Front. Environ. Sci. Eng. 2016, 10(6): 6
DOI 10.1007/s11783-016-0870-x

FEATURE ARTICLE

Engineering practice of mechanical soil aeration for the remediation of volatile organic compound-contaminated sites in China: Advantages and challenges

Yan Ma[1,2], Xiaoming Du[2], Yi Shi[2], Deyi Hou[3], Binbin Dong[1], Zhu Xu[2], Huiying Li[4], Yunfeng Xie[2], Jidun Fang[5], Zheng Li[2], Yunzhe Cao[2], Qingbao Gu[2], Fasheng Li (✉)[2]

1 School of Chemical and Environmental Engineering, China University of Mining & Technology, Beijing 100083, China
2 Department of Soil Pollution Control, State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
3 School of Environment, Tsinghua University, Beijing 100084, China
4 Beijing Key Laboratory for Risk Modeling and Remediation of Contaminated Sites, Beijing Municipal Research Institute of Environmental Protection, Beijing 100037, China
5 Shandong Key Laboratory of Eco-Environmental Science for Yellow River Delta, Binzhou University, Binzhou 256600, China

HIGHLIGHTS

- Engineering practice of mechanical soil aeration in China is reviewed.
- MSA is a cost-effective technique for VOC-contaminated sites.
- Limitations of MSA application have been summarized.

ARTICLE INFO

Article history:
Received 12 May 2016
Received in revised form 20 July 2016
Accepted 20 August 2016

Keywords:
Soil contamination
Volatile organic compound
Mechanical soil aeration
Engineering practice
China

GRAPHIC ABSTRACT

ABSTRACT

In recent years, many industrial enterprises located in the urban centers of China have been relocated owing to the rapid increase in urban development. At the sites abandoned by these enterprises, volatile organic compounds have frequently been detected, sometimes at high concentrations, particularly at sites abandoned by chemical manufacturing enterprises. With the redevelopment of sites and changes in land-use type associated with these sites, substantial amounts of contaminated soils now require remediation. Since China is a developing country, soil remediation warrants the usage of techniques that are suitable for addressing the unique challenges faced in this country. Land shortage is a common problem in China; the large numbers of contaminated sites, tight development schedules, and limited financial resources necessitate the development of cost-effective methods for land reclamation. Mechanical soil aeration is a simple, effective, and low-cost soil remediation technique that is particularly suitable for the remediation of large volatile organic compound-contaminated sites. Its effectiveness has been confirmed by conducting laboratory studies, pilot tests, and full-scale projects. This study reviews current engineering practice and developmental trends of mechanical soil aeration and analyzes the advantages and disadvantages of this technology for application in China as an emerging soil remediation market. The findings of this study might aid technology development in China, as well as assist other developing countries in the assessment and implementation of cost-effective hazardous waste site soil remediation programs.

1 Introduction

Volatile organic compounds (VOCs) are frequently detected organic contaminants in soils and groundwater. They are defined as organic chemicals that have a saturated vapor pressure greater than 70.91 Pa at room temperature or have a boiling point of less than 260°C, and are represented mainly by the benzene series and halogenated hydrocarbons. Because of their diversity, characteristic volatility, toxicity, lack of obvious color or odor at toxic

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✉ Corresponding author
E-mail: lifs@craes.org.cn

concentrations, tendency to accumulate in confined spaces, and adverse effects on the ozone layer, VOCs have been listed as high-priority contaminants in many countries [1–3]. VOCs are primarily generated in industries such as petroleum, chemical engineering, printing, building material, painting and coating, and machinery [4,5]. Owing to their widespread use and hazardous nature, regulations in many countries require that soils contaminated with VOCs be appropriately treated to ensure biologic and environmental safety.

With rapid urbanization in China, large numbers of companies in oil, chemical, coke, and other industries are being relocated, leaving a large number of abandoned industrial sites contaminated with VOCs. It is estimated that approximately 86,000 companies, including chemical, metallurgy, petroleum, transportation, and light manufacturing companies, closed down or relocated during the period 2001–2007. Thus, adopting effective, widely applicable, and cost-effective techniques has become essential for soil remediation at abandoned urban industrial sites [6,7].

In developed countries, VOCs have been controlled using various sophisticated technologies such as soil washing, soil vapor extraction, high-temperature incineration, thermal desorption, bioremediation, and bioventing [8–13]. Although widely used, these technologies have certain shortcomings. For some of these technologies, there is limited experience of their use on a large scale. Soil washing is relatively expensive and might only be suitable for coarse soil matrices. Soil vapor extraction requires a long treatment period, and contaminant concentration might rebound once the system is turned off. Bioremediation methods tend to be useful for sites with low levels of contamination, since microbial activities might be inhibited by high levels of toxic contaminants; therefore, they are less applicable at large-scale, highly contaminated, and highly heterogeneous sites found in China.

