EPA Public Access

Author manuscript

Chemosphere. Author manuscript; available in PMC 2019 December 01.

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Published in final edited form as:

Chemosphere. 2018 December ; 213: 551–558. doi:10.1016/j.chemosphere.2018.09.092.

Enhanced degradation of polycyclic aromatic hydrocarbons by indigenous microbes combined with chemical oxidation

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Abstract

In this study, the removal efficiency PAHs by chemical oxidation combined with microbe remediation was evaluated in two contaminated soils. The number of indigenous soil microbes decreased after the addition of chemical oxidants and then increased by nutrients addition. The total removal efficiencies of PAHs by chemical oxidation and nutrient addition followed the order: activated persulfate > potassium permanganate > modified Fenton reagent > Fenton reagent. There are 24.29–27.97%, 22.00–23.67%, 10.24–13.74% and 1.9–2.5% contributions separately due to nutrient treatment in Fenton, modified Fenton, activated persulfate and potassium permanganate treatment, which show significantly difference. The different chemical oxidants exhibited 78–90% removal efficiency for 5–6 rings PAHs, while 52–85% removal efficiency for 2–4 rings PAHs. With the addition of nutrients, the growth of indigenous microbes was enhanced significantly, and the contents of 2–4 rings PAHs in the soil were further decreased. Furthermore, the removal efficiencies of NAP and ANY were increased by more than 45%, while the removal efficiencies of ANE, FLE and PHE were about 30% at Fenton system. There was a complementary enhancing effect of microbial remediation for PAHs degradation after chemical oxidation.

Graphical Abstract

Keywords

soil; polycyclic aromatic hydrocarbons; microbe; chemical oxidation; combined strategies

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1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are widespread pollutants, commonly being found at sites associated with coking, petroleum production industries and incomplete combustion of wood (Kuppusamy et al., 2017). Studies have shown that 90% of environmental PAHs is present in soil (Wild and Jones 1995). Despite some innovative methods used to remediate contaminated soils, an increasing focus is placed on cost-effective in situ remediation technologies with less environmental impact, such as in-situ chemical oxidation (ISCO) and in-situ microbiological remediation. These technologies are considered as efficient and are adaptable to a variety of soil conditions and contaminant types (Singh 2009; Sutton et al. 2011).

The typical chemical oxidants used in the ISCO include Fenton, modified Fenton, potassium permanganate, persulfate and ozone (Siegrist et al. 2011). In addition to degrading contaminants, these technologies remove compounds that are adsorbed to soil organic matter (SOM) or in the form of light or pure product, as especially high molecular weight (HMW) PAH is present as solid. Chemical oxidation has demonstrated to remediate soil polluted with various organic contaminants, including PAHs (Sutton et al. 2014). Nevertheless, ISCO can adversely affect soils, including quantitative and structural changes to SOM, increases or decreases in pH, and reduction in soil microbial communities (Siegrist et al. 2011; Baciocchi et al. 2014).

Microbiological remediation depends on the natural metabolism of microbes to degrade organic contaminants. Researches on the current microbial degradation of PAHs have been well reviewed (Ghosal et al. 2016). Organisms belonging to genera *Pseudomonas*, *Alcanivorax*, *Microbulbifer*, *Sphingomonas*, *Micrococcus*, *Cellulomonas*, *Dietzia*, *Gordonia*, *Marinobacter*, *Mycobacterium*, *Haemophilus*, *Rhodococcus*, *Paenibacillus*, *Bacillus*, *Aeromonas*, *Burkholderia*, *Xanthomonas*, *Micrococcus*, *Arthrobacter*, *Acinetobacter*, *Corynebacterium* and *Enterobacter* have been documented to effectively degrade PAH compounds (Wu et al. 2013). Although chemical oxidation has a certain “destructive effect” to soil, in recent years, studies have shown that chemical oxidation combined with biodegradation is more effective and applicative to remove the pollutants in soil. It can be inferred that chemical oxidation will decrease the high concentrations of organic pollutants; and then, microbes can be used to deal with low concentrations of residual organic pollutants, thereby improving the remediation efficiency (Sutton et al. 2011; Chen et al. 2016; Xu et al. 2016).

In this study, the effectiveness of Fenton reagent, modified Fenton reagent, potassium permanganate, and activated persulfate coupled with bioremediation for PAHs degradation were evaluated by adding a nutrient substance after chemical oxidation. This study aimed at clarifying the role of the population of indigenous microorganisms in the overall remediation of residual PAHs-contaminated soil.

