quad (4)$$

The weights of remedial alternatives selection criteria are calculated by analytical hierarchy process in this study.

Analytical hierarchy process

Being used in almost all the applications related to decision-making process (Vaidya and Kumar 2006), AHP (Saaty 1980) is a structured MCDA approach which combines both qualitative and quantitative analysis. The primary principle/steps of AHP can be summarized as follows: first a hierarchic structure/network is proposed by recursively decomposing the decision problem, then the pairwise comparison matrix is constructed to establish the relative importance of the alternatives on criteria (scoring) and criteria among themselves (weighting) by the decision-maker(s) with a nine scale numerical rating (1 indicating equal importance and 9 extreme importance, intermediate values indicating increasing preference for one weight/alternative), and the priority weights of alternatives according to the pairwise comparison matrix can be calculated by following equation:

$$Aw = \lambda_{\max} w, \quad w = (w_1, w_2, \dots, w_n)^T \quad (5)$$

where A is a n dimensional comparison matrix, λ_{\max} is the largest eigenvalue of A , and w is the eigenvector corresponding to λ_{\max} .

Consistency index (CI) is defined to measure the inconsistency within the pairwise comparison matrix A in AHP and the consistency ratio (CR) is used to measure the degree of CI by the following equation:

$$CI = \frac{\lambda_{\max} - n}{n-1} \quad (6)$$

$$CR = \frac{CI}{RI} \quad (7)$$

where RI is the random consistency index and its value is related to the dimension of the matrix.

If $CR < 0.1$, the inconsistency degree of the comparison matrix A is considered to be acceptable and the eigenvector w is used as the weighting vector after normalization.

Otherwise, the comparison matrix should be adjusted and the process described above will conduct again until the consistency degree meets the need (Saaty 1980, 1987).

AHP is widely used by the researchers worldwide, it has the particular advantage of flexibility to be combined with different techniques like linear programming, quality function deployment, fuzzy logic, etc. (Vaidya and Kumar 2006). In this study, AHP is integrated with PROMETHEE and applied to determine the weights of criteria for remedial alternatives selection, the case study are discussed in “Remedial alternatives selection by AHP-PROMETHEE method” section.

Preliminary screening of the remedial alternatives and criteria framework construction

The groundwater remedial alternatives can be classified into categories according to the remedy implement of site conditions (in situ/ex situ) and the remedial target (the pollution source/the polluted groundwater) (U.S. EPA n.d.), the detailed description of remedial technologies being widely used currently can be found in the [supplement files](#). The alternatives are further evaluated on multiple aspects based on specific criteria, such as the long/short-term effectiveness and permanence, reduction of toxicity, overall cost of the remedial project, and so on, and which mainly focus on the following aspects: (1) overall cost of the remedial project; (2) pollutants removal efficiency; (3) associated risk to the residents, operated labors, and ecological environment throughout the remedial project; and (4) acceptance of the authorities and community. In this study, a criteria assessment system consists of 11 criteria is developed to evaluate the effectiveness and feasibility of variable remedial alternatives, and both of the criteria system developed by the U.S. EPA (1988) and the MEP of China (2014) are referred. The criteria system is illustrated in Fig. 2.

The U.S. EPA classified the criteria into three categories: threshold criteria (*overall protection of human health and the environment and compliance with applicable or relevant and appropriate requirement*, the minimum requirement that one alternative must meet); modifying criteria (*state acceptance and community acceptance*, which may be modified according to new information from state or community comments); and balancing criteria (the left five criteria which are used to assess the alternatives particularly) (U.S. EPA 1988). Some of the criteria are descriptive and not easy to be analyzed and determined quantitatively, the decision-maker(s)' assessment would probably depend on their own subjective judgment, which would increase the uncertainty of the selection, and it is where the difficulty of accuracy evaluation of the remedial alternative lies in. Therefore, the criteria for remedial alternatives selection are regrouped into three categories based on their analytic attributes in this study: qualitative, semi-quantitative, and quantitative criteria (Fig. 2). Qualitative criteria

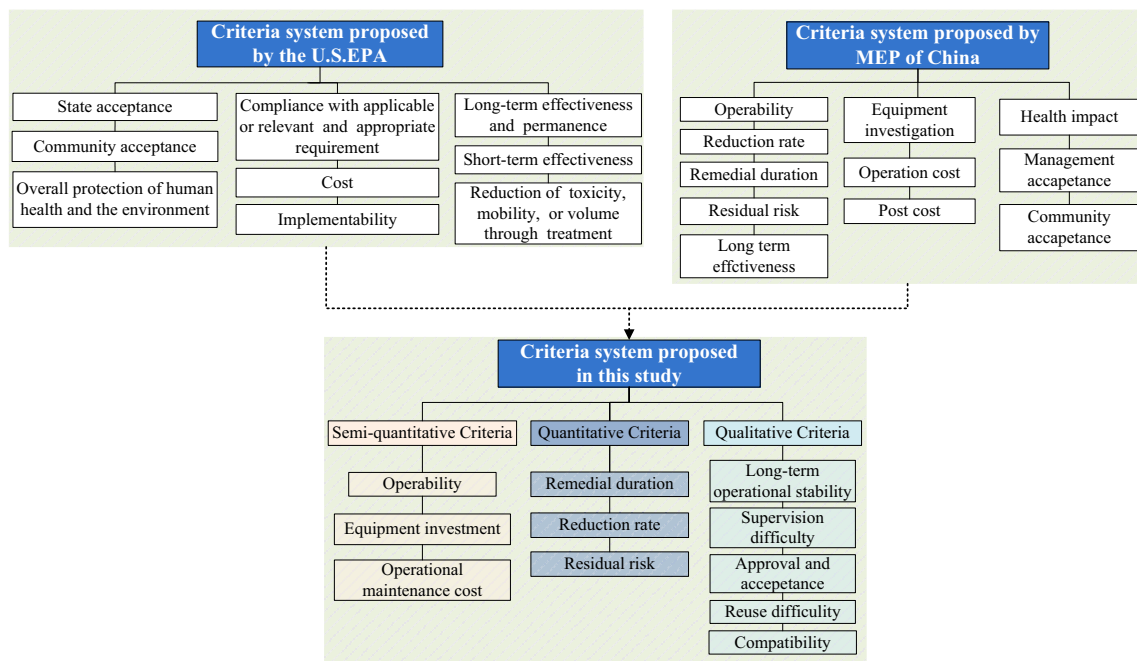


Fig. 2 Criteria framework proposed for remedial alternatives selection in this study