Mechanical soil aeration (MSA), as a remediation method suitable for VOC-contaminated sites, has many advantages, including being highly effective and rapid, and having a broad applicability for site-specific variations in soil quality and VOC concentration [14–16]. In addition, MSA offers simplicity and is less expensive than more capital-intensive methods such as thermal desorption or incineration. The US Environmental Protection Agency (EPA 2007) found that MSA provided high efficacy and success rates [17]. In China, MSA has been shown to be effective for the remediation of sites contaminated by volatile chlorinated hydrocarbons at the laboratory and pilot scales, and also in full-scale site projects [18–23]. Thus, MSA has been found to be an efficient and economical ex-situ remediation technique for VOC-contaminated sites at scales ranging from laboratory scale to full scale.

In this study, we review the strengths and challenges associated with using MSA for the remediation of VOC-contaminated sites in China. We assess the principles, processes, and present research status of this technique, as well as its cost-effectiveness, in the context of urban reclamation projects in China. In addition, we consider the secondary challenges associated with the method, including worker health and safety, and the environmental challenges involving processing, transportation, interim storage, disposal of removed toxics, and subsequent use or disposal of decontaminated soils. This study summarizes the experience gained with MSA and provides theoretical and technical support for future decision-making and sustainable use of this technology in China and other developing nations.

2 Characteristics of VOC-contaminated sites in China

As mentioned previously, many abandoned industrial sites in urban areas are contaminated by VOCs. Contaminated sites in China possess unique characteristics owing to significant differences in the history, levels of economic development, and regional characteristics between China and developed Western countries. There are expected to be many more contaminated sites in China than in most other countries. Existing contaminated sites were mainly used for mechanical processing or agrochemical and petrochemical industries. Industrially contaminated sites are mainly located in Chongqing, Jiangsu, Hubei, Beijing, Shandong, Hebei, Fujian, and other economically developed areas that are heavily populated [24]. The major contaminants from the mechanical processing industry are monocyclic and polycyclic aromatic hydrocarbons, whereas those from the agrochemical industry are phenols, monocyclic and polycyclic aromatic hydrocarbons, halogenated aliphatic hydrocarbons, and halogenated aromatic compounds, and those from the petrochemical industry are monocyclic and polycyclic aromatic hydrocarbons, and petroleum hydrocarbons [24,25]. Overall, the most common contaminants are aromatic and halogenated hydrocarbons such as 1,2-dichloroethane, trichloroethylene, benzene, and carbon tetrachloride [16,26,27]. Many of these compounds are highly toxic or carcinogenic and pose serious health and environmental risks when present in the air, soil, or groundwater [28]. The scope and scale of these sites pose serious technical and infrastructural challenges. VOCs disperse easily in soil, which further widens the contaminated area and complicates remediation efforts [27]. VOCs can also leach into groundwater, posing a contamination risk. Under certain conditions, some volatile organic pollutants exist as non-aqueous liquids at high concentrations, and these may be retained long-term in the soil and become new sources of pollution. High VOC concentrations in the soil can pose a high risk to construction workers if the soil is excavated and disturbed. In 2004, for example, construction workers at Songjiazhuang station on Beijing Subway Line 5 were intoxicated by semi-volatile pesticides from contaminated soil [25].

China has only recently begun to systematically remediate contaminated sites, and at present, only a small number of large-scale remediation projects have been conducted. The main techniques employed for VOC-contaminated sites include thermal desorption, MSA, and soil vapor extraction. In recent years, Chinese authorities have paid increasing attention to soil pollution control [29]. A current bottleneck in this regard is the lack of equipment and monetary resources available for large-scale projects [30]. Given the high demand for available land in urban centers and the large number of contaminated sites requiring remediation, it is particularly important to develop cost-effective soil remediation technologies characterized by safe operation, high throughput, low energy consumption, and high VOC removal efficiency.

3 Principles of MSA

MSA, also known as Ambient Temperature Desorption, is a remediation technique used in China for removing VOCs from contaminated soils. To remove contaminants by using MSA, mechanical stirring is used to mix soil aggregates so that low-boiling volatile contaminants are more rapidly emitted into the air. The released volatiles are treated by gas exchange and adsorption, and are discharged until the air quality passes pollution standards [14,15,21].

MSA involves several steps, which are summarized in Fig. 1. First, the contaminated soils are excavated, moved to temporary enclosed greenhouses, and piled into windrows. The soils are then stirred at regular time intervals by using heavy earth-moving equipment, and subsequently, the released volatiles are collected and treated with activated charcoal filters. Precautions are taken to prevent secondary contamination and to protect the health of workers. When testing verifies that the concentration of contaminants in the treated soil is reduced below the target concentration, treated soils are removed from the greenhouses and used for backfill or other purposes. Particulates from the contaminated air generated during mixing are first removed by settling and filtration, and then VOCs are removed by adsorption through activated carbon, or incinerated and discharged after the concentration of the toxic species drops below established thresholds [14,16,18]. The greenhouse itself is constructed using a single-framed design, and enclosed in a multi-functional gas impermeable membrane, secured with a ring girder around the foundation of the structure, which maintains the overall seal. The building is fitted with aluminum fixtures, blowers, check valves, hoses, and the control devices needed to collect, move, and monitor the accumulated toxic gases and force them out through the activated carbon filters [22].

