

1 **Pilot application of SVE enhanced bioremediation technology for in situ**
2 **clean up of light oil contaminated site**

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16 **Abstract**

17 Light oil (isooctane) removal using soil vapor extraction (SVE) enhanced
18 bioremediation (BR) was investigated by four steps including (i) amendment of substrates
19 in batches; (ii) continuous induction of contaminants for 15 days; (iii) *in situ* acclimation for
20 100 days; and (iv) biodegradation assisted with SVE venting for 120 h at $20 \text{ m}^3 \text{ h}^{-1}$. Results
21 showed that the total removal efficiency was up to 90% after BR-SVE treatments. The
22 contribution of SVE to the overall removal was initially 53% ~ 69% and decreased to 13%
23 ~ 30% after 36 h. This implied that it would be an important strategy to limit water content
24 at the early stage while increase water supply at the end stage during implementation of BR-
25 SVE because water content was a significant factor hindering SVE but favouring BR.
26 Additionally, SVE was observed to increase the bioavailability and biodegradation by one-
27 order of magnitude. The overall results demonstrated a good complementarity between SVE
28 and BR and a potential for their combination in real-world applications.

29

30 **Keywords:** Soil vapor extraction, Bioremediation, Biodegradation, Bioavailability,
31 Isooctane

32 **1. Introduction**

33 Leaking underground storage tank (LUSTs) in the unsaturated zone is extensively
34 present in gas station, chemical plant and dry cleaning laundry, which produces wide-
35 reaching negative environmental impacts and threatens to human health [1, 2].
36 Bioremediation (BR) and soil vapor extraction (SVE) are effective remediation
37 technologies for treating and disposal of oil contaminated soils [3-5]. Microbial
38 decontamination (or bioremediation) of oil-polluted soils is a versatile alternative to
39 physicochemical treatments [6], which involves microbial decomposition of complex
40 organic or inorganic matter into simple non-toxic compounds such as CO₂ and H₂O
41 by living organisms (both indigenous or extraneous) in the presence of oxygen . It is
42 perceived as an important mechanism in the natural attenuation of oil pollutants and
43 hence a natural or 'green solution' to oil pollution problems because of minimal
44 ecological impacts [7]. However, the rate of microbial degradation of hydrocarbons in
45 soils under natural conditions is usually limited by several physicochemical and
46 biological factors including soil characteristics; abundance and diversity of
47 indigenous microorganisms; conditions for microbial degradation activity (e.g.,
48 nutrients, oxygen, pH and temperature); and the quantity, quality and bioavailability
49 of contaminants [7]. In order to augment bioremediation, in situ SVE is an alternative
50 approach, which consists of the installation of vertical and/or horizontal wells in the
51 unsaturated zone and the application of vacuum to increase the air flow through the
52 pore spaces of the soil. The added air flow (oxygen) subsequently stimulates the
53 growth and activity of the indigenous microbes and encourages the desorption of
54 volatile organic contaminants (VOCs) from the soil. In the process, the off-gas is
55 either treated to recover or destroy the VOCs because of its ignitability and toxicity
56 (acute and long-term carcinogenicity). For the treatment of SVE off-gas, active
57 carbon adsorption is currently the most common treatment technology in terms of
58 both cost and waste management [8]. However, the main limitations of carbon
59 adsorption are that (i) it is not effective for treating VOCs with high polarity or high
60 vapour pressures, and (ii) it would suffer from the high operating cost associated with
61 adsorbent replacement or regeneration if the contaminants concentration in off-gas is
62 high.

63 . BR and SVE were demonstrated to complement each other in terms of the factors
64 (e.g. type of soil and contaminants, moisture, natural organic matter content)

65 influencing the effectiveness of their performance [9-13]. While SVE is limited to
66 cases involving VOCs in unsaturated zone that is relatively permeable and
67 homogeneous, BR is applicable to a wide range of organics in all environmental
68 media that are prone to degradation by microorganisms. In addition, the high level of
69 moisture is favourable for microbial degradation, but it would reduce the soil
70 permeability, restrict the air flow through soil pores, and lessen the SVE efficiency [9].
71 The presence of natural organic matter may be a source of nutrients and microbial
72 communities having a great potential in bioremediation [14], but it could also serve as
73 a compartment for strong sorption of contaminants resulting in the decrease of SVE
74 effectiveness [12]. Moreover, SVE has a relatively short treatment time while the
75 period of BR is normally long. Therefore, combination of these two technologies is an
76 attractive approach with the potentials to promote the advantages and circumvent the
77 drawbacks compared to the application of each method individually.