include *long-term operational stability*, *supervisor difficulty*, *approval and acceptance*, *compatibility*, and *reuse difficulty*, both of which are hard to determine by quantitative methods; in this study, crossing comparison and assumption are conducted among the alternatives and the expert’s opinion is considered to rank the scores; semi-quantitative criteria include *operability*, *equipment investment*, and *operational maintenance cost*, and the ranking scores of these criteria can be revealed by analysis of literatures and remedial reports associated to individual alternative; quantitative criteria include the rest three criteria related to the reduction of pollutants and which are determined by numerical simulation in this study. The weights of all the criteria are calculated by AHP method following the procedures mentioned above, and the comparison matrix is adjusted until the consistency degree of the criteria system meets the requirement. Besides the remedial effectiveness, the remedial duration and overall cost are two issues that the decision-makers are always concerned (Gavaskar et al. 2007; Critto and Agostini 2009); thus, sensitive analysis of these criteria is conducted. Based on the initial weights of the criteria calculated by AHP, another two scenarios of weights of the criteria, time-priority and cost-priority, are conducted to assess the sensitivity of the relative criteria. Time-priority scenario means the weight of remedial duration increased 20% and the weights of other criteria decreased proportionately while cost-priority scenario means the weights of equipment investment and operational maintenance cost increased 10% respectively and the weights of other criteria decreased proportionately. All the weights of the criteria framework are shown in Table 1.

Case study

Study area description

The study area is located in Cangzhou, northern China (Fig. 3), which is part of alluvial plain of the ancient Yellow River. The terrain of the study area is relatively flat with a slight slope from southwest to northeast, and the average elevation is about 15.9 m. The groundwater flow direction is mainly in accordant with the terrain and affected by irrigation seasonally. The area has a temperate semi-humid continental monsoon climate with an average receiver of 552.6 mm in rainfall which mainly occurs during June to September, and the average yearly evaporation is about 1252.4 mm. The boreholes data indicates that the shallow stratum in this area are composed of the Quaternary sediments, presenting a stratiform lithological structure of silty sand and silty clay, and two shallow aquifers can be recognized. The discontinuous phreatic aquifer consists of silt with a depth about 2~7 m and the thickness of which is about 0.5~2.9 m, and the confined aquifer consists of silt and silty sand with a depth about 12~16 m and the thickness of which is about 1.5~4.8 m.

The study area is mainly farmland with several villages spread around. A chemical plant which manufactured fluorescent whitening agents operated during 1990 to 2005 in this area, and was demolished and relocated in 2006, leaving an open pit (with the length, width, and depth about 208, 152, and 1.5 m, respectively) which was used to drain organic production wastewater during 1996 to 2000. The polluted soil and wastewater in the pit have been remedied by the local

Table 1 Weights of criteria of remedial alternatives selection determined by AHP method

Criteria weights	Operability	Equipment investment	Operational maintenance cost	Remedial duration	Reduction rate	Residual risk
Initial	0.107	0.113	0.040	0.045	0.202	0.197
Time-priority	0.085	0.089	0.032	0.245	0.160	0.156
Cost-priority	0.082	0.213	0.140	0.034	0.154	0.151
Criteria weights	Long-term operational stability	Supervision difficulty	Approval and acceptance	Reuse difficulty	Compatibility	
Initial	0.081	0.073	0.026	0.055	0.061	
Time-priority	0.064	0.058	0.021	0.043	0.047	
Cost-priority	0.062	0.056	0.020	0.042	0.046	

environmental authorities in 2011, but the contaminated groundwater is still a potential health threat to the residents nearby.

Samples collection and detection method

The sampling locations are shown in Fig. 3, field investigation and groundwater level monitoring were conducted in 2014 and 47 groundwater samples were collected (24 in July and

23 in December) from the drilling wells and local irrigation wells. The coordinates of the sampled wells were recorded by a portable GPS receiver; total dissolved solids (TDS), electrical conductivity (EC), dissolved oxygen (DO), pH, and temperature were tested on-site by a portable test instrument (YSI^{proplus}, USA). The groundwater samples were collected by high-density polyethylene containers for the test of major ions and heavy metals and by 1 L brown glass containers with PTEF caps for organic contaminants.