Because VOCs are released in their gas phase from soils, different sizes of pores formed between soil particles act as diffusion channels for these gases [31]. The composition and size of soil particles directly affect the pore size in soil [32,33]. When larger soil particles or aggregates are present in soils, a greater number of larger pores are formed; when smaller soil particles or aggregates are

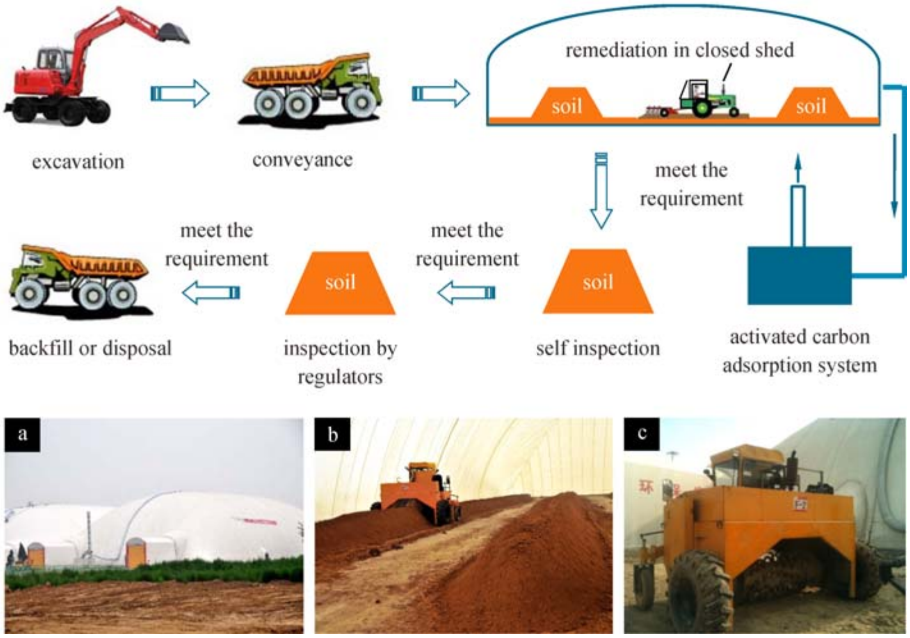

Fig. 1 Technological process of remediation with mechanical soil aeration. Closed sheds at a remediation field (a) and stirring devices at a remediation field (b and c)

present, the pores in soils are relatively small (Fig. 2). In such porous media, the pores between soil particles are occupied by soil vapor and soil water to form channels with different sizes and connection patterns. The gaseous contaminants migrate along channels, and eventually evaporate from the soil matrix. With the same water content, the size and number of pores in a porous medium are important factors that influence the diffusion characteristics of volatile contaminants in soils.

The size and structure of soil particles have a great impact on the migration of contaminants [34]. The existing forms of the organic contaminants in soil particles are shown in Fig. 2(b) [35,36]. Organic contaminants usually occur in various forms in soil particles. The gas phase contaminants often occur in the pores of soil particles. They adsorb on the soil particles via the distribution of vapor/solid phases and diffuse into the liquid phase of soils via the distribution of vapor/liquid phases, and eventually enter other areas by gas transportation and steam convection. The liquid and non-aqueous phase liquid phase of contaminants can dissolve in soil water, volatilize into soil air, or enter pores by vapor diffusion. Since the pore size and distribution of soil particles are not uniform, mobile phases with vapor phase, water phase, or vapor/water coexistence phase form in the large pores. Even when soil water content is low, smaller pores are usually occupied by static liquids because of the capillary effect, making it difficult for contaminants dissolved in soil water to volatilize from soil particles. Therefore, the concentration of contaminant residuals in soil increases.