78 The performance of this combined approach have been currently investigated by
79 Soares et al. [11] in which benzene was removed by SVE followed by BR in *ex situ*
80 column experiments. However, it remains unclear whether this approach would be
81 efficient for *in situ* remediation in which the site disturbance is minimal. Additionally,
82 it is of particular interest to investigate the effectiveness of implementing SVE after
83 BR with the potential to degrade the contaminants to a lower concentration and
84 thereby reduce the cost associated with active carbon replacement during the SVE off-
85 gas treatment. In this work the BR coupled with SVE was proposed for the *in situ*
86 remediation of light oil contaminated soils and the mass distribution of contaminants
87 into soil matrix was evaluated by a simple mathematical fitting. In order to investigate
88 the feasibility of field application, four stages were proposed as follows: (i) injection
89 of substrates to the soil in order to induce the real and potential metabolic activity of
90 indigenous microorganisms; (ii) addition of contaminants to formulate a simulated
91 contaminated zone; (iii) *in situ* acclimation for the adaption of microorganisms to the
92 artificially modified atmosphere; (iv) biodegradation assisted with SVE. Isooctane
93 was selected as a representative compound to illustrate the performance of this
94 method. Other contaminants such as cyclohexane, benzene, xylene, biphenyl,
95 perchloroethylene, trichloroethane, and gasoline may be effectively removed in the
96 same way.

97 **2. Materials and methods**

98 **2.1 Location of wells**

99 The experimental plot (10 m × 10 m) is located in the east of Tanggu District
100 (Tianjin, China) and soil samples were collected from the perched aquifer where
101 rainfall was the predominant water source. International standard methods were used
102 for the characterization of the soils including pH [15], moisture content [16], soil
103 organic matter [17], particle size [18], particle density [19]. The infiltration property
104 was assessed using drip infiltrometer [20].

105 The location of wells instrumented in the test field for implementing the BR-SVE
106 treatment is shown in Fig. 1. One vapor extraction well (EW1) was centrally located,
107 screened from 1 to 2 m below ground surface and connected to an air pump. The other
108 two wells (MW1 and MW2) were used as monitoring wells. Three 15 mm diameter
109 PVC wells (N1 to N3) were installed at 1 m intervals for injection of contaminants
110 and nutrients solution. At 11 locations (P1 to P4 and S1 to S7) in the test area, 4 gas
111 sampling wells were installed to sample soil vapor and to measure the pressure
112 drawdown throughout the test plot, and 7 solid sampling wells consisted of 15 mm
113 diameter stainless steel pipes with 20 slots (4 mm diameter) were installed to sample
114 soil and to measure the removal rate of contaminants. The intervals between ground
115 surface and wells were sealed off with bentonite pellets and covered with cement
116 grout.

117 **2.2 Experimental process**

118 The nutrients solutions consisted of $(\text{NH}_4)_2\text{SO}_4$ (50 g L⁻¹), K_2HPO_4 (5 g L⁻¹) and
119 MgSO_4 (0.06 g L⁻¹) were injected from injection wells after 6, 18, 24, 34, 48, 58 and
120 73 days in the experiments. Total 1.5 L (500 mL × 3 injection wells) nutrients
121 solutions were injected in batch on each injection day. The contaminants isooctane
122 (23 kg) was injected continuously from day 18 to 33. The contaminated zone was then
123 allowed for acclimation for 100 days when the amount of bacteria recovered to the
124 initial order of magnitude (10^7). The dispersion of isooctane underground was
125 calculated using software 'PetraSim' [21]. Briefly, the simulation zone (10 m × 10 m ×
126 3 m) was divided into 9464 (26 × 26 × 14) grids. The T2VOC programme was selected
127 as the numerical simulator which is a module designed to simulate 3-phase non-

128 isothermal flow of water, air and a volatile organic compound in multidimensional
129 heterogeneous porous media [22].

130 After the 100-day acclimation period, BR enhanced by SVE was performed by
131 venting which last for 120 h until the end of the experiments. The air (viscosity: $1.8 \times$
132 10^{-5} Pa · s) flow was monitored by a flow meter and controlled at 20 ± 1 m³ h⁻¹ as
133 reported in previous studies [23, 24]. The vacuum degree at the intake of air pump
134 and the WE1 well was 17 and 13 kPa, respectively. The pressure drawdown at various
135 monitor wells showed that the radius of influence (ROI) was between 1.2 and 4.0 m
136 [25]. The effective air permeability (k_a) within the range of ROI was estimated to be
137 at the order of magnitude of 10^{-12} m² using the model suggested by Johnson et al. [26].
138 The overall removal of isooctane during this period was determined by the
139 concentration in the soil phase. The isooctane removed by SVE was monitored by
140 measuring the concentration in the gas phase. The contribution of BR to the isooctane
141 loss was identified by the difference between the total isooctane loss in soil phase and
142 the amount removed by SVE.

