



# Bioaugmented soil aquifer treatment for P-nitrophenol removal in wastewater unique for cold regions

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## ABSTRACT

P-nitrophenol (PNP) is a toxic and recalcitrant organic pollutant and a usual intermediate in the production of fine chemicals, which has posed a significant threat to subsurface environment safety. Soil aquifer treatment (SAT) is a promising method to remove and remediate contamination in vadose zone with low cost and high efficiency. However, there are still research gaps for the treatment of recalcitrant contaminants by SAT in cold regions, such as un-robust indigenous microbes and low temperature constraint in vadose zone. The bioaugmentation technology was first introduced into SAT in order to enhance the removal ability of PNP by SAT operated in cold regions in this study. A high-efficiency PNP-degrading bacterium was successfully isolated, which can efficiently degrade PNP below 200 mg L<sup>-1</sup> with a degradation rate above 99% at 15 °C close to the real subsurface temperature in cold regions, and added into SAT for bioaugmentation. The feasibility of bioaugmented SAT and associated PNP removal process were investigated by laboratory sand columns, along with effects of the SAT operative parameters (namely PNP loading concentration, flow rate and soil saturation level of SAT). Within the range of PNP loading stresses tested (1–200 mg L<sup>-1</sup>), PNP removal efficiency was optimal at constant flow rate of 219 mL d<sup>-1</sup> in unsaturated operating condition of SAT under 15 °C among all the investigated experimental conditions. Longer hydraulic residence time increased the PNP removal rate, although the accumulated mass removed reduced and the removal efficiencies remained constant in unsaturated operating condition of SAT. It is found from the comparison between the PNP removals via both unsaturated and saturated columns that slight difference only in the removal rate of PNP was observed and the highly efficient bioaugmented SAT can completely degrade PNP of 10 mg L<sup>-1</sup> within 5 wetting/drying cycles under both scenarios.

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## 1. Introduction

P-nitrophenol (PNP), i.e. 4-nitrophenol is a significant xenobiotic nitro-aromatic compound (Chi et al., 2013; Kovacic and Somanathan, 2014). Due to the toxicity and mutagenic potential and most deleterious effects to ecosystem, PNP has been blacklisted for priority contaminants by USEPA since 1980 (Yue et al., 2018; Zhao and Kong, 2018; Zheng et al., 2009). PNP can be found as a common contaminant in natural environment because of its extensive and frequent occurrence in the synthesis and application of pharmaceuticals, dyes, pesticides, industrial solvents and explosives (Sahoo et al., 2011; Wang et al., 2017; Yue et al., 2018; Zhao

and Kong, 2018). Owing to the recalcitrance of PNP in subsurface environment, PNP wastewater could easily penetrate vadose zone soil with rainfall or irrigation (Sahoo et al., 2011), and then transport quite a long distance in groundwater system (Zheng et al., 2009). Therefore, to control PNP's dispersion for further contamination, it is better to eliminate PNP at or close to the sources in the vadose zone before it enters into groundwater.

Soil aquifer treatment (SAT), an in-situ remediation technology working mainly in the natural vadose zone, is proved to be a cost-effective and environmental friendly wastewater treatment and reclamation system whilst undertaking groundwater artificial recharge (Essandoh et al., 2011; Zucker et al., 2015). The specific contamination removal mechanisms in SAT are shown in Fig. 1. Wastewater is supplied to an infiltration tank excavated in soil of good permeability; a series of natural physical, chemical and biological reactions could then take place leading to great reduction of

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### List of abbreviations

Abi-SAT	abiotic soil aquifer treatment
Bio-SAT	bioaugmented soil aquifer treatment
DO	dissolved oxygen
HEPD-bacterium	high-efficiency PNP-degrading bacterium
HPLC	high performance liquid chromatography
PNP	P-nitrophenol
PPCP	pharmaceuticals and personal care product
SAT	soil aquifer treatment
TOC	Total organic carbon
UV	ultraviolet

inorganic and dissolved organic contaminants during the process of wastewater permeating through the vadose zone (Ak and Gunduz, 2013; Mienis and Arye, 2018; Sopilniak et al., 2018). Removal efficiency could be further improved by enhanced methods in the vadose zone of SAT system, such as improving adsorption capacity of SAT via adding special materials, e.g. steel slag (Cha et al., 2006); enhancing infiltration regime of SAT via rapid flooding (Nadav et al., 2012) and so on. To ensure water safety, groundwater in aquifer beneath the vadose zone would generally be withdrawn for further use such as unrestricted irrigation or industrial purposes months or years later (Onesios and Bouwer, 2012).