2 Materials and Methods

2.1 Experimental design

Soil samples were collected from a coal gas plant field (abbreviated G) in Nanjing City and coking plant field (abbreviated C) in Shijiazhuang City were selected in this study. The physical and chemical characteristics of coal gas plant field soil (G) were shown in table 1.

The optimal oxidant dosage (Liao et al., 2011) of Fenton reagent (Fenton), modified Fenton reagent (M-Fenton), potassium permanganate (KMnO₄) and activated persulfate (Active-PS) was used to oxidize the contaminated soils. The composition of the 4 oxidizing solutions was: Fenton reagent (H₂O₂ 30%, 1 mmol·g⁻¹ and FeSO₄ 0.5 mol·L⁻¹, 0.2 mmol·g⁻¹); modified Fenton (H₂O₂ 30%, 2 mmol·g⁻¹ and FeSO₄ 0.5 mol·L⁻¹, 0.4 mmol·g⁻¹ and citric acid 0.4 mmol·g⁻¹); Potassium permanganate (KMnO₄ 0.4 mol·L⁻¹, 1.5 mmol·g⁻¹); Activated persulfate (Sodium persulfate 1 mol·L⁻¹ 2 mmol·g⁻¹ and FeSO₄ 0.5 mol·L⁻¹ 0.2 mmol·g⁻¹ and citric acid 0.04 mmol·g⁻¹).

Sixty tests were conducted to investigate the changes in indigenous soil microorganism population by chemical oxidant stress. For each test, 50g of contaminated soil was weighted into a 250-mL Erlenmeyer flask. Milli-Q water was first added into the flask to make the final volume of the Milli-Q water and oxidant to 100 mL, and the flask was sealed to form slurry by a magnetic stirrer. A glass tube was connected to another flask containing methylene chloride to collect the released gas. Then the chemical oxidant was added slowly by the set amount. To make a homogeneous suspension, the Erlenmeyer flasks were placed on the magnetic stirrer for about 2 h at 150 rpm. After standing for 22 h, the flasks were covered with a sterile breathable film, and microbes were cultured in a sterile dark room at room temperature. On the sixteenth day, 10 mL nutrients (0.126 mol·L⁻¹ NH₄Cl and 0.0126 mol·L⁻¹ NaH₂PO₄/Na₂HPO₄) were added into the soil suspensions. Control group was treated with 10 mL sterile water. Of each suspended soil sample, 10 mL was collected using a pipette on 0d, 0.5d, 1d, 3d, 5d, 7d, 10d, 15d, 18d, 21d, 24d for further analysis. All experiments in this study were performed in triplicate.

2.2 Chemicals

Milli-Q water was used in the experiments. Sodium persulfate (Na₂S₂O₈, >99%; Beijing chemical reagent, China), hydrogen peroxide (H₂O₂, 30%; Beijing chemical reagent, China), and potassium permanganate (KMnO₄, >99%; Beijing chemical reagent, China) were used as the oxidants. Citric acid (C₆H₈O₇, >99.8%; Tianjin chemical reagent, China) and ferrous sulfate (FeSO₄·7H₂O, >99.8%; Tianjin Chemical Reagent, China) were used as the activated reagent. Acetone (HPLC), dichloromethane (HPLC) and n-hexane (HPLC), purchased from Duksan Pure Chemicals Co., Ltd (Korea), were used for GC-MS analysis.

2.3 PAHs analysis

From the samples, 2.0 g of freeze-dried soil was extracted by ultra-sonication with acetone and dichloromethane mixture. The extracts were concentrated and then transferred to silica gel column for cleanup by washing with hexane and dichloromethane mixture. The eluate was concentrated and analyzed by Gas Chromatography-Mass Spectrometry (GC-MS)

(Agilent 7890A GC coupled with a 5975C MS, USA). The average recoveries of PAHs were 95–110% ($n=6$, relative standard deviation $< 2.61\%$). The detection limit for PAHs was $1 \text{ ng}\cdot\text{g}^{-1}$ (Sun et al., 2014).

2.4 Microbial counts

Bacterial counts were determined according to ISO 6222 (1999). An amount of 10 g of the sample was mixed with 20 mL of physiological solution (0.9% NaCl) in the sterile tube. After diluted log 1, 2, 3, 4, 5, 6, 7, 8, 9 times with 0.9% saline solution, 1 mL sample was spread on plate count agar without dextrose (BIO-RAD) and incubated at 22°C for 4 d. Then the sample was counted for the colony forming unit (CFU).