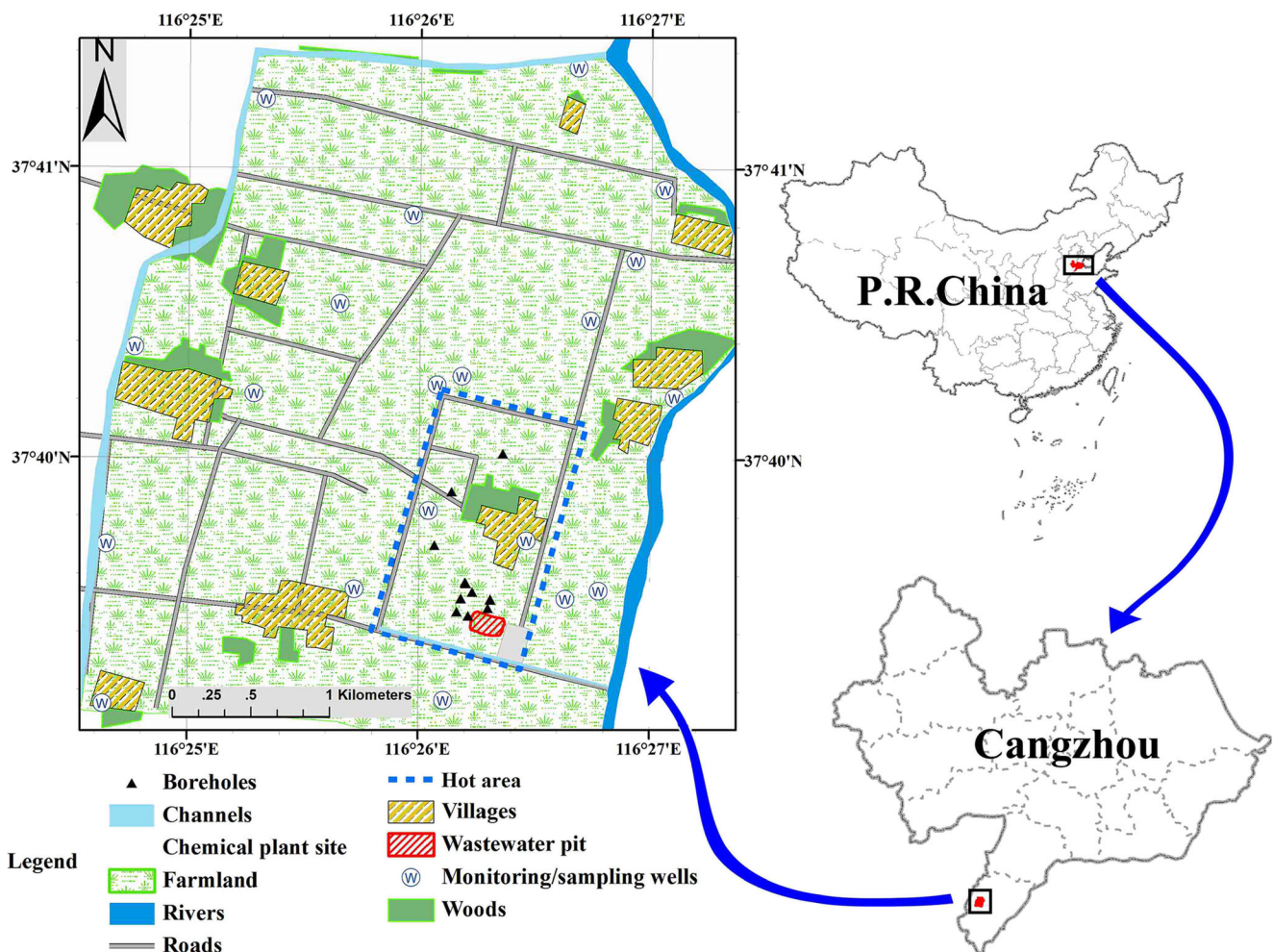


Fig. 3 Geographical location and the land use type of the study area

Table 2 Descriptive statistics of groundwater parameters in the study area

Value/parameters	Wells elevation (m, m.a.s.l.)	Sampled groundwater table (m, m.a.s.l.)	Temperature (°C)	pH
Min	15.1	7.2	14.0	6.2
Max	17.0	14.9	16.1	9.3
Average	15.9	13.6	14.9	7.3
Value/parameters	EC (μS/cm)	TDS (mg/L)	ORP (mV)	DO (mg/L)
Min	1898.0	1485.3	-176.9	0.9
Max	19,833.0	15,746.3	138.6	5.1
Average	5092.2	4079.4	19.5	2.5

The concentrations of contaminants in groundwater samples were determined by laboratory test. Fluoride, chloride, and sulfate were tested by ion chromatography (Dionex ICS-90, USA); VOCs were tested by gas chromatographic/mass spectrometry (Trace DSQ II, USA); and chromium, manganese, arsenic, ammonia-N, nitrate-N, and chemical oxygen demand (COD) were tested by spectrophotometer (Hach DR2800, USA). The test procedures followed the *Standard examination methods for drinking water* (MEP 2006).

Results and discussion

Samples analysis and human health risk assessment

The on-site test results of conventional groundwater parameters are summarized in Table 2. pH ranges from 6.2 to 9.3, with an average value of 7.3, indicating a neutral to alkaline condition of local groundwater; EC, TDS, and ORP show relative large variation, for instance, the maximum value of TDS is about tenfold of the minimum one. Moreover, relatively low ORP and high EC/TDS indicate anaerobic/reductive condition of groundwater in some sampling area, and which may probably results from pollution.

Groundwater quality test results show that the major detected contaminants in local groundwater can be divided into three types: inorganic contaminants (fluoride, nitrate-N, and arsenic), heavy metal (chromium), and organic contaminants (1,2-dichloroethane [1,2-DCA], o-dichlorobenzene [1,2-DCB], p-dichlorobenzene [1,4-DCB], and chlorobenzene [CB]), the statistical results are shown in Fig. 4.

Firstly the local groundwater quality is assessed based on the *quality standard for groundwater of China* (MEP 1993), the *standards for drinking water quality of China* (MEP 2006), and the *drinking water standards and the health advisories of the USA* (EPA 2012). The statistical results (Fig. 3) indicate that the mean concentrations of the most detected pollutants are within the water quality standards. However, the mean and maximum concentrations of 1,2-DCA are about 1~2 orders of magnitude larger than the water quality standards (36% of the samples exceed the MEP standards and the highest concentration is about 40-fold of the maximum contamination level), demonstrating serious groundwater organic pollution in the study area which may be attributed to the wastewater discharged into the pit in the early time. Moreover, high 1,2-DCA concentration detected wells distribute surrounding the wastewater pit and chemical plant site

Fig. 4 Statistical results of the primary detected pollutants in local groundwater (the black symbols) and the water quality standard in China and the USA (the color lines)

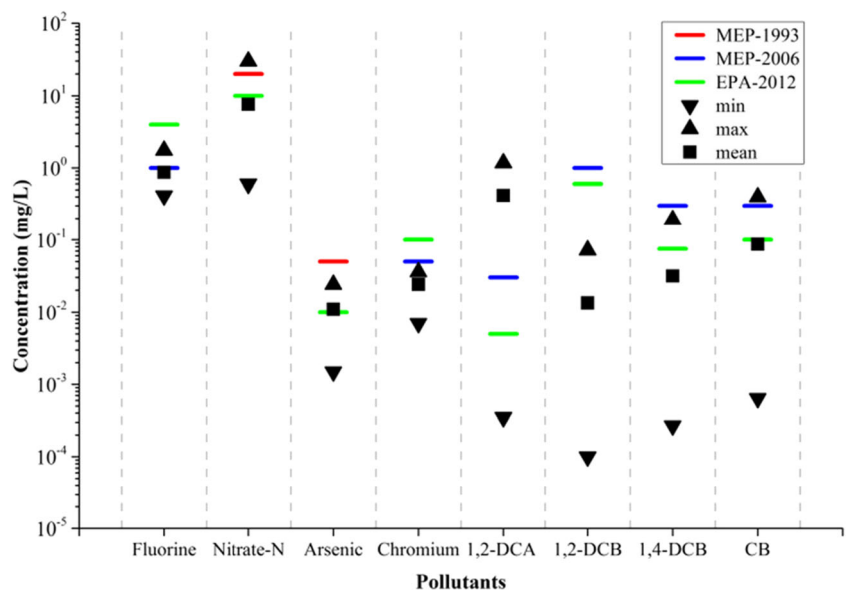


Fig. 5 Spatial distribution diagram of the detected concentrations of 1,2-DCA and the chemical oxygen demand in local groundwater, the blue lines represent the water table investigated in July 2014 (confined aquifer). The data interpolation was conducted by the inverse distance weighted method