SAT has been employed in many subtropical regions around the world with the purpose of treated wastewater's reclamation, such as California US, South France and Tel Aviv, Israel (Arye et al., 2011; Brissaud et al., 1991; Mienis and Arye, 2018; Nadav et al., 2012; Quanrud et al., 2003). In recent years, many laboratory studies were

carried out on the removal of some recalcitrant organic contaminants in wastewater using SAT at room temperature, apart from easily degradable contaminants (Guizani et al., 2011; Hübner et al., 2014). For example, Onesios and Bouwer (2012) attempted to remove 14 PPCPs (Pharmaceuticals and Personal Care Products) by SAT system and the results showed near 95% reductions in 10 of 14 PPCPs were obtained in laboratory soil column tests, and the bio-film was detected to take effect on the SAT capacity enhancement until nearly 256 days of in-situ microorganism growth (Onesios and Bouwer, 2012). The SAT soil columns have also been used by Essandoh et al. (2012) for estrogens removal both in unsaturated and saturated operating conditions. The removal efficiencies were improved by increased transport length in unsaturated zone. The improved removal efficiencies were also obtained at lower flow rates in both unsaturated and saturated conditions (Essandoh et al., 2012).

Among all the removal mechanisms of SAT shown in Fig. 1, adsorption and biodegradation are considered as the major ones for dissolved organic compounds (Essandoh et al., 2012, 2013; Mienis and Arye, 2018). Moreover, biodegradation is normally the dominant removal mechanism for recalcitrant contaminants (Ak and Gunduz, 2013). Most biodegradation in the SAT systems were achieved by acclimatizing the microorganisms existing in soil or wastewater (Essandoh et al., 2012, 2013; Mienis and Arye, 2018; Onesios and Bouwer, 2012). However, such process would take too long time and may be much difficult to work on toxic organic contaminants of high concentration. As PNP is toxic to most microbes, rare indigenous microorganisms in soil and wastewater are capable of degrading PNP (Rodrigues et al., 2017). Therefore, it is difficult to cleanup PNP in wastewater by conventional SAT-biological treatment.

Attempts in enhancing the PNP elimination artificially have

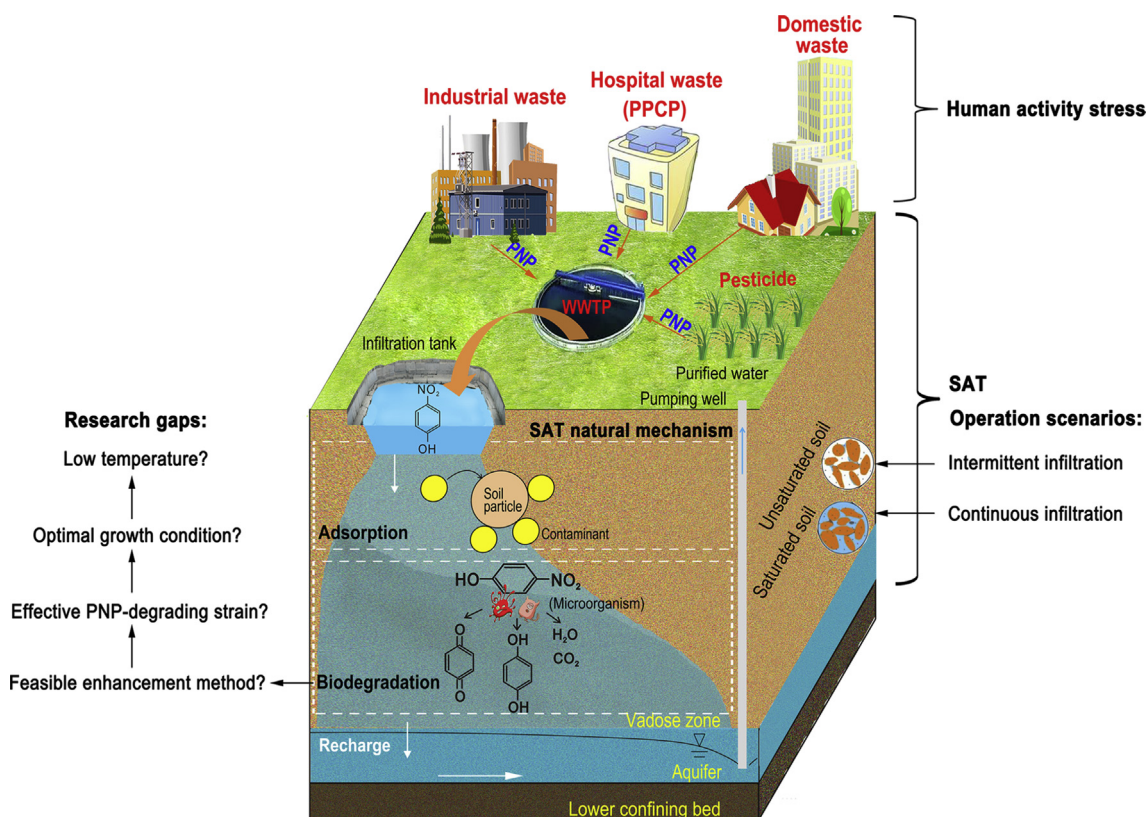


Fig. 1. The conceptual model of the SAT remediation process for PNP.

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