4 Engineering practice of MSA

VOC-contaminated sites are of great concern throughout China. According to incomplete data (Table 1 and Fig. 3), MSA is considered an efficient technology that is currently being adapted or developed for the remediation of VOC-contaminated sites in the major provinces of China, such as Beijing, Jiangsu, Shandong, Hubei, and Liaoning. Analysis of data from nine completed soil remediation projects in China suggests that MSA is being extensively used. These sites were formerly used by the chemical industry. During remediation, a large volume of contaminated soils was identified at these sites, ranging from 17,474 m^3 to 1,530,000 m^3, and the largest five sites accounted for

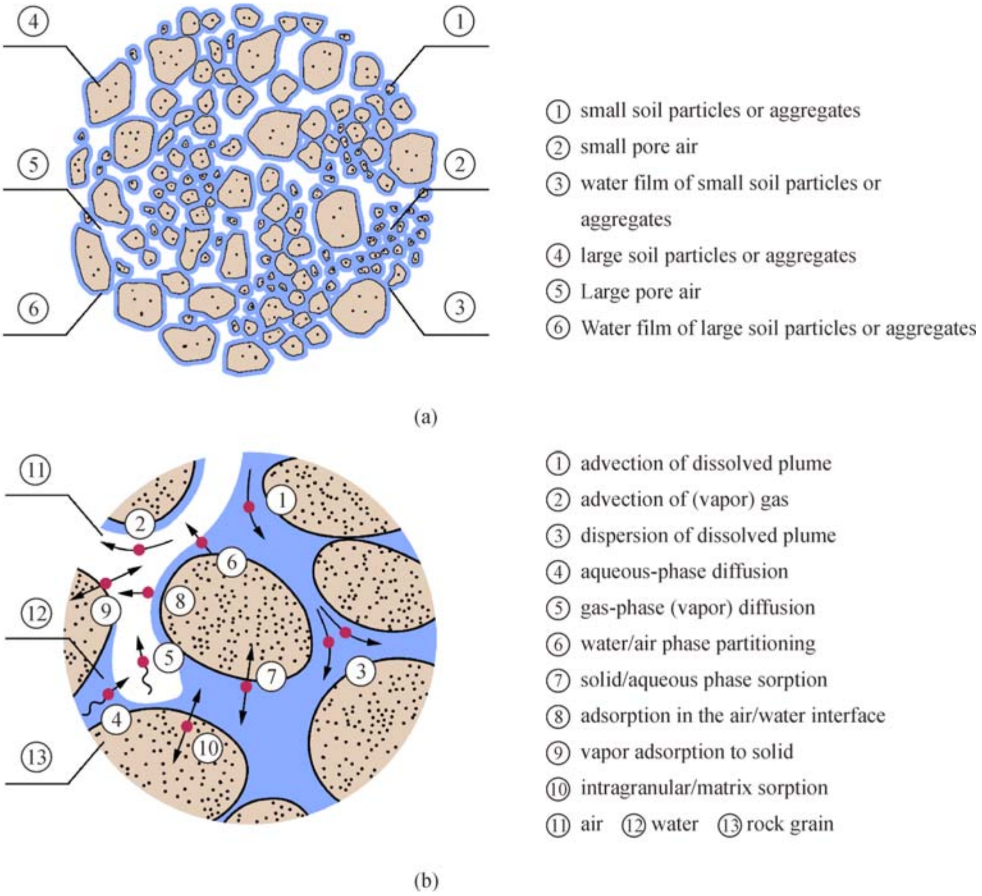

Fig. 2 Conceptualization of soil particles and pores (a) [32,33] and volatile organic compounds (VOC) in the unsaturated zone (b) [35,36]

Table 1 Characteristics of typical contaminated sites in China at which mechanical soil aeration (MSA) was used or partially used for remediation

provinces	main contaminants in the soil	soil remediation technologies	total volume/m^3	volume by MSA/m^3
Liaoning (S1)	heavy metals, PAHs [a], and BTEX [b]	MSA, soil washing/flushing, and chemical oxidation	279290	5721
Hubei (S2)	heavy metals, VOCs [c] (aniline, benzene, and chlorobenzene, etc.) and SVOCs [d] (Benzo [a] anthracene)	MSA, solidification/stabilization, and chemical oxidation	376000	57000
Shandong (S3)	VOCs (benzene, chloroform) and SVOCs (hexachlorobenzene, benzex, and DDT)	MSA and cement kiln incineration	17474	7863
Jiangsu (S4)	PAHs [a], BTEX [b], chlorinated hydrocarbons, and organic phosphorus pesticides	MSA, thermal desorption, and chemical oxidation	245742	116081
Beijing (S5)	PAHs [a] and benzene	MSA-thermal desorption	1530000	1144200
Beijing (S6)	VOCs (1,2-dichloroethane, chloroform, trichloroethylene, etc.)	MSA	57500	57500
Beijing (S7)	VOCs (chloroform, dichloromethane, and benzene)	MSA	334000	334000
Beijing (S8)	VOCs (1,2-dichloroethane, chloroform, benzene, etc.)	MSA	416100	416100
Beijing (S9)	VOCs (1,2-dichloroethane, chloroform, benzene, etc.)	MSA	1230000	1230000

Notes: a) PAHs = polycyclic aromatic hydrocarbons; b) BTEX = benzene series; c) VOCs = volatile organic compounds; d) SVOCs = semi-volatile organic compounds