2.5 Statistical analysis

The mean and standard deviations of triplicate independent experiments were calculated. The mean values were compared by a parametric one-way ANOVA test. Parts of the statistical analyses and graphing were performed using the Origin 2016 software program (OriginLab Corporation, USA).

3 Results and Discussion

3.1 Counts of indigenous microbes

The variations of counts of indigenous microbes in soils were described in Fig. 1 and Fig. 2. The number of indigenous soil microbes decreased due to the chemical oxidant and then increased by nutrients addition. With the consumption of chemical oxidants, the microbes began to recover on the 7th day, and the growth of microbes was promoted significantly on the 16th day by adding nutrients. In both the coking plant and coal gas plant soils, the number of indigenous microbes was reduced sharply from 10^6 – $10^7 \text{ CFU}\cdot\text{g}^{-1}$ to 10^2 – $10^3 \text{ CFU}\cdot\text{g}^{-1}$ after the addition of chemical oxidants in the soil. After the 7th day, the number of microbes in the two soils began to recover, increasing from 10^2 – $10^3 \text{ CFU}\cdot\text{g}^{-1}$ to 10^5 – $10^6 \text{ CFU}\cdot\text{g}^{-1}$ (except the treatment of potassium permanganate). On the 16th day, the number of microbes increased significantly with the addition of nutrients. The addition of nutrients increased the number of microbes in the soil even more than the microbial level in the original soil that was treated with sodium persulfate reagent, Fenton reagent and modified Fenton reagent. The number of microbes in the soil pre-oxidized by the Fenton reagent was increased from $(2.6\pm 0.2)10^2 \text{ CFU}\cdot\text{g}^{-1}$ to $(6.8\pm 2.4)10^7 \text{ CFU}\cdot\text{g}^{-1}$, the number of microbes in the soil pre-oxidized by the modified Fenton reagent was increased from $(5.3\pm 1.3)10^2 \text{ CFU}\cdot\text{g}^{-1}$ to $(6.1\pm 1.2)10^7 \text{ CFU}\cdot\text{g}^{-1}$, and the number of microbes in the soil pre-oxidized by the activated sodium persulfate reagent treatment was increased from $(1.3\pm 0.3)10^3 \text{ CFU}\cdot\text{g}^{-1}$ to $(2.1\pm 0.7)10^7 \text{ CFU}\cdot\text{g}^{-1}$. The number of microbes in coking plant soil also showed a similar trend with these treatments. The addition of nutrients significantly increased the number of microbes in the soil treated with potassium permanganate, but it did not restore it to the original microbial level on the 24th day.

Nutrient is one of the limiting factors for microbial degradation of PAHs. In addition to carbon sources, microbes also require a variety of minerals, such as nitrogen and phosphorus for normal metabolism and growth. Thus, exogenous nutrients can be supplemented to

stimulate the growth of indigenous micro-organisms, and enhance the bioremediation of contaminants, which is also known as biostimulation (Atagana et al., 2003; Trindade et al., 2005). This remediation technology is not suitable for soils with high concentrations of contaminants and poor permeability, due to the low tolerance of microbes and liquid form of oxidants (Kuppusamy et al., 2016). In this study, the concentration of PAHs in soil was reduced by chemical oxidation, and the growth of indigenous microbes was stimulated by adding nutrients. The residual PAHs after chemical oxidation were further reduced by microbial degradation. Chen et al. (2016) showed that the addition of nutrients to 1 to 5% (w/w) hydrogen peroxide, potassium permanganate, and persulfate resulted in an increase in the order of microbes from 10^4 CFU·g⁻¹ to 10^7 – 10^8 CFU·g⁻¹. In this study, the dose of potassium permanganate was different, resulting in a slower increase in microbial growth compared with other oxidants. However, previous studies have shown that not all cases require exogenous nutrients to promote bioremediation. Sirguey et al. (2008) showed that the phosphate concentration in the soil after oxidation of Fenton or permanganate was increased due to the oxidation and release of soil organic matter. Kulik et al. (2006) pointed out that, compared with sand, pre-oxidation significantly improved the effect of microbial remediation due to higher nutrient content in peat. Therefore, the effects of soil type, nutrient content, and chemical oxidation treatment on the subsequent microbial remediation can be complicated and need to be studied case by case for practical site remediation.