143 **2.3 Instrument analysis**

144 The concentration of isooctane in gas phase was monitored in an AutoSystem XL
145 Gas Chromatograph (PerkinElmer GC, USA) equipped with a FFAP capillary column
146 (30 m × 0.25 mm × 1.0 μm) and flame ionization detector (FID). Vapor samples (1
147 mL) were taken at the gas sampling wells (P1~ P4) using syringe (PerkinElmer, USA)
148 and injected into the GC for determinative analysis. Vapor was pumped from each
149 sampling well to reach a steady-state vapor concentration before sampling. The
150 temperature of injector, column and detector were set at 230 °C, 100 °C and 300 °C,
151 respectively. Chromatographic data were collected and handled by the Software
152 Turbochrom 4.1.

153 The concentration of isooctane in soil was determined by HP 5890N GC equipped
154 with Agilent 7694E Headspace Sampler and FID. The soil samples (5 g) were
155 prepared from the sampling points (S1~ S4) to a depth between 1.2 and 1.4 m using
156 standard method [27]. The headspace sample (1 mL) was injected into the GC-FID
157 instrument using splitless injection. The HP-624 capillary column (25 m × 0.2 mm ×
158 1.12 μm) was used for the GC analysis. The injector and detector were set at 250 °C
159 and the column worked isothermally at 100 °C. The isooctane quantification was
160 performed by direct calibration method.

161 **3. Results and discussion**

162 The physicochemical characteristics of the soils are presented in Table 1. The soil
163 texture was recognized as loam, clay, silt clay and silt clay loam at sampling depth
164 from 0.3 to 2.3 m below the surface. Insignificant difference was found in the density,
165 pH and porosity between soils at different depths. The largest difference was observed
166 on the infiltration rate which decreased by 95% at 2.3 m depth compared to the top
167 subsurface. The pH values of the soils were slightly alkaline and within the preferable
168 ranges for bioremediation [28]. The sufficient soil water content (~ 22%) was
169 beneficial to biodegradation [11] but in contrast it may decrease the mass transfer
170 coefficient between the non-aqueous liquid phase and gas phase during the
171 implementation of SVE [9, 10]. Therefore, the relatively high vapor rate ($20 \text{ m}^3 \text{ h}^{-1}$)
172 used in this study was expected to favour SVE as previous study showed that the
173 impact of water content on SVE efficiency could be reduced by increasing the airflow
174 rate [11].

175 During the acclimation period, the first-order degradation reaction model provided
176 a good fit to the experimental data ($R^2 = 0.9937$, Fig. 2). At the end of 100-day
177 acclimation, the concentration of isooctane decreased by up to 63%. The estimated
178 areal distribution of the remaining isooctane from a single injection well indicated the
179 contamination was predominantly within the area of 0.5 m from the centre of injection
180 wells (Fig. 3a). Vertical profile of the relative concentration demonstrated that
181 isooctane diminished to undetectable levels within only 0.2 m below the ground water
182 table (1.8 m) during the sampling period (Fig. 3b).

183 The subsequent BR-SVE treatment resulted in a significant decrease in the
184 concentration of isooctane in both soil and gas phases (Fig. 4). The percentage loss of
185 isooctane resulted from BR was determined using the percentage loss of concentration
186 in soil (Fig. 4a) subtracted by the fractions removed by SVE that was estimated by the
187 area under the venting curve (Fig. 4b). Results demonstrated that SVE dominated the
188 isooctane removal in the first 36 h when its contribution to the overall removal ranged
189 from 53% to 69% (Fig. 5). On the contrary the remaining isooctane was mainly
190 removed by BR which contributed to 70 ~ 87% of the overall efficiency. This finding
191 was partially attributed to the increase of soil water content from 25 to 37% (data not
192 shown) due to the entering into rain season (August - September) in the test site.
193 Therefore, it is an important strategy to control water content at the early stage but

194 increase water supply at the end stage during the implementation of BR-SVE as water
195 content is a significant factor hindering SVE but enhancing BR.

196 In order to compare the influence of SVE on BR, the percentage of isooctane
197 removed by BR in absent of SVE (Fig. 5) was predicted using the degradation model
198 developed during the acclimation period (Fig. 2). Results indicated that the presence
199 of SVE significantly increased the biodegradation by one-order of magnitude (Fig. 5).
200 This may be attributed to the fact that the strong airflow accelerates biodegradation by
201 stimulating the transfer of the volatile fractions that was sequestered in the micro- or
202 nano- pores in the soils from solid phase into aqueous phase, increasing the degree to
203 which the compounds are free to move into or onto microorganisms, and
204 consequently increasing the dissolved mass available for uptake by the indigenous
205 bacterial populations. This finding coupled with the observation of insignificant
206 changes in the number of bacteria during BR-SVE process (Fig. 6) without nutrients
207 amendment suggested that complement of vapor extraction at the final stage of
208 bioremediation was beneficial for shortening the lag phase of biodegradation.