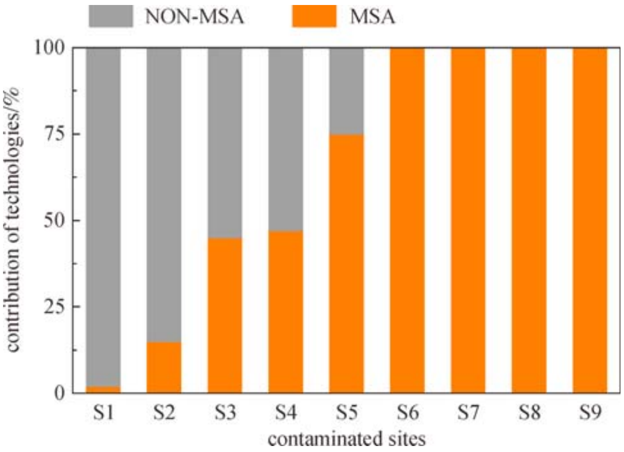

Fig. 3 Percentage of contaminated soil treated using mechanical soil aeration (MSA) at nine sites in China

over three million cubic meters of contaminated soils. The total volume of soils treated by MSA reached 3.4 million m^3 (including 1.1 million m^3 soils by the joint remediation technology of mechanical soil aeration-thermal desorption), accounting for 75% of the total amount of treated contaminated soil. In four of the five remediation sites in Beijing, MSA was used exclusively. As one of the oldest industrial bases in China, Beijing is currently the cultural, political, and educational center of China with a population of over 21,000,000, and is regarded as one of the typical and more developed cities in China [37]. Therefore, there is a great urgency to use land to its full potential and repurpose obsolete industrial sites [30]. At present, numerous sites in Beijing require decontamination and VOC removal. As the main pollutants in the other five contaminated sites included not only VOCs but also semi-volatile organic compounds (SVOCs) and heavy metals, MSA was combined with other soil remediation technologies for their removal. For example, at a dyestuff plant in Jiangsu Province, MSA and thermal desorption were used to remediate soil contaminated by VOCs and SVOCs, respectively, and the total volume of soils treated by MSA accounted for 47% of the total amount of treated contaminated soil.

The use of MSA in Western countries is rarely reported. For instance, the US Superfund program only reported four cases of MSA application in ex-situ source zone treatment, whereas some other technologies, such as incineration and solidification/stabilization, have been used in over 100 cases [38]. This may be because the Superfund program only addresses the most toxic sites, for which MSA may not be a suitable technology. MSA may have been used for less toxic sites that were addressed by voluntary cleanup programs; however, application data for such programs are often not published. Moreover, as an ex-situ remediation technique, MSA is associated with both excavation costs and increased risk of secondary contamination during soil excavation, transportation, temporary storage, stacking for examination, and other processes [39]. Owing to these concerns, alternatives to MSA, such as soil vapor extraction and land farming, are more often used in western countries. Although the soil vapor extraction technique is currently one of the most commonly used remediation techniques for volatile contaminants in the world, it has some limitations. It is very sensitive to the hydrogeological environment of the whole field and is only effective for unsaturated soils with relatively high permeability. The effects of vapor extraction on layered soils or low permeability soils are uncertain. Land farming is an above-ground remediation technology that reduces the concentration of contaminants present in soils through processes associated with bioremediation, and involves the excavation of the contaminated soil, which is similar to MSA using mechanical aeration devices [13,39,40].

5 Advantages of MSA

MSA is characterized by simple operation, excellent performance in removing volatile contaminants, low energy consumption and low cost over a wide range of VOC contaminant levels. An MSA reclamation project can be completed in a relatively short period. Because of these attributes, MSA is ideal for use in China, particularly in urban centers that have numerous VOC-contaminated sites. In addition, mechanically stirred soils are uniform and satisfy the physical and chemical standards of reclamation projects. As China is developing its remediation equipment manufacturing capability and is continuously improving process management, MSA has become an efficient and economical technology for the remediation of VOC-contaminated sites.

The data from a field site at a large abandoned domestic chlor-alkali chemical company were analyzed to confirm the advantages of MSA. This remediation site had a test processing capacity of 1,000–1,500 $m^3 \cdot h^{-1}$, a stirring frequency of 12 times$\cdot$day^{-1} and required 5–7 days for the remediation. In all, 1,109 soil samples were collected, including 95 control samples, and 507,000 m^3 of soil was treated. The performance of contaminant removal from soil at this site is shown in Table 2 and Fig. 4 [18,21,22]. The concentration of contaminants in the soil at the site was significantly reduced to levels substantially below the target concentration thresholds for site remediation. Reduction of toxic VOCs ranged from 17% for 1,1,2,2-tetrachloroethane to more than 99% for 1,2-dichloroethane (1,2-DCA). Overall, the concentration of seven of the 11 compounds tested was reduced to below their detection limits, and the percentage of test samples that showed reduction of contaminants below the required thresholds was above 90%, indicating successful remediation of VOC contamination at this site by using MSA [18].