3.2 Degradation of total PAHs after adding nutrients

Fig. 3 and Fig. 4 showed the total PAHs content and removal efficiency after remediation by chemical oxidation combined with nutrient addition in the coal gas plant soils and coking plant soils. The results indicated that the four chemical oxidants reduced the content of PAHs significantly in the soils, however the PAH concentrations were rather stable after 3 d, when probably all oxidants were depleted. After addition of nutrients on day 16 the decrease in PAH concentrations was resumed for soil G (Fig. 3). This occurred together with the increase of the number of indigenous microbes (Fig. 1 and Fig. 2), whereas in the controls the PAH decrease was not resumed. For soil C only after the modified Fenton treatment a further lowering of PAH occurred (Fig. 3).

It can be seen from Fig. 3 and Fig. 4 that the removal efficiency of PAHs by chemical oxidation was in the order: potassium permanganate (91.30–92.40%) > activated persulfate (80.90–85.60%) > Modified Fenton reagent (67.20–69.60%) > Fenton reagent (54.10–59.80%), which show significantly difference. Chemical oxidation by potassium permanganate and activated persulfate showed to be most effective for PAH removal in soil G and C. The total removal efficiencies of PAHs with the addition of oxidant and nutrient followed the order: activated persulfate (94.64–95.84%) > potassium permanganate (93.80–94.30%) > modified Fenton reagent (90.87–91.90%) > Fenton reagent (78.39–87.77%). Nutrient addition after Modified Fenton treatment showed a similar results as for potassium permanganate and activated persulfate. And the Fenton treatment was less effective and after nutrient addition still higher PAHs concentrations were left.

Among them, the removal efficiency increased by 24.29–27.97%, 22.00–23.67%, 10.24–13.74% and 1.9–2.5% respectively after adding nutrients in the Fenton, modified Fenton,

activated persulfate and potassium permanganate treatment. It was found that the highest total removal rate of PAHs was found in the activated persulfate pre-treatment. In contrast, the addition of nutrients did not increase the removal rate of PAHs in the soil after pre-treatment with potassium permanganate (1.9–2.5%); however, because of its high efficiency of chemical oxidation to remove PAHs (91.30–92.40%), PAHs removal rate for potassium permanganate was still higher than those of Fenton reagent and modified Fenton reagent.

Nutrients were used to stimulate microbial growth as a standard method of microbial remediation, especially in the soil low in nutrients (Sutton et al., 2014). Previous experiments under laboratory conditions have shown enhanced microbial growth by simultaneously controlling pH and providing phosphate (Liang et al., 2009; Valderrama et al., 2009). The addition of trace metals, minerals or vitamins can also help create favorable conditions for the growth of microbes (Sahl et al., 2007; Xie and Barcelona 2003). Sutton et al. (2014) reported that the removal of TPH was almost zero after the addition of nutrients for 80 d pre-treatment with potassium permanganate. In that study, after the pre-treatment with Fenton, soil was incubated for 80 d with the nutrients, and TPH removal efficiency was about 25%; in the modified Fenton reagent pre-treatment after adding nutrients for 80 d, the microbial residual TPH removal efficiency was about 57%; in the pre-treatment of persulfate after adding nutrients for 80 d, the removal efficiency of residual TPH by microbes was about 8%. Our results are consistent with their findings in that the removal of residual pollutants is higher by adding nutrients after Fenton reagent and modified Fenton reagent pre-treatment, while the activated persulfate and potassium permanganate show opposite effect. Consequently, the choice of chemical oxidant has a direct effect on the subsequent recovery of microbial quantity and activity and overall remediation efficiency. Also, the Fenton reagent and modified Fenton reagent have little effect on the respiration rate of microbes compared to potassium permanganate (Sutton et al., 2014). Thus, microbes are more effective with Fenton reagents, and the removal rate of residual PAHs after pre-treatment with modified Fenton reagent was higher. It is worth noting that the activated persulfate showed the highest PAHs removal efficiency with an increase of 10.24–13.74% after the addition of nutrients, which may have wider practical applications for site remediation.

3.3 Removal of different PAH after adding nutrients

It can be found from table 2 and table 3 that (1) there was a difference in removal of PAHs among different chemical oxidants, (2) the removal efficiency of PAHs with low and middle number rings was increased significantly after adding nutrients, and the PAHs with high rings removal efficiency of microbes was increased only slightly.

