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A mobile, modular and rapidly-acting treatment system for optimizing and improving the removal of non-aqueous phase liquids (NAPLs) in groundwater

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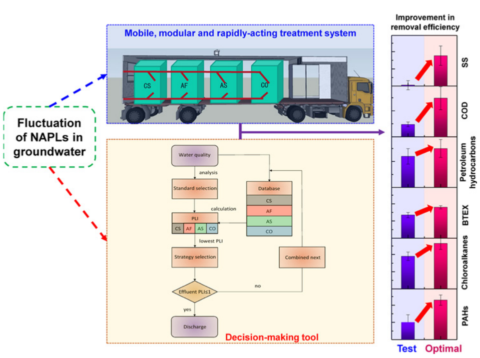
GRAPHICAL ABSTRACT



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ABSTRACT

Non-aqueous phase liquids (NAPLs) in pumped groundwater are highly variable, challenging the selection of above-ground treatment strategies in pump-and-treat system. Adjustable systems with multiple treatment units are urgently required. In the present study, a mobile, modular and rapidly-acting treatment system was developed to treat groundwater contaminated by NAPLs at a chemical industrial site. The system integrated four units of coagulation sedimentation, air flotation, air stripping and chemical oxidation. During a 3-month onsite operation, the composition of groundwater NAPLs had huge fluctuations and different treatment units had unique advantages in eliminating some components. For instance, air stripping exhibited satisfactory removal efficiencies ($> 80\%$) for short-chain petroleum hydrocarbons and chloroalkanes, but poor performance for others comparing to other units. A decision-making tool and a central control system were further developed to combine and adjust the four units in proper orders, achieving satisfied removal efficiency (70–90%) for multi-component NAPLs, regardless of composition fluctuation. These findings raise the state-of-the-art modular and rapidly-acting groundwater treatment system to clean up NAPLs contaminated groundwater through pump-and-treat strategy, help in better understanding on the decision and management to improve the treatment performance, and provide guidelines for its implication at other sites contaminated with multi-component NAPLs or undergoing accidental contamination.

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1. Introduction

The rapid industrialization and long-term anthropogenic activities have generated numerous contaminated sites globally, around 294,000 in US [1], 342,000 in Europe [2] and over 1,000,000 in China [3]. Caused by accidental spills or chronic leaks, subsurface accumulation of organic contaminants including both non-aqueous phase liquids (NAPLs) and hydrophilic substances poses serious threats to human health and ecosystem [4–8]. Pertaining the hydrogeological conditions, some contaminants are retarded in the vadose zone [9], whereas the others migrate deeper and eventually reach the groundwater table, leading to the subsequent contamination in groundwater. Additionally, frequently detected NAPLs at chemical industrial sites have multiple compositions, *e.g.*, petroleum hydrocarbons, benzenes, chlorinated compounds and aromatic hydrocarbons, accounting for around 55% of the groundwater contamination at sites in 16 European countries [2]. The majority of them are priority pollutants owing to their teratogenic, carcinogenic and mutagenic traits [1,10]. At the contaminated sites, NAPLs have long-term persistence (> 30 years) and facile transport, resulted from the low oxygen, limited nutrients (*e.g.*, C, N, P) and heterogeneous aquifer parameters with seasonal variation [11] and challenging the onsite cleanup activities.

From 206 documents for decision on groundwater treatment strategy reported by United State Environmental Protection Agency (USEPA), the most applied and well-accepted remediation strategies include Monitored Natural Attenuation (MNA), *in situ* treatment and Pump-and-Treat (P&T) [12]. As an *ex situ* strategy, P&T can remove contaminants in groundwater through manipulating hydraulic gradient, successfully cleaning hundreds of plumes of organic contaminants in groundwater since 1960s. Most previous studies and cases have addressed the decisions for the design, operation and optimization of the 'pump' portion in P&T, as well as the 'exit' strategy, *via* evaluating the groundwater extraction system and monitoring the long-term performance [13–15]. Only limited works attempt to maximize the removal efficiency, minimize the treatment cost and meet a set of technical and economic constraints in the above-ground treatment system [14]. The four widely applied physical-chemical approaches for the above-ground treatment include coagulation sedimentation, air flotation, air stripping and chemical oxidation, owing to their high removal efficiency and low allotted reaction time [11,16–20]. Coagulation sedimentation and air flotation are by far the most accepted processes for removing suspended solids and some hydrophobic macromolecular contaminants [20–22]. Over 90% of PAHs and mineral oils can be effectively removed by coagulation sedimentation and air flotation, respectively [21,22]. The selection of coagulation sedimentation or air flotation depends on the turbidity, state of floc aggregates and suspended solid density relative to water [20,23]. For highly volatile contaminants with high Henry's law constant (K_H), air stripping can evaporate them from the dissolved state in groundwater across the air/water boundary layer [6,17,24]. NAPLs suitable for air stripping include BTEX (benzene, toluene, ethylbenzene and xylene) and chloralkanes [11]. Chemical oxidation is another alternative to remove NAPLs, using chemical oxidants as electron accepters [6,16,24–26]. Some persistent organic pollutants (POPs) can be efficiently eliminated by chemical oxidation [27], and its efficiencies are highly dependent on pH, temperature, and the presence and effectiveness of oxidants (*e.g.*, electrode potential, E^o) [11,16].