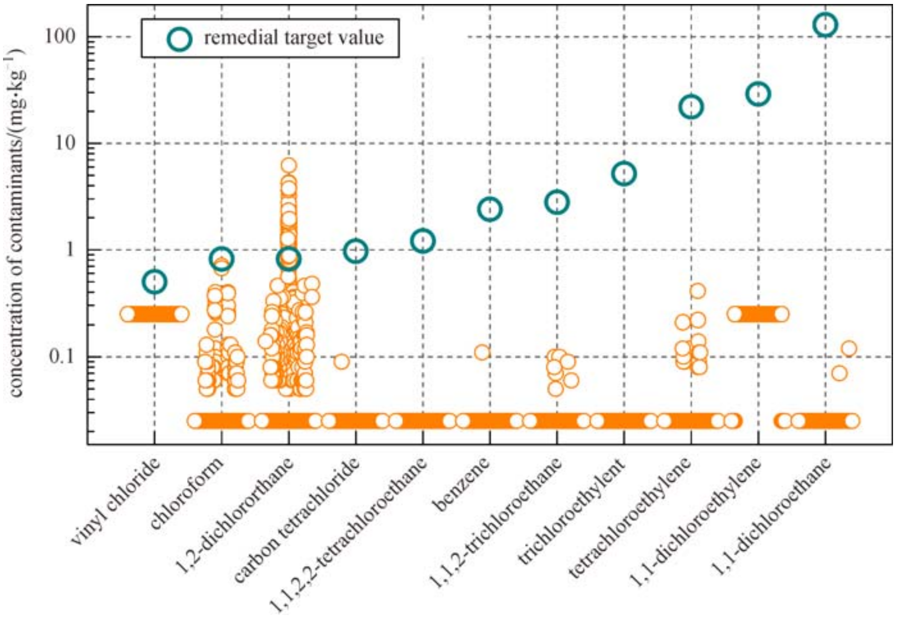

Fig. 4 Residual levels of contaminants in soil after remediation with mechanical soil aeration and comparison with their remedial target values

The cost of MSA is approximately 150 CNY/m^3 (not including the costs of soil excavation and transportation). The cost includes the construction of a closed greenhouse, the equipment and operating costs required to remove the released VOCs, and the stirring equipment and its operating costs. Several different remediation technologies for VOC-contaminated soils were compared with MSA (Table 3) [7,41–44]. As can be seen from Table 3, MSA methodology requires less time and is significantly more cost-effective than other commonly used methods for soil remediation. Nevertheless, it is clear that in order to solve the problem of sites contaminated with organic chemicals in China, it will be necessary to use MSA technology in combination with other remediation technologies, such as thermal desorption techniques.

6 Technical problems and challenges

6.1 Influence of tailing on remediation efficiency

The VOC concentration in the soil during mechanical aeration follows an inverse log function, a phenomenon referred to as tailing (Fig. 5) [14]. The tailing phenomenon becomes further pronounced with a decrease in temperature, increase in soil moisture, and elevation of soil density, such as in soils rich in clay. Tailing not only increases the time and labor required but also makes MSA less efficient, leaving higher levels of contaminant residues in the soil. Further studies are needed to improve the process of MSA under these conditions and to address the lower efficiency of aeration when contaminant concentrations are relatively low [41,45].

As mentioned above, temperature greatly influences the removal of volatiles from soil during aeration. To illustrate this phenomenon, we analyzed the data from a selected domestic large-scale contaminated site over a 1-year period [22]. The efficiency of removal of 1,2-DCA varied markedly depending on the season (Fig. 6). In the spring and summer (April to September), when average temperatures are relatively high, 1,2-DCA concentrations in the treated soils were below the remediation target level, whereas in the winter and autumn, when average temperatures are relatively low, the concentrations of 1,2-DCA remained above the required threshold. At lower temperatures, the residual concentration of 1,2-DCA exceeds contamination limits, indicating that temperature might markedly affect the efficiency of MSA. Thus, MSA might be more efficient at higher temperatures; at lower temperatures, it might be necessary to increase the processing time or take other measures to enhance the remediation of volatiles [14,46,47].