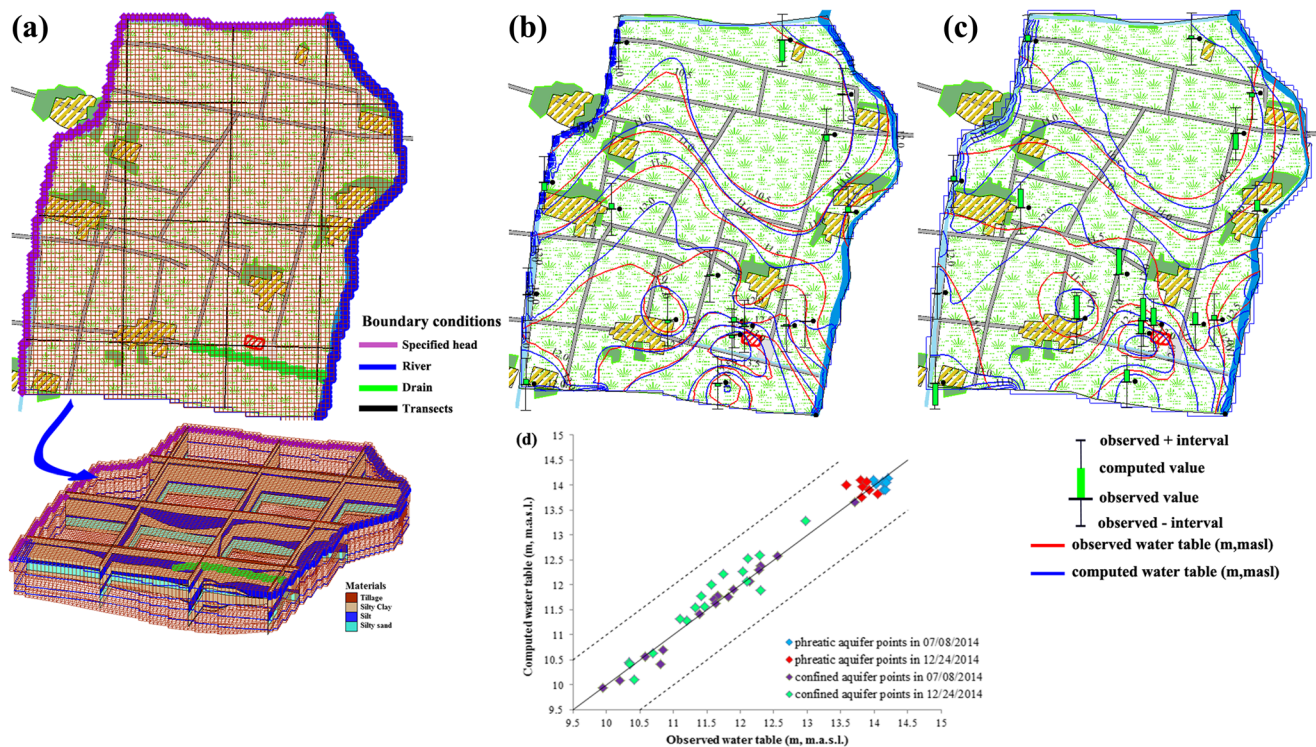
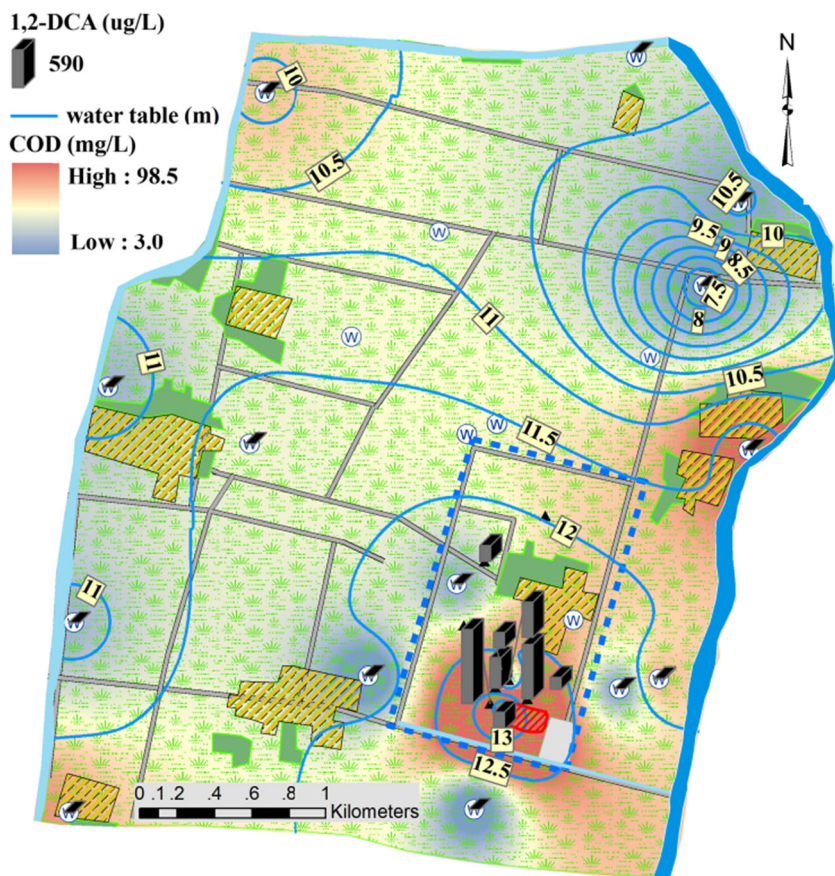


Fig. 6 Lithological structure, boundary conditions, grid division of the study area (a), the observed vs computed water heads in the confined aquifer (b, 08 July, 2014) and (c, 24 December, 2014), and the transient water head intervals (d)

(Fig. 5); the spatial distribution of which is in accordant with COD in groundwater, and shows a remarkable decreased trend with the increase of the distance between the sampling wells and the pit, which confirms the assumption of pollution mentioned above, and the organic substances in groundwater would be further evaluated to determine the potential threat to human health.

Human health risk is generally based on the quantification of the risk level in relation to two types of adverse effects: chronic/non-carcinogenic and carcinogenic effects (De Miguel et al. 2007; Li et al. 2015). According to the agents classification by the International Agency for Research on Cancer (IARC 2016) and the substances listed in the Integrated Risk Information System (IRIS 2016), the carcinogenic potential of the four substances detected in the site are ordered as follows: 1,2-DCA (possibly carcinogenic/probable human carcinogen) > 1,4-DCB (possibly carcinogenic/not assessed under the IRIS program) > 1,2-DCB (not classifiable as to its carcinogenicity/not classifiable as to human carcinogenicity) > CB (not classified/not assessed under the IRIS program). Therefore, 1,2-DCA and 1,4-DCB are conducted for the human health risk evaluation, and the maximum acceptable level for carcinogenic and non-carcinogenic risk is set as 1×10^{-6} and 1, according to the technical guideline for risk assessment in contaminated sites (MEP 2014).

