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CONTENTS

Aquatic environment

Applicable models for multi-component adsorption of dyes: A review Babak Noroozi, George A. Sorial	419
Effects of sludge dredging on the prevention and control of algae-caused black bloom in Taihu Lake, China Wei He, Jingge Shang, Xin Lu, Chengxin Fan	430
Distribution characteristics and source identification of polychlorinated dibenzo- <i>p</i> -dioxin and dibenzofurans, and dioxin-like polychlorinated biphenyls in the waters from River Kanzaki, running through Osaka urban area, Japan Masao Kishida	441
Pre-oxidation with KMnO ₄ changes extra-cellular organic matter's secretion characteristics to improve algal removal by coagulation with a low dosage of polyaluminium chloride Lei Wang (female), Junlian Qiao, Yinghui Hu, Lei Wang (male), Long Zhang, Qiaoli Zhou, Naiyun Gao	452
Identification of causative compounds and microorganisms for musty odor occurrence in the Huangpu River, China Daolin Sun, Jianwei Yu, Wei An, Min Yang, Guoguang Chen, Shujun Zhang	460
Influences of perfluorooctanoic acid on the aggregation of multi-walled carbon nanotubes Chengliang Li, Andreas Schäffer, Harry Vereecken, Marc Heggen, Rong Ji, Erwin Klumpp	466
Rapid degradation of hexachlorobenzene by micron Ag/Fe bimetal particles Xiaoqin Nie, Jianguo Liu, Xianwei Zeng, Dongbei Yue	473
Removal of Pb(II) from aqueous solution by hydrous manganese dioxide: Adsorption behavior and mechanism Meng Xu, Hongjie Wang, Di Lei, Dan Qu, Yujia Zhai, Yili Wang	479
Cr(VI) reduction capability of humic acid extracted from the organic component of municipal solid waste Barbara Scaglia, Fulvia Tambone, Fabrizio Adani	487
Off-flavor compounds from decaying cyanobacterial blooms of Lake Taihu Zhimei Ma, Yuan Niu, Ping Xie, Jun Chen, Min Tao, Xuwei Deng	495
Pollutant concentrations and pollution loads in stormwater runoff from different land uses in Chongqing Shumin Wang, Qiang He, Hainan Ai, Zhentao Wang, Qianqian Zhang	502

Atmospheric environment

Influence of fuel mass load, oxygen supply and burning rate on emission factor and size distribution of carbonaceous particulate matter from indoor corn straw burning (Cover story) Guofeng Shen, Miao Xue, Siye Wei, Yuanchen Chen, Bin Wang, Rong Wang, Huizhong Shen, Wei Li, Yanyan Zhang, Ye Huang, Han Chen, Wen Wei, Quyu Zhao, Bin Li, Haisu Wu, Shu Tao	511
Synergistic impacts of anthropogenic and biogenic emissions on summer surface O ₃ in East Asia Yu Qu, Junling An, Jian Li	520
Effect of central ventilation and air conditioner system on the concentration and health risk from airborne polycyclic aromatic hydrocarbons Jinze Lv, Lizhong Zhu	531
Emission inventory evaluation using observations of regional atmospheric background stations of China Xingqin An, Zhaobin Sun, Weili Lin, Min Jin, Nan Li	537
An improved GC-ECD method for measuring atmospheric N ₂ O Yuan Yuan Zhang, Yujing Mu, Shuangxi Fang, Junfeng Liu	547
Adsorption of carbon dioxide on amine-modified TiO ₂ nanotubes Fujiao Song, Yunxia Zhao, Qin Zhong	554

Terrestrial environment

Factors influencing the contents of metals and As in soils around the watershed of Guanting Reservoir, China Li Xu, Tiejue Wang, Wei Luo, Kun Ni, Shijie Liu, Lin Wang, Qiushuang Li, Yonglong Lu	561
Photolysis of polycyclic aromatic hydrocarbons on soil surfaces under UV irradiation Chengbin Xu, Dianbo Dong, Xuelian Meng, Xin Su, Xu Zheng, Yaoyao Li	569
Sorption and transport studies of cetyl trimethylammonium bromide (CTAB) and Triton X-100 in clayey soil Sivaram Harendra, Kumaraswamy Vipulanandan	576

Environmental biology

Effects of soil water and nitrogen availability on photosynthesis and water use efficiency of <i>Robinia pseudoacacia</i> seedlings Xiping Liu, Yangyang Fan, Junxia Long, Ruifeng Wei, Roger Kjellgren, Chunmei Gong, Jun Zhao	585
Phytoremediation potential of charophytes: Bioaccumulation and toxicity studies of cadmium, lead and zinc Najjapak Sooksawat, Metha Meetam, Maleeya Kruatrachue, Prayad Pokethitiyook, Koravid Nathalang	596
Sulfur speciation and bioaccumulation in camphor tree leaves as atmospheric sulfur indicator analyzed by synchrotron radiation XRF and XANES Jianrong Zeng, Guilin Zhang, Liangman Bao, Shilei Long, Mingguang Tan, Yan Li, Chenyan Ma, Yidong Zhao	605
Hydrocarbon biodegradation and dynamic laser speckle for detecting chemotactic responses at low bacterial concentration Melina Nisenbaum, Gonzalo Hernán Sendra, Gastón Alfredo Cerdá Gilbert, Marcelo Scagliola, Jorge Froilán González, Silvia Elena Murialdo	613