Table 2 Contaminant removal performance for soils treated with mechanical soil aeration

pollutants	initial concentration a) /(mg·kg^{-1})	concentration after treatment a)/(mg·kg^{-1})	remedial target value /(mg·kg^{-1})	average of removal efficiency/%	pass rate/%
1,1,2,2-tetrachloroethane	0.03	0.025 b)	1.21	16.67	100
1,1-dichloroethylene	0.34	0.22	29.1	35.29	100
trichloroethylene	0.06	0.025 b)	5.19	58.33	100
benzene	0.06	0.025 b)	2.4	58.33	100
vinyl chloride	0.62	0.25 b)	0.5	59.68	100
carbon tetrachloride	0.11	0.025 b)	0.97	77.27	100
tetrachlorethylene	0.17	0.026	22	84.71	100
chloroform	0.52	0.032	0.82	93.85	100
1,1,2-trichloroethane	0.41	0.025 b)	2.8	93.90	100
1,1-dichloroethane	0.72	0.025 b)	130	96.53	100
1,2-dichloroethane	30.59	0.17	0.82	99.44	93.8

Notes: a) The concentration is an average value; b) According to the environmental statistical requirements, concentrations below the detection limit are expressed as half of the detection limit

Table 3 Comparison of commonly used technologies for volatile organic compound-contaminated soils in China

technology	remediation period	remediation cost/(CNY·m^{-3})	engineering practice in China
soil washing	3–12 months	600–3000	few applications
thermal desorption	several weeks to years	350–2100	a few applications
chemical treatment	several weeks to months	500–1500	few applications
soil blocking landfill	several weeks to months	300–800	a few applications
mechanical soil aeration	several days to months	90–200	frequent applications

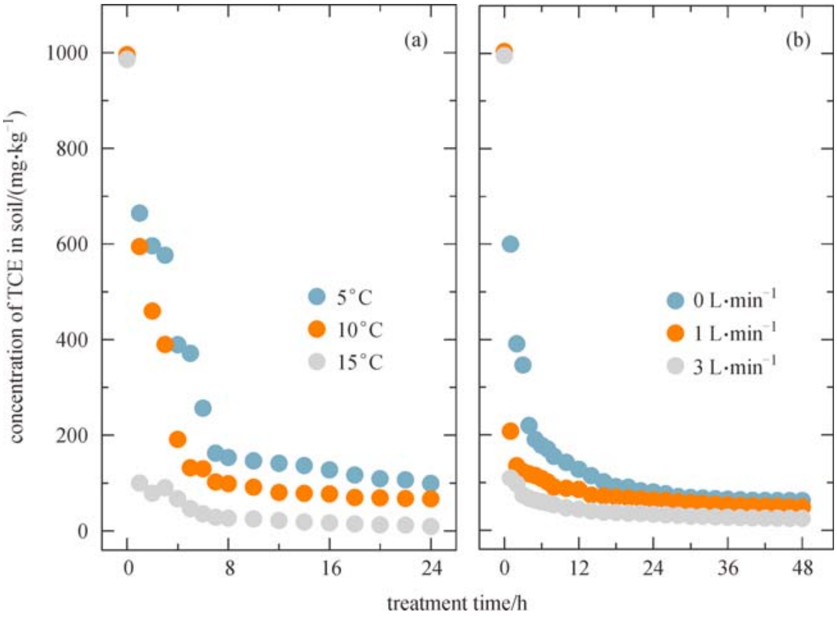

Fig. 5 Tailing of contaminant concentration in soil during mechanical soil aeration at different temperatures and aeration conditions. (a) Temperature-controlled experiment for clayey silt, with aeration rate, agitation interval, speed, and agitation time of 3 L·min^{-1}, 2 h, 200 r·min^{-1}, and 10 s, respectively; (b) Aeration-controlled experiment for clayey silt, with soil temperature, agitation interval, speed, and agitation time of 20°C, 2 h, 200 r·min^{-1}, and 10 s, respectively

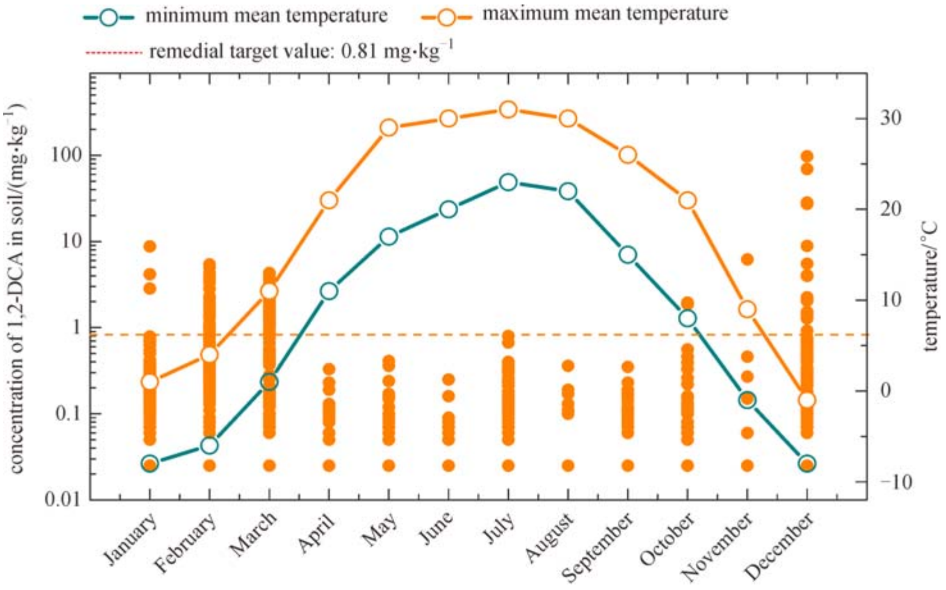

Fig. 6 Effect of temperature (seasonal change) on the removal of 1,2-dichloroethane (1,2-DCA) in soils under mechanical soil aeration treatment