Field investigation indicates that the shallow aquifer in the study area is mainly exploited for irrigation, which means the residents are exposed to the contaminated groundwater indirectly, thus the inhalation of gaseous pollutants of outdoor air from groundwater is ascertained to be the exposure pathway (MEP 2014) and the risk assessment is conducted. The results show that the carcinogenic risk of 1,2-DCA ranges from 2.85×10^{-10} to 9.64×10^{-7} , with an average value of 3.30×10^{-7} , and the carcinogenic risk of 1,4-DCB ranges from 9.10×10^{-11} to 6.47×10^{-8} , with an average value of 9.19×10^{-9} . The chronic/non-carcinogenic risk of 1,2-DCA and 1,4-DCB ranges from 2.26×10^{-5} to 7.55×10^{-2} and from 1.49×10^{-7} to 1.06×10^{-4} , and the average values of which are 2.61×10^{-2} and 1.50×10^{-5} , respectively. It should be concerned that the maximum calculated carcinogenic risk of 1,2-DCA reaches 9.64×10^{-7} , and which is very close to the maximum acceptable level; furthermore, being one kind of dense non-aqueous phase liquid (DNAPL), 1,2-DCA tends to migrate downward and penetrate the aquitard gradually once it enters the shallow aquifer (ITRC 2005), and which may probably polluted the deep groundwater that abstracted for domestic supply in the study area. Therefore, to maximally protect the local environmental system and the health of the residents, 1,2-DCA in groundwater is considered as the target contaminant that should be remedied and the cleanup alternatives are set as 30 $\mu\text{g/L}$ (with mean and maximum values of 1,2-DCA detected in the study site of 184 and 1170 $\mu\text{g/L}$,

respectively) based on *the standards for drinking water quality of China* (MEP 2006).

Remedial alternatives selection by AHP-PROMETHEE method

First, a lithological structure conceptual model of the shallow aquifer is conducted based on the boreholes and drilled irrigation wells data collected in the study area by GMS 7.1 and transferred into MODFLOW 2000 for groundwater flow simulation. The model covers the whole study area which is divided by $50 \text{ m} \times 50 \text{ m}$ grid horizontally and four layers vertically according to the stratum structure, lithology, and pumping test results: phreatic aquifer (layer 1), aquitard 1 (layer 2), confined aquifer (layer 3), and aquitard 2 (layer 4) (Fig. 6a). The irrigation channels in the west and north of the study area are set as specified head boundary condition, the river in the east and the drain in the south of the wastewater pit are set as river and drain boundary conditions. The hydraulic conductivity (HK), specific yield (SY), and storage (SS) are set divisionally according to the pumping test results and historical data collected in the study area for layers 1 and 3, the values of which range from 0.15 to 0.49 m/d for HK, 0.006 to 0.008 for SY, and 0.007 to 0.009 for SS, respectively. The recharge and discharge of the model mainly consist of precipitation, irrigation, lateral flow and evaporation, and river and drain discharge, and which are set in sub-regions monthly based on the monitoring results and land use types and controlled by the boundary conditions (Wang et al. 2015). The model is conducted by transient type flow and the whole simulating period is 6 months (started on 1 July and ended on 31 December) and the stress periods are divided monthly with 100 time steps for each stress period. The model is calibrated and adjusted by inverse parameter estimation (PEST module in GMS 7.1); 7 observed wells of the phreatic aquifer (layer 1) and 18 observed wells of the confined aquifer (layer 3) are monitored and the water head intervals are used to verify the accuracy of the simulation results, and the computed and observed water heads on 8 July and 24 December (Fig. 6b, c, close to the start and end time of simulation) are interpolated into contours. The simulated results shows similar groundwater flow direction and water heads with the observed results, and the water head intervals between observed and computed wells are very close (Fig. 6d), all of which are below 0.5 m, indicating the simulated results are accurate and acceptable, which can reveal the local groundwater flow and is further used for solute transport simulation.

Based on analysis of the literatures and remedy reports of field remedial projects (see in the [supplement files](#)), the applicable condition of different alternatives is summarized and improper alternatives are excluded and the results are summarized in Table 3.

Based on the preliminary screening of the alternatives mentioned above, monitored natural attenuation, bioremediation, and chemical reduction are considered more appropriate and the evaluation is conducted among these remedial

Table 3 Preliminary screening of alternatives conducting in the study site

Alternatives	Suitable	Not suitable	Reason
Solidification/stabilization		✓	Mainly used to remedy the pollutants in unsaturated zone (the vadose zone/soil) (U.S. EPA n.d.)
Permeable reactive barriers		✓	The aquifer in the study area mainly consists of silty clay and silt, of which the hydraulic conductivity and flow velocity are relatively small, leading a low efficient performance of PRB
Monitored natural attenuation	✓		Based on efficient monitoring, pollutants are degraded to acceptable levels by natural physical, chemical, and biochemical reactions, which is more cost-saving and successfully conducted in many brownfield in the USA (U.S. EPA 2016a, b)
Multi-phase extraction		✓	Mainly used for extract liquid petroleum products from groundwater (U.S. EPA n.d.)
Chemical reduction	✓		Hazardous compounds are converted to nonhazardous or less toxic compounds that are more stable, less mobile or inert by redox reactions (adsorption/precipitation of heavy metals), and which is suitable for most of the contaminated sites (U.S. EPA n.d.)
Air sparging		✓	Can volatilize the organic contaminants into the unsaturated zone, thus the exposure concentration of 1,2-DCA in the air and associated health risk caused by inhalation would probably increase in short period, and soil vapor extraction is required to combine with air sparging/thermal treatment, which would significantly increase the overall cost of remedial project (Gavaskar et al. 2007)
Thermal treatment		✓	
Pump and treat		✓	The remedial effectiveness would probably decrease and a tailing phenomenon appears with pumping relatively low concentrations of contaminants in the later period of remediation
Bioremediation	✓		1,2-DCA could be degraded by microbial reactions; the microorganisms break down it by using it as an energy source or co-metabolizing it with an energy source, thus it is suitable for the remediation of the study site

technologies; thus, three corresponding solute transport models are conducted to simulate the migration and degradation of 1,2-DCA in groundwater and the quantitative criteria (remedial duration, reduction of toxicity and mobility, reduction rate and secondary pollution risk) are calculated based on the simulation results. The degradation of 1,2-DCA at the site can be described by first-order kinetics reaction with following equation (Nobre and Nobre 2004):