The average removal efficiency of the 2–4 rings PAHs (NAP, ANY and ANE, FLE, BaA, CHR) was 52–58% in Fenton reagent treated soils, while the average removal efficiency of 5–6 rings PAHs (BbF, BkF, BaP, I(cd)P, D(ah)A, B(ghi)P) were 79–81% (Fig. 2). In the modified Fenton reagent treated soil, the average removal efficiency of 2–4 rings PAHs was 64–74%, and the removal efficiency of 5–6 rings PAHs was 78–86%. In the activated persulfates and potassium permanganate, the average removal efficiency of 2–4 rings PAHs was about 85%, and the removal efficiency of 5–6 rings PAHs was better than 90%. In

general, the PAHs, such as BkF, BaP, I(cd)P, D(ah)A, B(ghi)P, were most easily oxidized and removed. Whatever oxidant was used, the total removal efficiency can reach more than 70%, and BaA and CHR were difficult to oxidize under the dose conditions of this study.

The removal efficiency of 2–4 rings PAHs was significantly increased under the pre-oxidation conditions of Fenton reagent and modified Fenton reagent, but pre-oxidation did not effect on the removal efficiency of 5–6 rings PAHs. The removal efficiency of NAP and ANY was increased by more than 45%, and the removal efficiency of ANE, FLE, and PHE was increased by about 30% (Fig. 2). The removal efficiency of PAHs increased by about 10% after adding nutrients under the condition of activated persulfate pre-oxidation, and the removal efficiency of a NAP, ANY, ANE and FLE was about 3% higher by potassium permanganate pre-oxidation. In summary, the removal efficiency of 2–4 rings PAHs were enhanced by the addition of nutrients.

The results of this study show that chemical oxidation can remove the PAHs from contaminated soils effectively, and there is a difference in removal efficiency with different chemical oxidants. The modified Fenton reagent removed 55% PAHs in the soil. The normal Fenton reagent was less effective, with only about 50% removal, which was slightly higher than the total removal efficiency (0–43%) of 16 PAHs treated by Fenton reagent in a previous study (Jonsson et al., 2007). Fenton reagents and modified Fenton reagents showed low removal efficiency for the 2-rings PAHs, which were in contrast to the results of Jonsson et al. (2007) that show low-ring PAHs were more susceptible to oxidation than high-ring PAHs. The reason is that some of the high-ring PAHs are decomposed to form low-ring PAHs during the degradation, and these low-ring PAHs have not been completely reacted under the condition of the limited oxidant dose, which makes the removal of low-ring PAHs correspondingly lower. The oxidative effect of the modified Fenton reagent (i.e., the addition of the chelator citric acid on the basis of the Fenton reagent) was significantly higher than that of the Fenton reagent because the Fe^{2+} in the Fenton reagent was quickly oxidized and added with no chelating agent. The mixture can stabilize the ferrous ions, allowing the ferrous ions to be released slowly and to react with hydrogen peroxide to form hydroxyl radicals, thereby effectively removing PAHs in the soil.

Potassium permanganate and activated sodium persulfate are very effective for the removal of most PAHs but are less effective for some PAHs such as CHR and BaA. Brown et al. (2003) and de Souza e Silva et al. (2009) also showed that the removal efficiency of PYR, PHE, ANT, and BaP was more than 50% for both potassium permanganate and activated sodium persulfate, while the removal efficiency of CHR was only 7.8% for potassium permanganate. The difference of PAHs removal efficiency is caused by the difference of PAHs reactivity, which is closely related to the electronic structure (Clar structure) of PAHs (Brown et al., 2003). Therefore, selection of appropriate type of oxidant in an actual remediation project must be based on the remediation objectives and the type of target pollutants.

Microbial degradation depends on the chemical structure of PAHs, the content of pollutants, microbial enzyme activity and soil texture and other factors. Each microbe has a different tolerance, degradation rate and degradation ability for specific PAHs. In general, low PAHs

are relatively more susceptible to biodegradation relative to high-ring PAHs (Pannu et al., 2003). Studies have shown that microbial degradation of PAHs have two pathways: one is to use PAHs as the sole carbon source, such as NAP, PHE and other small molecules that can be directly consumed by microbial degradation; the other is to use PAHs and other organic matter for metabolism, such as BaP. Microbial degradation of macromolecular PAHs is generally carried out in a co-metabolic manner. In the presence of other carbon sources, the microbial activity is enhanced, and the efficiency of degradation of nonessential growing substances is increased, known as co-metabolism (Ghosal et al., 2016). Juhasz et al. (2000) demonstrated that the addition of nutrients to the soil can improve the activity of microbial enzymes and promote the degradation of PYR metabolic degradation process. Kuppusamy et al. (2017) reviewed that the addition of nutrients resulted in the degradation of PAHs in the soil, but only in the low and medium ring PAHs, which was similar to the results of this study.