6.2 Potential environmental risks

MSA has significant advantages, including low cost and shorter processing time. However, there are some drawbacks associated with this technology [48]. First, handling of contaminated soils is associated with an increased risk of human toxic exposure. Workers must wear appropriate personal protective equipment. Handling of contaminated soils during excavation, transportation, temporary storage, and stacking of soils into windrows all increase the risk of secondary contamination. Processing of soils containing high concentrations of VOCs inevitably increases the risks of atmospheric contamination [49–51]. Another risk is that the excavation process might pose structural risks to neighboring buildings. In addition, unforeseen events such as extreme weather and continuous rain might lead to high water content in the contaminated soils (causing longer processing time).

To mitigate these risks, the following measures need to be taken during soil excavation and transportation: 1) reducing the excavation surface, ensuring negative pressure within the greenhouse, spraying odor inhibitors, etc.; 2) ensuring that workers wear appropriate personal protection gear that provides sufficient protection under the measured concentrations of VOCs; 3) effective protection and containment of the excavation pit; 4) control of contaminants and dust during the excavation; 5) regular atmospheric monitoring of the area surrounding the excavation site, taking into consideration wind velocities and direction; and 6) development of strict environmental protection measures during the transportation of contaminated soils. Developing and implementing a coordinated and comprehensive plan, including environmentally sensitive procedures, monitoring of air and toxics, and emergency response, are necessary prior to the formal implementation of remediation projects in urban centers.

7 Future prospects

Unlike in developed countries, the development of soil remediation methods in China is still in the early stages. Formulation of regulation-based best practices and relevant experience might be essential for the safe and efficient reclamation of abandoned chemical sites, particularly in urban centers. Remediation programs for contaminated sites need to be optimized based on the characteristics of each site, taking into consideration the types of contamination and the unique hydro-geological characteristics of the site. Despite these challenges, MSA seems to be an effective and low-cost remediation option that has a great potential in developing countries such as China. MSA is a particularly promising method for land reclamation that addresses the challenges posed by limited land resources, large quantities of contaminated soil, demanding time constraints, and limited financial resources. However, although this technique has been successfully applied at some large-scale VOC-contaminated sites in China, it does have certain limitations. The prevention and management of secondary contamination need to be strengthened throughout the entire process of excavation, transportation, storage, and remediation. Equipment needs to be improved and perfected according to the specifics of the project, in order to achieve the goal of broad adoption and optimization of MSA. The engineering practice and development of MSA in China can, nevertheless, serve as a model for the employment of this technique in other developing countries.

Acknowledgements The present study was supported by National Environmental Protection Public Welfare projects (Nos. 201409047 and 201109017). We are grateful to Prof. Oliver J. Hao for his valuable comments and suggestions. We also acknowledge Dr. Shupeng Li from Beijing Construction Environmental Remediation Co., Ltd., Dr. Yong Yang from Zhongke Dingshi Environmental Engineering Co., Ltd., Sinopec Fifth Construction Co., Ltd., and Beijing Eco-Island Science and Technology Co., Ltd. for providing cases, material, and a basis for the preparation of this paper.

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Short biography- Fasheng Li

Professor Fasheng Li received his Bachelor degree in Department of Chemistry and Chemical Engineering of Hunan University in 1987 and PhD in Department of Chemistry of Kazan Lenin State University of Russia in 1993. Since 2001 he has been professor of Soil Pollution Control and Remediation in Chinese Research Academy of Environmental Sciences (CRAES) in Beijing, China. He established the Department of Soil Pollution Control at CRAES and was the director of the department in the period of 2005–2015. He is currently the Chief Engineer of CRAES.

Professor Li's research is aimed at developing cost-effective techniques and process for the remediation and redevelopment of contaminated sites as well as strategies and methodologies for risk assessment of industrially contaminated sites. He was responsible for the development of several national guidelines for risk control of contaminated soils in China and remediation of several megasites. He was the member of the international expert group on developing toolkit for risk assessment and remediation of POPs contaminated sites of UNIDO.

Professor Li has written 7 monographs on risk assessment and remediation of contaminated sites and more than 160 papers out of which more than 70 refereed papers published internationally.