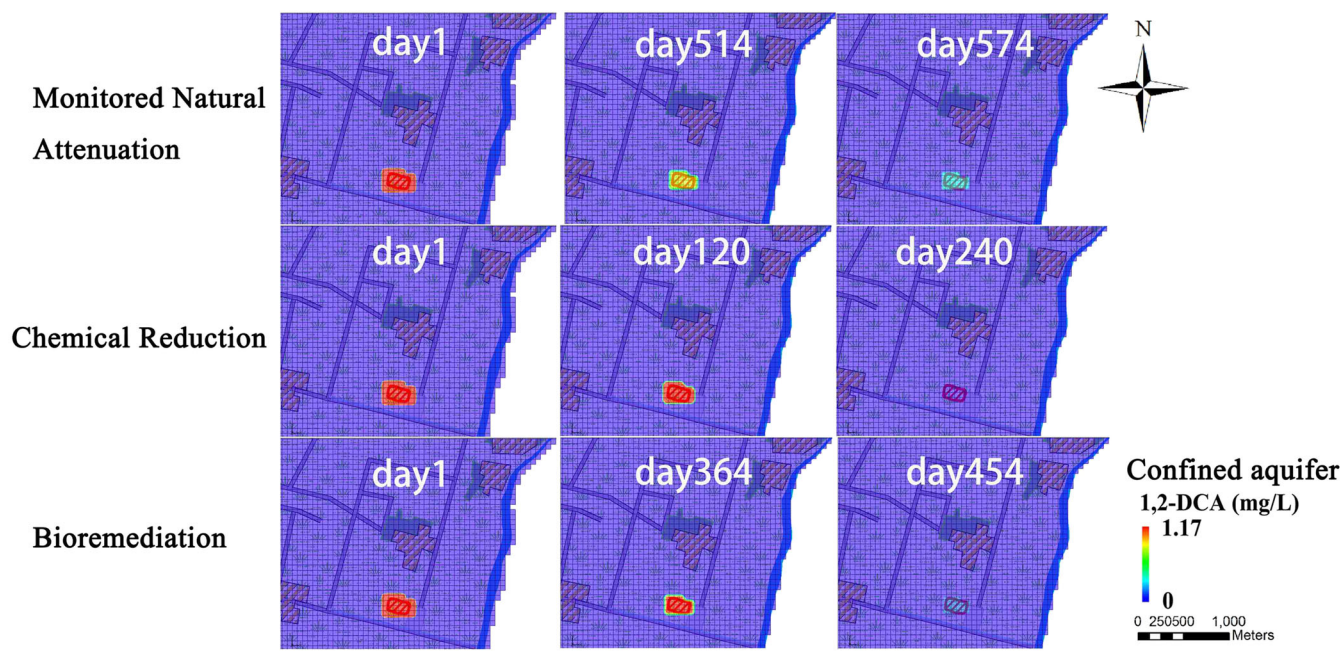
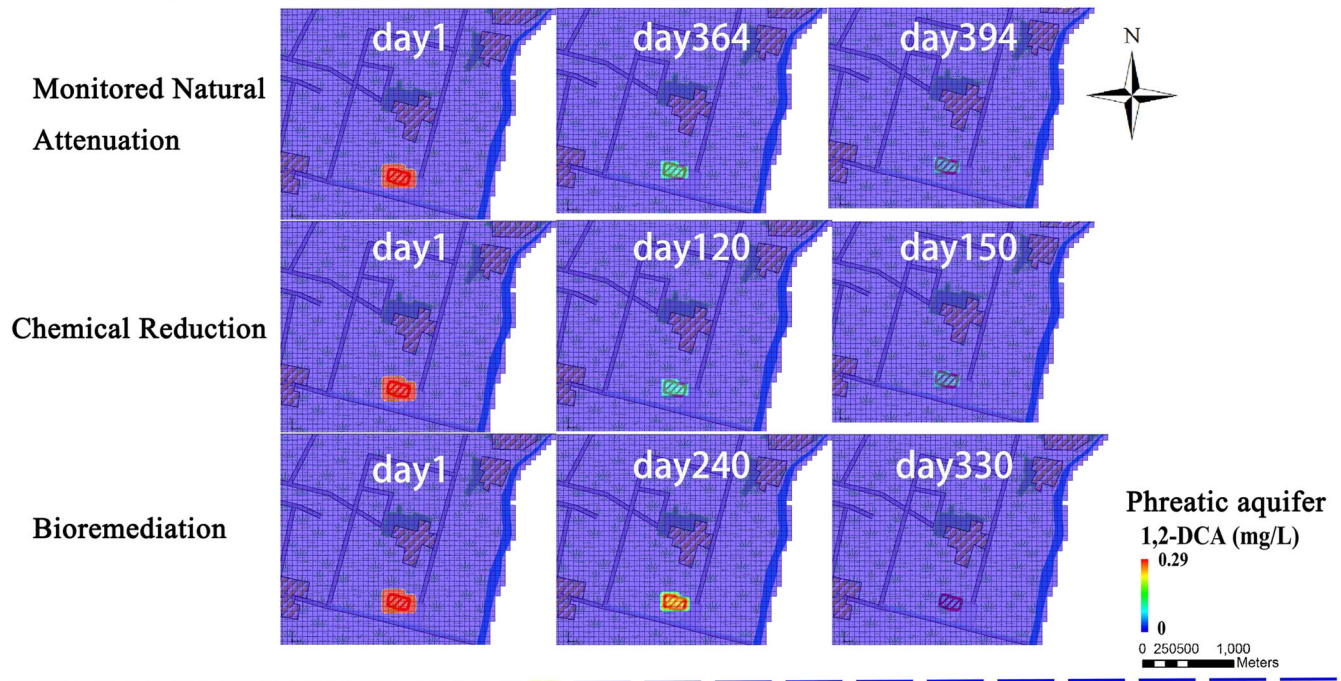
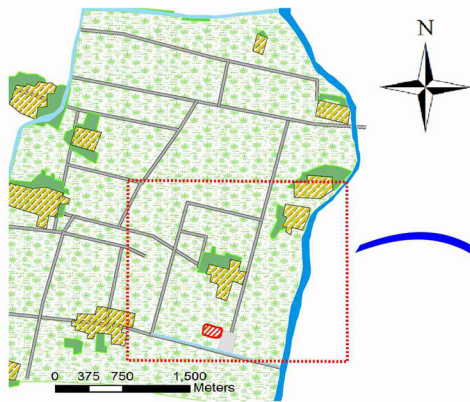
$$C = C_0 \times e^{-\lambda t} \quad (8)$$

where C is degraded concentration of the contaminant at time t , C_0 is the initial concentration of the contaminant, λ is the first-order decay coefficient and which can be expressed by

the chemical half-life $t_{1/2}$: $\lambda = \ln 2 / t_{1/2}$ (Eq. 9). The first-order decay coefficient of the contaminant at the site may be affected by variable factors and vary from individual to individual; in this study, the decay coefficients under monitored natural attenuation is calculated using Eq. 9 based on the concentration variation of 1,2-DCA in the nearest monitoring well of the wastewater pit (the sampling time interval of the phreatic aquifer and the confined aquifer are 696 and 412 days, and the 1,2-DCA concentration ranges from 585 to 6.3 $\mu\text{g/L}$ and 281 to 61.9 $\mu\text{g/L}$, respectively); the variation of 1,2-DCA concentration under chemical reduction is obtained by lab test (the contaminated water was sampled from the nearest monitoring well of the wastewater pit and the chemical reduction test

Table 4 Parameters used in the solute transport simulation

	Porosity	Bulk density (g/m^3)	Longitudinal dispersivity (m)	Starting concentration (mg/L)	Foc (%)	1st sorption const. (m^3/g)	Rate const. (dissolved/sorbed) (1/d)		
							Monitored natural attenuation	Bioremediation	Chemical reduction
Phreatic aquifer	3.50×10^{-1}	1.93×10^6	50	0.29	0.876	1.69×10^{-7}	2.29×10^{-3}	1.41×10^{-2}	3.74×10^{-2}
Confined aquifer	3.10×10^{-1}	1.92×10^6	50	1.17	0.720	1.69×10^{-7}			



◀ **Fig. 7** Concentration variation of 1,2-DCA vs time in the phreatic aquifer under different remedial conditions

was conducted using a glass sealed container with 1% H₂O₂ and a molar ratio of 1:100 as oxidant, the 1,2-DCA concentration on test day 0, day 1, and day 30 are 61.9, 54.3, and 15.2 µg/L, respectively) and the decay coefficient is calculated using Eq. 9; the decay coefficient under bioremediation is referred from literatures of field and laboratory test under different remedial conditions is used for solute transport simulation (Nobre and Nobre 2004; Arjoon et al. 2013; Huang et al. 2014), the primary parameters are summarized in Table 4.