209 The overall results allowed concluding that the application of SVE would enhance
210 the removal of contaminants in two aspects such as (i) the vapor evaporates and drives
211 out the volatile components and (ii) the high speed air flow greatly increased the
212 bioavailability and biodegradation of the initially adsorbed components. The latter
213 appears predominant in the process. Future works are needed to (i) examine the
214 factors and mechanisms limiting the multiphase distribution of contaminants into soil
215 matrix, and (ii) develop mathematical models simulating the fate of contaminants
216 during the BR-SVE process.

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- 292
- 293

294

295 **Table 1** Physicochemical properties of soils at different depths below the surface

296

Depth (m)	Density (g mL ⁻³)	Moisture (%)	pH	SOM (%)	Porosity (%)	Infiltration rate (mm min ⁻¹)	Soil texture (%)		
							Sand	Silt	Clay
0.3 ± 0.1	1.48	22.3	7.8	0.6	45.1	0.63	47	27	26
1.2 ± 0.1	1.48	22.3	8.1	1.2	45.1	0.17	19	31	50
1.8 ± 0.1	1.47	26.4	8.2	1.1	45.4	0.14	0	58	42
2.3 ± 0.1	1.49	24.4	8.2	1.7	44.8	0.03	0	67	33

297

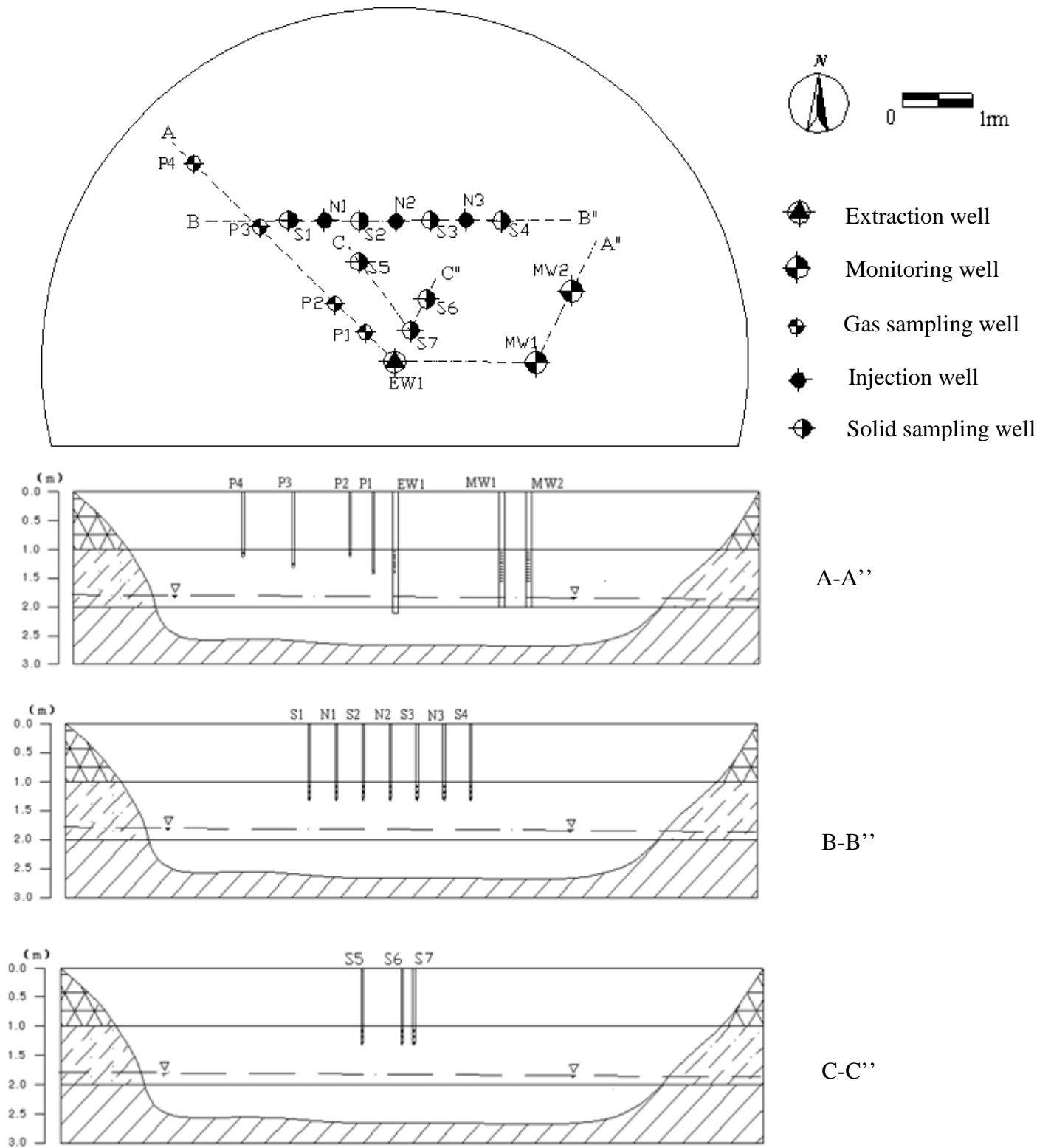


Fig. 1 Schematic of wells location

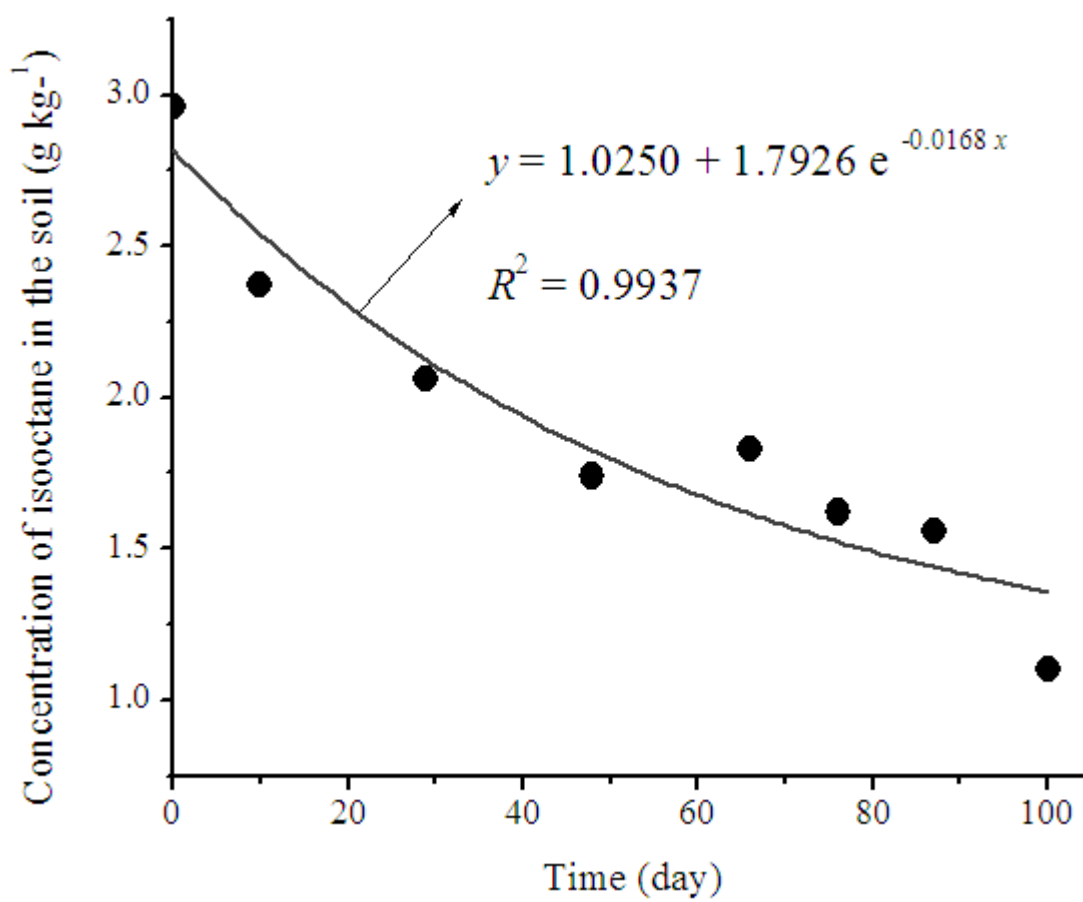
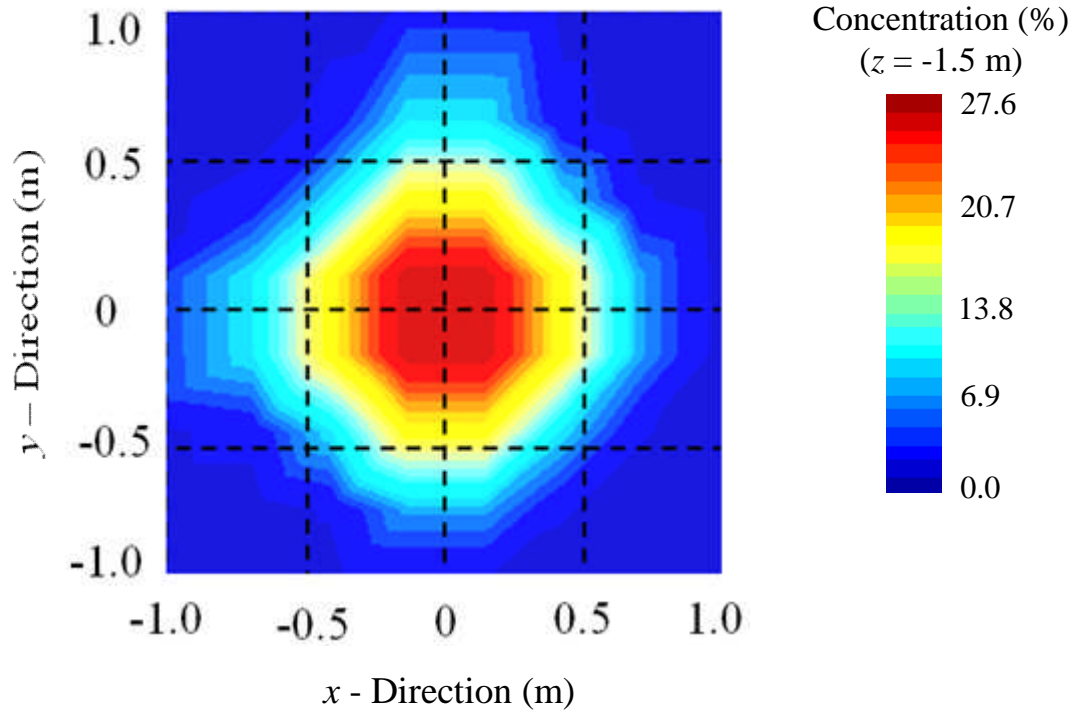