Nevertheless, one of the greatest unsolved challenges in P&T is the fluctuation of contaminant compositions and aquifer conditions [11]. Hydrological conditions and chemical properties of NAPLs significantly affect the performance of P&T, such as the matrix diffusion and octanol-water partition coefficient (K_{ow}) [11,14,28]. Besides the seasonal fluctuation of groundwater table, the groundwater extraction process also alters the groundwater hydraulic conditions and the contaminant partition in subsurface, such as desorption, dissolution and infiltration of NAPLs within the vadose zones and aquifers [5,14]. Accordingly, the common decisions on the above-ground treatment attempt to select single or a set of combinatory treatment units, which have the potential to tolerate the high degree uncertainty in NAPLs composition and hydraulic fluctuation [29,30]. However, most of these systems can neither achieve the treatment objectives nor match the cost-effectiveness [4], even worse for rapidly-acting remediation in accidental contamination events in which fast decision-making for the selection of treatment units is required and the time for cleanup is limited [31–33]. Hence, it is of great urgency to develop decision-making tools to construct the above-ground treatment system with combinatory units, select appropriate treatment orders and achieve satisfactory removal efficiencies for a board range of NAPLs.

In the present study, a mobile, modular and rapidly-acting treatment system coupling with multiple physical-chemical treatment units was innovated. During the 3-month operation at a chemical industrial site in China, the performance of individual and combinatory treatment units (coagulation sedimentation, air flotation, air stripping and chemical oxidation) was evaluated in removing NAPLs in groundwater with huge fluctuation. A decision-making tool was developed for an intelligent central control system to combine and adjust the four treatment units in proper orders, significantly improving the removal efficiencies, regardless of the fluctuation in NAPLs composition. This work aims to improve the understanding of optimizing the treatment performance in P&T by appropriate decision-making strategies and provide guidelines for its application at other sites contaminated with multi-component NAPLs.

2. Method and materials

2.1. Site description

The study area is located at a chemical industrial site in a Chinese coastal city (29°53′10.38″N, 121°34′45.09″E). The site used to be a pesticide plant and has been abandoned for over 10 years. According to hydrogeological investigation, the lithology of subsurface soils is approximately divided into 5 layers, namely backfill soil (0–3.8 m), yellow clay (1.1–4.6 m), silty clay (1.8–24.8 m), sandy silt (16.2–23.1 m) and grey clay (18.9–28.9 m). The shallow groundwater level is 2.5 to 4.0 m, and the direction of groundwater flow is generally from west to east (Fig. 1A). Based on the distribution of NAPLs and the flow direction of groundwater, 21 pumping wells were drilled and their locations were illustrated in Fig. 1A.

2.2. Groundwater treatment configuration

The mobile, modular and rapidly-acting treatment system (Fig. 1B) was consisted of four treatment units, including coagulation sedimentation (CS), air flotation (AF), air stripping (AS) and chemical oxidation (CO), as illustrated in Fig. 1C.

CS unit (grey box in Fig. 1C) contained a pipeline mixer (101 in Fig. 1C) with both ends connected to a chemical dosing system (brown box in Fig. 1C) and an inclined tube sedimentation tank, respectively. The coagulants and flocculants were pumped from the chemical dosing system and mixed with the influents by the pipeline mixer to destabilize the particles and enhance the floc agglomerates. The treated groundwater was finally pumped into the inclined tube sedimentation tank to settle down.

AF unit (red box in Fig. 1C) was consisted of a coagulation/flocculation chamber, an air flotation chamber, and an effluent chamber. A microbubble generator (Karyu Turbo Mixer, Nikuni, Japan, 202 in Fig. 1C) recirculated and aerated groundwater from the effluent chamber to the air flotation chamber (reflux ratio, 1:10). The coagulants and flocculants were pumped from the chemical dosing system and injected into the coagulation/flocculation chamber with an automatic stirrer. A froth scraper (201 in Fig. 1C) was equipped in the air flotation chamber to remove the unwanted part during the air flotation process.