Due to the difficulty in removing all kinds of PAHs by chemical oxidation, it is interesting to consider the selective addition of nutrients or exogenous bacteria after chemical oxidation, to more effectively remove PAHs in the soil. We speculate that in the process of reaction, some high-ring PAHs will break down to form a low-ring PAHs. Upon the limited dose of oxidant, the low-ring PAHs has not been completely reacted, which makes the low ring PAHs removal rate decrease, and the specific mechanism remains to be further studied. And, microbes are more likely to use low-ring PAHs (such as phenanthrene) as substrates. According to the results of this study, it is possible to use chemical oxidation pre-treatment to degrade PAHs in the soil, and then add nutrients to activate the growth and activity of indigenous microbes, so as to achieve a more a complete degradation of PAHs.

4 Conclusions

A complementary strategy of chemical oxidation combined with microbial remediation was proposed for treating PAHs contaminated soils. The population of microbes in the soils decreased first with the addition of chemical oxidants and then rebounded after the addition of nutrients. The removal efficiency of PAHs by chemical oxidation was in the order: potassium permanganate > activated persulfate > Modified Fenton reagent > Fenton reagent. However, the total removal efficiency of PAHs with the complementary strategy is higher under activated persulfate treatment than that of potassium permanganate oxidation. All tested chemical oxidants exhibited higher removal efficiencies for the 5–6 ring PAHs, but microbial remediation is beneficial to a further degradation for 2–4 rings PAHs. As a whole, combined chemical oxidation with microbial degradation show promising results that may apply to real PAHs contaminated site remediation.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgements

The authors acknowledge the main financial support from the Key Research Program of the Chinese Academy of Sciences (KFZD-SW-303) and the Science and Technology Plan of Beijing (D16110900470000). This research was

not funded by U.S. Environmental Protection Agency (EPA); the views, interpretations, and conclusions expressed in the article are solely those of the authors and do not necessarily reflect or represent the EPA's views or policies. Any use of trade, product, or firm names is for descriptive purposes only and does not imply endorsement by the EPA or the US Government.

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Fig. 1.

Changes of microbe counts in soil G under different treatment conditions.

Fig. 2. Changes of microbe counts in soil C under different treatment conditions. Note: The inner plot indicate the number of microbes treated by potassiumpermanganate.

Fig. 3.
Concentration of total PAHs in soil G under various oxidations.

Fig. 4.
Concentration of total PAHs in soil C under various oxidations.

Table 1.

Physical and chemical characteristics of soils used in this study (n = 3).

Parameters	Coal gas plant soil (G)	Coal plant soil (C)
pH	7.41 ± 0.13	7.57 ± 0.16
TOC/%	0.27 ± 0.05	2.29 ± 0.56
Depth of soil	20–25 cm	20–25 cm
Texture classification	Loam	Sandy loam
NAP/mg·kg ⁻¹	16.9 ± 9.6	16.4 ± 3.96
ANY/mg·kg ⁻¹	15.2 ± 2.1	14.69 ± 8.8
ANE/mg·kg ⁻¹	39.6 ± 5.5	101.98 ± 6.8
FLE/mg·kg ⁻¹	26.3 ± 5.5	75.2 ± 6.49
PHE/mg·kg ⁻¹	28.1 ± 5.4	51.1 ± 16.7
ANT/mg·kg ⁻¹	18.2 ± 6.0	29.18 ± 7.76
FLA/mg·kg ⁻¹	18.5 ± 6.9	34.29 ± 12.6
PYR/mg·kg ⁻¹	15.3 ± 3.6	17.29 ± 3.7
BaA/mg·kg ⁻¹	12.7 ± 5.5	11.1 ± 1.58
CHR/mg·kg ⁻¹	15.0 ± 6.6	9.2 ± 1.1
BbF/mg·kg ⁻¹	10.8 ± 4.0	9.37 ± 1.1
BkF/mg·kg ⁻¹	9.7 ± 2.6	6.4 ± 3.1
BaP/mg·kg ⁻¹	12.0 ± 3.0	5.8 ± 2.9
I(cd)P/mg·kg ⁻¹	7.7 ± 4.0	1.3 ± 0.3
D(ah)A/mg·kg ⁻¹	7.5 ± 3.1	1.0 ± 0.3
B(ghi)P/mg·kg ⁻¹	10.1 ± 8.8	0.9 ± 0.1
Σ16 PAHs/mg·kg ⁻¹	263.6 ± 73.3	385.2 ± 39.6

Table 2.