Fig. 2 Concentration of isooctane in the soil during the 100-day acclimation period

a



b

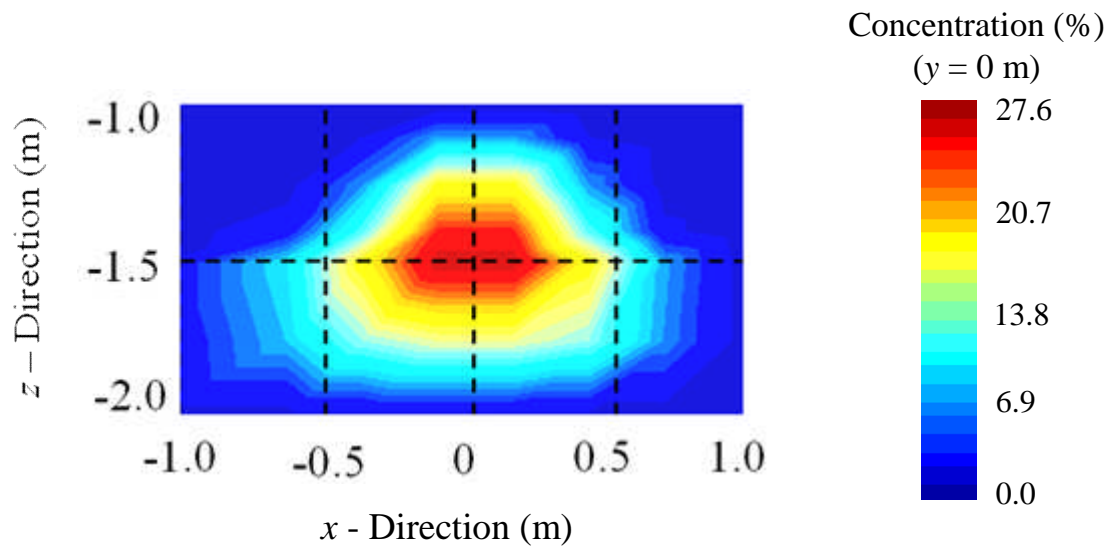


Fig. 3 The estimated (a) horizontal and (b) vertical dispersion of iso-octane near the injection well (single well) after 100-day *in situ* acclimation.