The initial conditions for the three solute transport models are the same, which means the concentration of 1,2-DCA is set as the highest value that detected in local groundwater (0.29 mg/L for the phreatic aquifer and 1.17 mg/L for the confined aquifer) and cover the whole area of the wastewater pit, the solute transport model assumes that 1,2-DCA could be reduced immediately while the remedial alternative is conducted and degraded with the rate constant mentioned above. The whole simulation time for solute transport is set as 5 years (July 1, 2014 to July 1, 2019), the stress period for the first half year (July 1 to December 31, 2014) is set as 1 day, and the rest is set as 30 days (see the simulation results in Fig. 7).

Similar trend of 1,2-DCA concentration variation is observed in both the phreatic aquifer and the confined aquifer, the remedial alternatives show the order of remedial duration that monitored natural attenuation > bioremediation > chemical reduction, decrease of 1,2-DCA reaches the remedial target (below 10 µg/L) within 150 days under chemical reduction condition while the remedial duration of monitored natural attenuation is about 1~2 years (see in Fig. 7). Owing to the relatively higher groundwater flow velocity and lower 1,2-DCA initial concentration, the remedial alternatives show higher efficiency in the phreatic aquifer, the average remedial duration of which is 60% of the confined aquifer. The quantitative criteria scores are thus determined based on the simulation results; for instance, the remedial duration is the average value of the phreatic and confined aquifer, the reduction rate is calculated with the reduced mass of 1,2-DCA at the minim remedial duration, and the residual mass of 1,2-DCA indicates the residual risk. Meanwhile, the semi-quantitative criteria scores (operability, equipment investment, and operational maintenance cost) are referred and determined by the number of superfund sites (U.S. EPA n.d.) being remediated by the alternative and the related remedial reports (supplement files), and the qualitative criteria scores are determined by experts with 9-points scoring methods (the score ranks from 1 to 9, corresponding worst to best). The criteria scores are summarized in Table 5, the initial scores are further normalized by Eq. 10 and Eq. 11 to reduce the error caused by the differences of units and order of magnitudes among the criteria.

Table 5 Scores of remedial alternative selection criteria under different remedial conditions (normalized values are shown in the brackets)

	Semi-quantitative criteria			Quantitative criteria				Qualitative criteria			
	Operability (number of recorded remedial projects)	Equipment investment (\$)	Operational maintenance cost (\$)	Remedial duration (day)	Reduction rate (%)	Residual risk (%)	Long-term operational stability	Supervisor difficulty	Approval and acceptance	Reuse difficulty	Compatibility
Monitored natural attenuation	172 (0.09)	11.6 × 10 ⁴ (1)	17.4 × 10 ⁴ (1)	484 (0)	9.4 (0)	90.6 (0)	9 (1)	1 (1)	2 (0)	4 (1)	9 (1)
Bioremediation	298 (1)	35.8 × 10 ⁴ (0.22)	53.8 × 10 ⁴ (0.22)	392 (0.32)	17 (0.08)	83 (0.08)	6 (0)	3 (0.29)	5 (1)	7 (0)	4 (0)
Chemical reduction	159 (0)	174.6 × 10 ⁴ (0)	261.7 × 10 ⁴ (0)	195 (1)	100 (1)	0 (1)	8 (0.67)	8 (0)	5 (1)	7 (0)	6 (0.40)

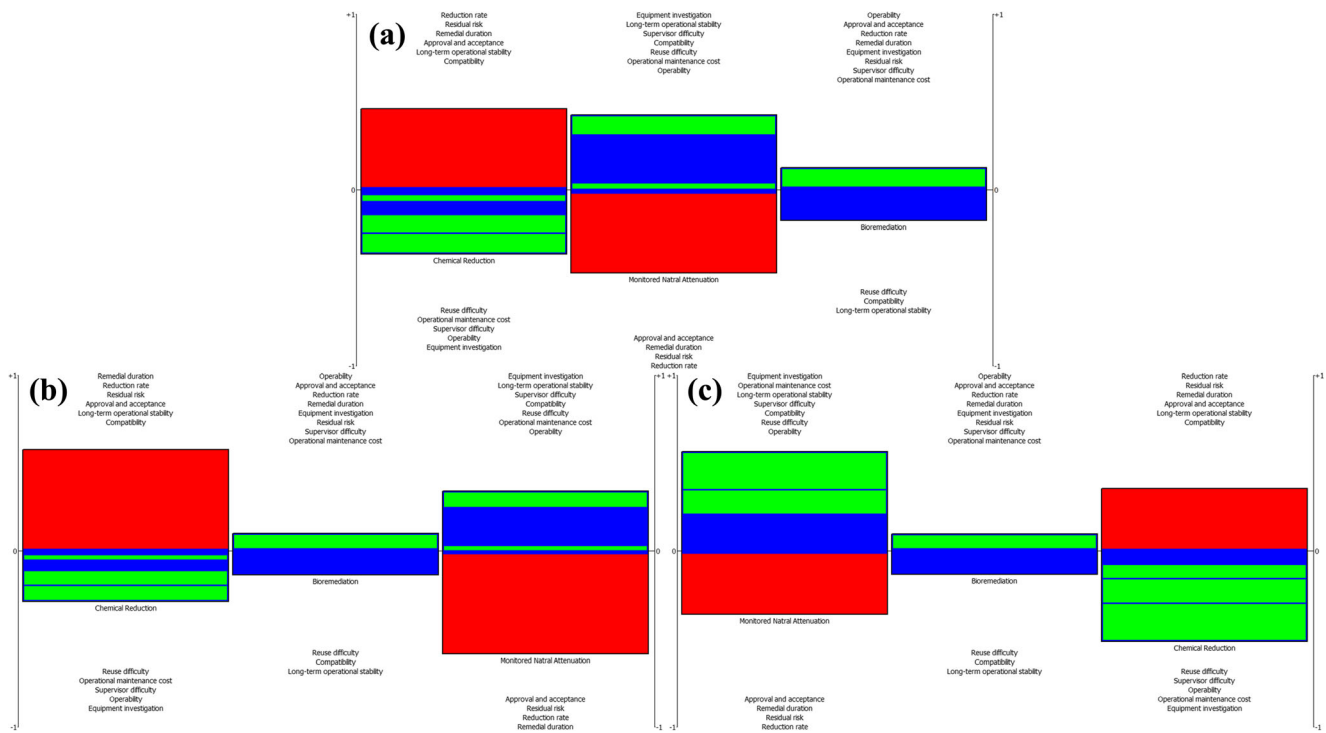


Fig. 8 Priority ranking scores under different remedial conditions (a, b, and c are ranking scores diagrams of initial criteria weights, time-priority weights, and cost-priority weights, respectively; the red/green/blue blocks