Removal efficiency of coal gas plant soil (G) of PAHs for different chemical oxidants (%).

	Fenton-CK	Fenton-Nutrition	M-Fenton-CK	M-Fenton-Nutrition	KMnO ₄ -CK	KMnO ₄ -Nutrition	A-PS-CK	A-PS-Nutrition
NAP	51.0 ± 5.6	94.9 ± 3.7	83.6 ± 4.5	99.6 ± 0.6	87.6 ± 2.5	90.1 ± 2.7	89.0 ± 2.6	97.7 ± 3.7
ANY	49.7 ± 2.0	93.0 ± 4.2	79.0 ± 4.1	97.0 ± 4.4	86.1 ± 2.5	87.8 ± 2.6	97.2 ± 2.1	97.1 ± 5.0
ANE	55.7 ± 1.8	85.5 ± 1.4	80.7 ± 2.3	89.4 ± 1.8	85.9 ± 1.2	88.7 ± 1.8	97.4 ± 2.1	97.4 ± 4.1
FLE	55.3 ± 4.5	77.5 ± 2.3	81.3 ± 3.2	90.1 ± 1.0	85.8 ± 1.2	88.1 ± 1.2	91.8 ± 2.2	94.6 ± 1.7
PHE	65.3 ± 5.4	69.0 ± 8.6	73.2 ± 1.9	93.6 ± 2.9	88.1 ± 7.2	84.5 ± 6.3	91.1 ± 2.3	95.4 ± 0.6
ANT	78.1 ± 5.3	93.8 ± 5.8	84.9 ± 3.0	89.3 ± 4.2	93.8 ± 3.6	93.7 ± 3.6	85.4 ± 3.0	93.0 ± 2.1
FLA	74.3 ± 3.9	81.7 ± 3.9	76.6 ± 4.2	92.4 ± 1.6	96.6 ± 1.0	95.7 ± 0.6	92.8 ± 3.0	95.3 ± 1.5
PYR	73.4 ± 3.0	77.7 ± 5.8	86.1 ± 1.1	96.6 ± 2.6	95.0 ± 2.8	95.0 ± 2.2	87.3 ± 4.0	94.5 ± 2.2
BaA	58.5 ± 5.4	58.9 ± 6.3	81.8 ± 3.3	95.2 ± 3.6	96.7 ± 1.6	94.4 ± 2.6	84.1 ± 4.4	89.0 ± 0.5
CHR	59.0 ± 5.8	46.5 ± 11.3	75.0 ± 5.5	90.9 ± 6.5	93.0 ± 1.5	92.5 ± 2.5	85.3 ± 4.5	87.1 ± 3.7
BbF	63.7 ± 4.3	65.0 ± 11.8	79.0 ± 2.0	93.5 ± 5.2	91.9 ± 0.9	92.9 ± 1.4	88.6 ± 4.2	86.2 ± 2.2
BkF	74.7 ± 2.5	64.4 ± 11.1	83.56 ± 2.0	88.0 ± 7.5	94.1 ± 0.9	96.5 ± 0.3	90.06 ± 4.0	84.0 ± 2.6
BaP	67.5 ± 4.2	65.9 ± 1.6	76.9 ± 2.4	82.8 ± 3.9	95.4 ± 0.5	95.6 ± 1.0	88.1 ± 3.7	89.9 ± 1.8
I(cd)P	75.8 ± 2.7	69.4 ± 3.0	77.5 ± 2.7	85.8 ± 0.4	93.9 ± 2.0	96.9 ± 0.2	87.1 ± 3.6	86.4 ± 0.2
D(ah)A	79.6 ± 3.8	70.8 ± 2.0	85.3 ± 1.9	85.2 ± 3.8	97.1 ± 2.3	98.3 ± 1.3	90.0 ± 3.5	94.0 ± 2.1
B(ghi)P	85.5 ± 3.0	83.9 ± 0.5	91.3 ± 2.2	87.2 ± 2.2	99.3 ± 0.7	99.6 ± 0.3	98.5 ± 3.7	99.7 ± 0.2

Note: "CK" means treated with sterile water.