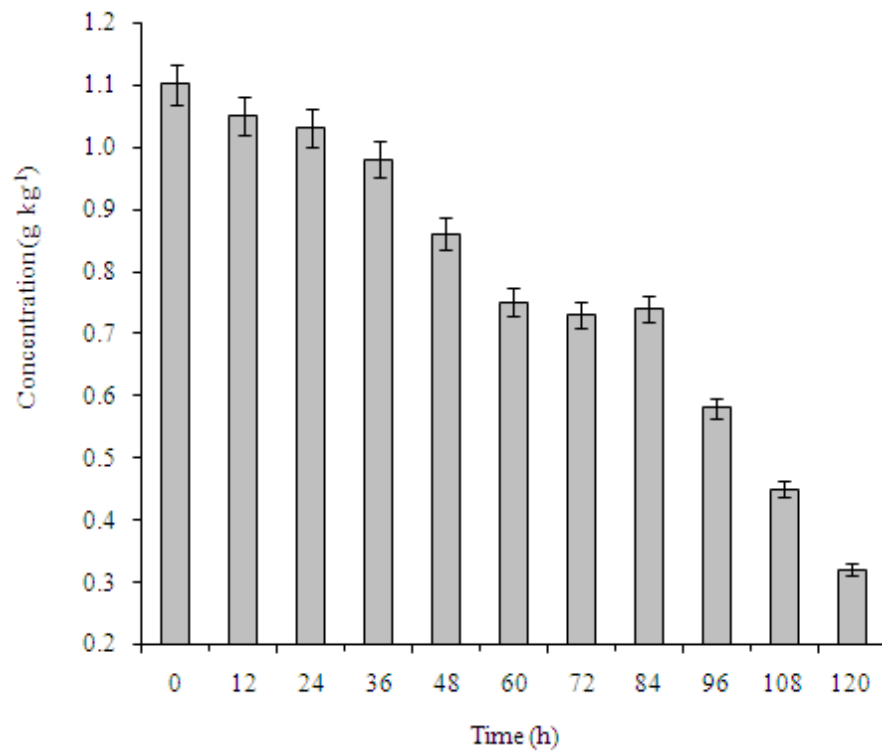
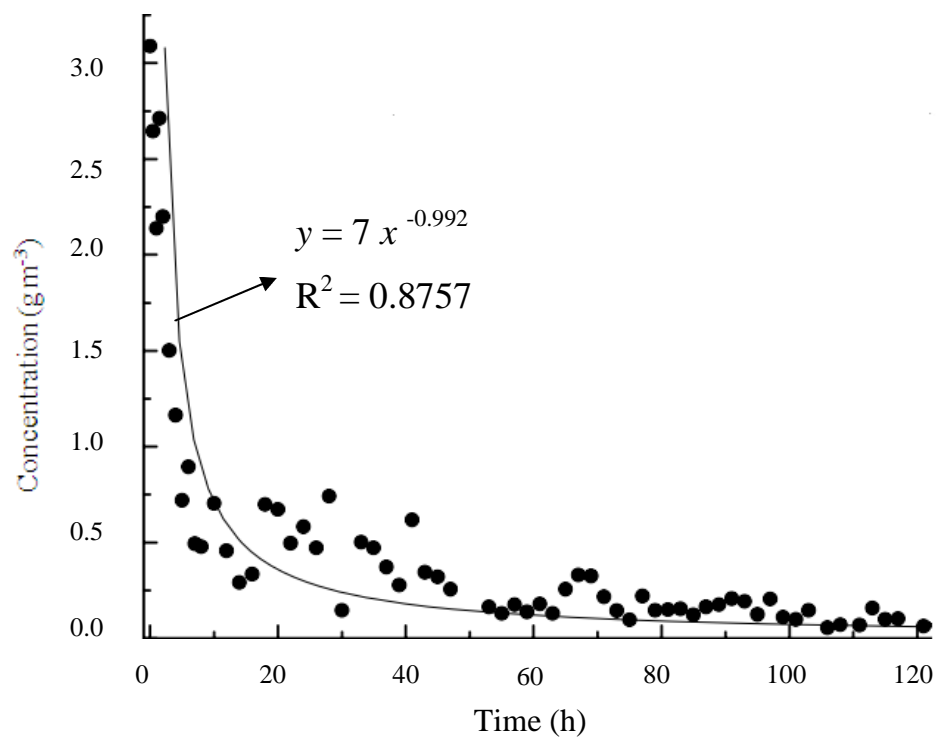
a**b**

Fig. 4 Concentration of isooctane in the (a) soil and (b) gas phase during the BR-SVE treatment