represent the ranking scores of quantitative, semi-quantitative, and qualitative criteria)

$$\bar{x}_i = \frac{x_i - \min(x_i)}{\max(x_i) - \min(x_i)} \tag{10}$$

$$\bar{x}_j = \frac{\max(x_j) - x_j}{\max(x_j) - \min(x_j)} \tag{11}$$

where x_i/x_j is the initial criteria scores and \bar{x}_i/\bar{x}_j is the normalized scores, the positive criteria is normalized by Eq. 10 (operability, reduction rate, long-term operational stability, approval and acceptance, and compatibility), and the negative criteria is normalized by Eq. 11 (equipment investment, operational maintenance cost, remedial duration, residual risk, supervisor difficulty, and reuse difficulty).

The weights of the criteria and the scores under different remedial conditions are further applied by PROMETHEE method to determine the priority ranking of the remedial alternatives, here Visual PROMETHEE (Mareschal 2015) is used to accomplish the priority ranking score calculation and the results are illustrated in Fig. 8.

The priority ranking scores of remedial alternatives with initial criteria weights follow the order: chemical reduction (0.10) > monitored natural attenuation (-0.04) > bioremediation (-0.05), indicating chemical reduction is more suitable than bioremediation and monitored natural attenuation for groundwater 1,2-DCA remediation in this site. The scores of quantitative and qualitative criteria of chemical reduction

(reduction rate, residual risk, remedial duration, approval and acceptance, long-term operational stability and compatibility) are positive while the semi-quantitative criteria scores (equipment investment and operational maintenance cost) are negative, indicating the advantages of chemical reaction on the aspects of remedial performance but higher remedial cost than bioremediation and monitored natural attenuation, and which are in corresponding with the simulation results and the sensitive analysis results of the criteria weights related to time cost and cost. On time-priority scenario, the ranking scores follow the order: chemical reduction (0.29) > bioremediation (-0.04) > monitored natural attenuation (-0.29), while on cost-priority scenario the ranking scores of these remedial alternatives show the opposite order (0.20 for monitored natural attenuation, -0.04 for bioremediation, and -0.16 for chemical reduction), indicating relatively strong impacts of the criteria on ranking the remedial alternatives especially for chemical reduction and monitored natural attenuation. It should be noticed that the solute transport simulation results are limited by the amount and accuracy of the input data, for instance, the reduction rate under monitored natural attenuation will probably be different with time, and the remedial rate would be overestimated by simulation. Some remedial projects reported that the groundwater remedial duration under monitored natural attenuation last for several or dozens of years (Yu et al. 2015), thus the screening results should be further assessed by lab test or pilot test.

Anyways, the method integrated MCDA with numerical simulation demonstrates the priority ranking of different remedial alternatives considering aspects involved economy, policy, and technology, and the criteria framework and weights could be adjusted and modified with the development of remedial technology and policy change, which help the decision-makers select and assess the remedial alternatives efficiently.

Conclusion

This paper proposed a systematic method which integrated human health risk assessment, numerical simulation, and MCDA for screening and assessment of brownfield site groundwater remedial alternatives. In briefly, filed investigation and human health risk assessment was conducted to determine the targeted remedial pollutants, and solute transport under different remedial conditions was simulated by GMS to evaluate the remedial effectiveness, MCDA (AHP-PROMETHEE method) was further applied based on the simulated results and experts' judges to rank the priority of the remedial alternatives, and this method was used in an abandoned chemical factory polluted site as a case study. The results indicated that local groundwater was polluted by 1,2-DCA which probably originated from the wastewater discharge of the chemical factory, and the human health risk under inhalation of gaseous pollutants from groundwater reached the risk threshold. Chemical reduction, bioremediation, and monitored natural attenuation were suitable for groundwater 1,2-DCA remediation of the site, the construction and operation cost of chemical reduction was highest while the total cost of monitored natural attenuation was lowest, the remedial duration by numerical simulation shown the opposite. The results calculated by PROMETHEE shown and ranked the priority of comparable remedial alternatives, and the ranking score of chemical reduction was followed by bioremediation and monitored natural attenuation. The framework and weights of the screening criteria could be modified and improved to fit the development of remedial technique and the change of policy, helping the decision-makers to select the remedial alternatives efficiently.

The presented methodology has been demonstrated in the case with a single contaminant is selected as target (1,2-DCA), and it is applicable in other cases if more contaminants may be of concern. In that cases, a cumulative risk should be considered (by risk assessment mentioned in “[Human health risk assessment](#)” section), the scores of qualitative criteria could be judged by the experts' options and the literatures and remedy reports of field remedial projects, the scores of the semi-quantitative and the quantitative criteria should be obtained by lab or pilot test for accurate results, and the effectiveness of the remedial alternatives could be revealed by the spatial and

temporal calculating results of the GMS owing to its powerful functions (which can simulate the multi-interactions occur in the aquifer), the research outcomes create a practical tools for the stakeholders working in the contaminated sites regeneration.

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Software availability

1. Name: Groundwater modeling system (GMS)
Developers: Aquaveo, LLC, Provo, Utah 84,604, USA
Operation system: Windows XP, Vista, 7, 8/8.1, 10
The latest versions: GMS 10.2.4 (released on March 2017)
Availability: Contact the developers (<http://www.aquaveo.com/downloads>.)
2. Name: Visual PROMETHEE
Developers: Prof. Bertrand Mareschal (bmaresc@ulb.ac.be), Solvay Brussels School of Economics & Management, Centre Emile Bernheim-Department of Quantitative Methods, Université Libre de Bruxelles, Belgium
Operation system: Windows XP, Vista, 7, 8 and later
The latest versions: Visual PROMETHEE 1.4 (Academic Edition, released on September 2013)
Availability: Contact the developers (<http://www.promethee-gaia.net/software.html>.)
3. Name: ArcGIS
Developers: ESRI, Inc. 380 New York Street, Redlands, CA 92373, USA
Operation system: Windows 7, 8.1, 10
The latest versions: ArcGIS 10.5 (released on December 2016)
Availability: Contact the developers (<http://www.esri.com>.)