Table 3.

Removal efficiency of coking plant soil (C) of PAHs for different chemical oxidants (%).

	Fenton-CK	Fenton-Nutrition	M-Fenton-CK	M-Fenton-Nutrition	KMnO ₄ -CK	KMnO ₄ -Nutrition	A-PS-CK	A-PS-Nutrition
NAP	48.7 ± 6.6	95.2 ± 2.5	65.1 ± 1.7	98.3 ± 1.6	90.9 ± 4.6	88.8 ± 4.1	98.2 ± 5.2	99.6 ± 0.1
ANY	45.0 ± 5.5	90.3 ± 5.1	61.5 ± 5.1	90.1 ± 2.9	87.8 ± 3.4	88.3 ± 2.5	80.9 ± 11.0	93.5 ± 3.1
ANE	56.4 ± 2.5	90.5 ± 0.6	65.3 ± 6.3	93.8 ± 0.9	90.0 ± 3.5	92.4 ± 3.4	80.6 ± 7.2	96.6 ± 0.7
FLE	60.4 ± 3.0	89.2 ± 0.5	66.0 ± 7.6	93.4 ± 1.4	93.6 ± 2.8	96.4 ± 1.7	84.2 ± 7.4	96.9 ± 1.3
PHE	60.6 ± 4.3	80.1 ± 3.1	74.4 ± 5.7	91.5 ± 3.2	97.8 ± 3.2	90.3 ± 2.5	84.6 ± 4.4	97.1 ± 0.5
ANT	70.0 ± 3.8	60.6 ± 2.6	75.8 ± 3.0	91.1 ± 3.0	93.7 ± 1.2	95.3 ± 0.9	78.0 ± 4.3	95.9 ± 1.5
FLA	65.2 ± 2.5	64.4 ± 3.2	61.1 ± 1.3	67.1 ± 2.4	91.2 ± 2.2	92.9 ± 2.5	88.2 ± 3.3	86.4 ± 1.0
PYR	56.9 ± 5.7	55.8 ± 4.2	72.8 ± 3.5	69.2 ± 10.1	86.2 ± 2.4	87.7 ± 2.7	83.3 ± 6.1	85.0 ± 3.2
BaA	59.5 ± 3.9	46.0 ± 4.8	71.8 ± 5.2	67.9 ± 7.3	85.5 ± 1.5	86.2 ± 1.1	83.6 ± 5.0	83.8 ± 6.7
CHR	60.7 ± 7.2	76.2 ± 4.7	68.4 ± 7.2	65.1 ± 1.7	91.5 ± 2.1	91.9 ± 2.1	85.6 ± 4.7	87.1 ± 1.8
BbF	63.0 ± 4.2	77.7 ± 5.1	79.9 ± 2.7	78.1 ± 4.4	91.3 ± 1.3	91.9 ± 1.5	91.4 ± 2.4	89.7 ± 1.7
BkF	65.0 ± 1.3	65.4 ± 3.1	81.9 ± 2.4	84.6 ± 4.8	94.4 ± 1.0	94.9 ± 1.0	91.8 ± 3.5	90.0 ± 4.1
BaP	74.0 ± 5.0	67.5 ± 0.5	76.9 ± 3.5	84.8 ± 2.9	95.7 ± 0.6	95.9 ± 0.7	93.6 ± 1.2	93.0 ± 1.9
I(cd)P	74.8 ± 6.6	74.8 ± 3.3	75.7 ± 2.5	81.9 ± 2.9	95.2 ± 0.6	95.5 ± 0.6	94.0 ± 1.0	91.1 ± 2.0
D(ah)A	75.7 ± 1.7	75.5 ± 3.3	82.4 ± 2.5	82.8 ± 7.6	89.0 ± 3.9	90.4 ± 4.1	89.5 ± 6.8	91.8 ± 3.6
B(ghi)P	84.6 ± 2.9	73.6 ± 2.4	84.9 ± 5.0	69.3 ± 8.3	98.2 ± 0.5	99.1 ± 0.3	98.2 ± 1.9	96.3 ± 2.3

Note: "CK" means treated with sterile water.