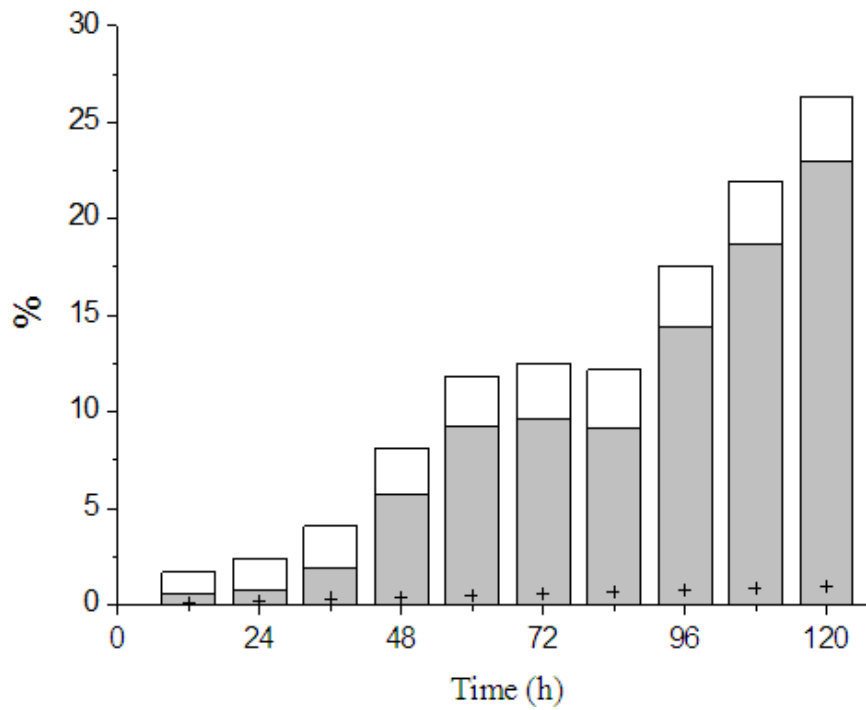
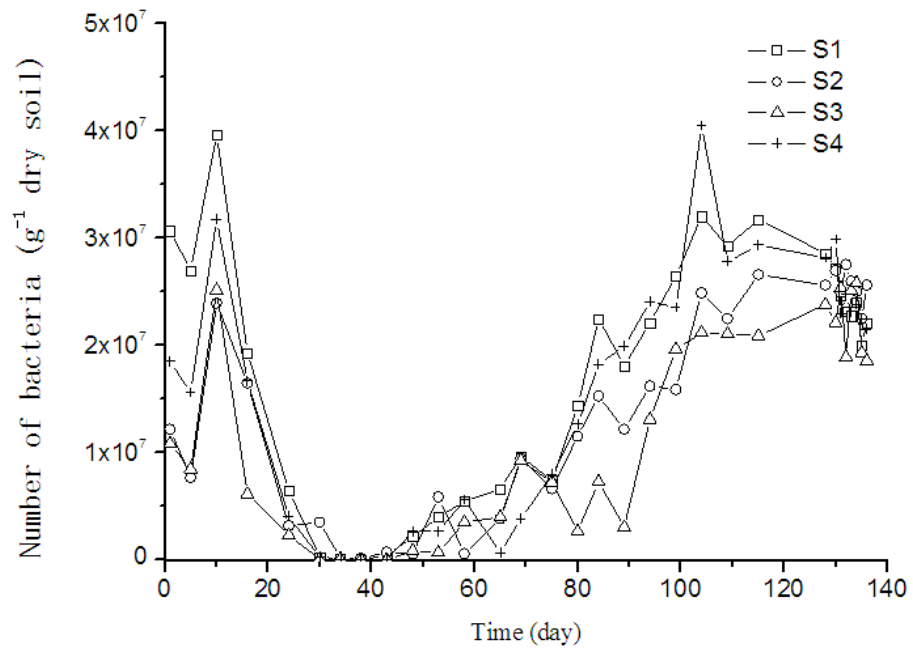


Fig. 5 Percentage of isooctane removed by BR (■) and SVE (□) during the BR-SVE treatment. The percentage removal by BR in absent of SVE (+) was estimated by the biodegradation curve during acclimation period.

a



b

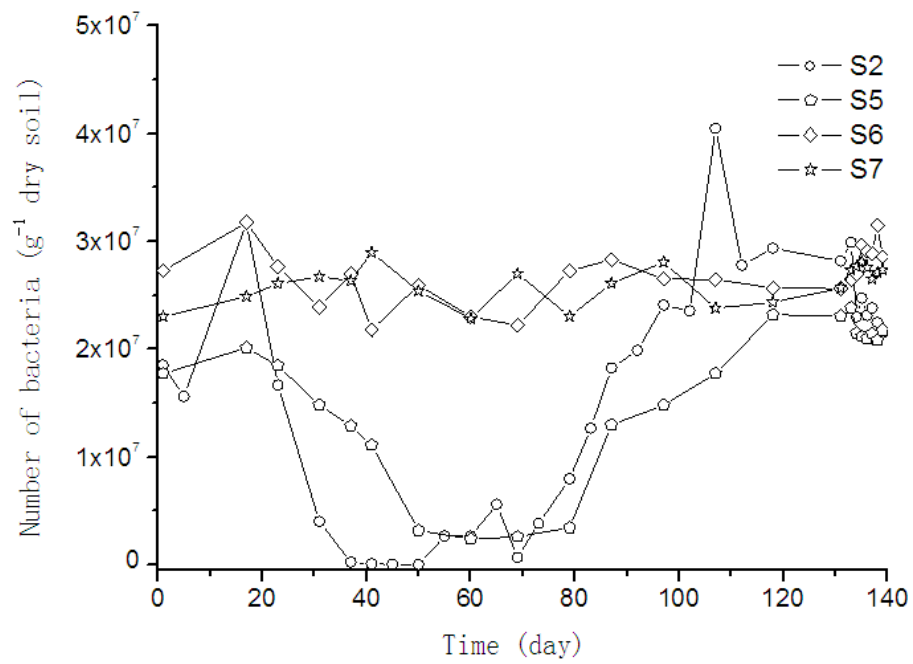


Fig. 6 Number of bacteria around the sampling wells

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