Environmental health and toxicology

Biogeochemical reductive release of soil embedded arsenate around a crater area (Guandu) in northern Taiwan using X-ray absorption near-edge spectroscopy Kai-Ying Chiang, Tsan-Yao Chen, Chih-Hao Lee, Tsang-Lang Lin, Ming-Kuang Wang, Ling-Yun Jang, Jyh-Fu Lee	626
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Photolysis of polycyclic aromatic hydrocarbons on soil surfaces under UV irradiation

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Abstract

Photolysis of some polycyclic aromatic hydrocarbons (PAHs) on soil surfaces may play an important role in the fate of PAHs in the environment. Photolysis of PAHs on soil surfaces under UV irradiation was investigated. The effects of oxygen, irradiation intensity and soil moisture on the degradation of the three PAHs were observed. The results showed that oxygen, soil moisture and irradiation intensity enhanced the photolysis of the three PAHs on soil surfaces. The degradation of the three PAHs on soil surfaces is related to their absorption spectra and the oxidation-half-wave potential. The photolysis of PAHs on soil surfaces in the presence of oxygen followed pseudo first-order kinetics. The photolysis half-lives ranged from 37.87 days for benzo[a]pyrene to 58.73 days for phenanthrene. The results indicate that photolysis is a successful way to remediate PAHs-contaminated soils.

Key words: photolysis; soil surfaces; polycyclic aromatic hydrocarbons; UV irradiation

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Introduction

Polycyclic aromatic hydrocarbons (PAHs), produced mainly by the incomplete combustion of fossil fuels, are known to be widespread pollutants with high toxicity. Many PAHs are carcinogenic and mutagenic (Liu et al., 2007; Ma and Zhou, 2011; Na et al., 2011; Sun et al., 2010). Some studies in recent years showed that quite a few PAHs have photo-induced toxicity (Huovinen et al., 2001; Arulazhagan et al., 2011; Yan et al., 2012). In the US EPA priority pollutants list, there are 16 PAHs included. Thus it is of great importance to investigate the transport and transformation of PAHs in the environment.

The main processes which successfully remove and eliminate PAHs from the environment include: microbiological transformation and degradation, volatilization, and photolysis (Dong et al., 2010a, 2010b; Ding et al., 2008). However, some of the high molecular weight (HMW) PAHs are refractory, and biodegradation is difficult for most PAHs (Zhang et al., 2006; Gu et al., 2010; He et al., 2009). Photolysis is one of the major abiotic transformation processes for PAHs in the environment (Zepp and Schlotzhauer, 1979). Over the past few years, much

work has been done in characterizing photolytic reactions of PAHs in the aqueous phase (Lehto et al., 2000; Sanches et al., 2011; Leite et al., 2011). The photolysis mechanisms of PAHs in the liquid phase were described by Miller and Olejnik (2001) and An and Carraway (2002). Niu et al. (2007) reported the photolysis of PAHs associated with fly ash particles under simulated sunlight irradiation. Sotero and Arce (2008) reported the major products in the photochemistry of perylene adsorbed on models of atmospheric particulate matter. A number of studies on the adsorption of PAHs on silica and TiO₂ have also been reported (David and Boule, 1993; Reyes et al., 1998; Kefi et al., 2011). However, comparatively little research effort has been directed at examining the photolysis of PAHs on soil surfaces, and the fate of PAHs adsorbed on soil surfaces has not been fully explored.

In China, the background levels of PAHs in arable soils vary between 10–20 µg/kg, while soil PAH concentrations have been found to be as high as 10²–10⁴ µg/kg in some contaminated areas (Li et al., 2006, 2011), which may pose great threats to agricultural food quality, human health, and ecological security. Thus considerable attention has been paid to the remediation of contaminated soils. The purpose of this study therefore was to investigate the photolysis of

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PAHs on soil surfaces under UV irradiation. Phenanthrene, pyrene and benzo[a]pyrene were chosen as representative compounds for PAHs. The kinetic model and the influence of several parameters on the course of photolysis are discussed.

1 Materials and methods

1.1 Chemicals

Phenanthrene, pyrene and benzo[a]pyrene were purchased from Fluka, Germany, and used without further purification. Methanol (HPLC grade) was purchased from Shandong Yuwang Company, China. Hexane and dichloromethane (analytical grade) were purchased from Tianjin Concord Technical Company, China. NaN_3 was purchased from Zhejiang Hailan Chemical Co., Ltd., China.

1.2 Soil samples

Surface soil samples (0–10 cm) were collected from the Ecological Station of the Shenyang, Institute of Applied Ecology, Chinese Academy of Sciences. The soil samples were sterilized by soaking in NaN_3 solution for 48 hr and then dried in an oven at 105°C (Zhao et al., 2009). After drying, the soil samples were passed through a 1-mm sieve. The sieved soil samples were then stored in a dark chamber before use. The characteristics were as follows: the contents of sand, silt, and clay were 21.4 wt.%, 46.5 wt.%, and 32.1 wt.%, respectively; organic matter 1.78 wt.%; density 2.53 g/cm³; pH 6.8. Mineral components of the experimental soil samples were analyzed by X-ray fluorescence (XRF) and the results are listed in **Table 1**.

For the individual experiments, the soil samples were treated with methanol solutions of phenanthrene, pyrene and benzo[a]pyrene, mixed thoroughly and then air-dried to evaporate the methanol. The concentrations of phenanthrene, pyrene and benzo[a]pyrene in the resulting soil samples were 40 mg/kg, respectively.

1.3 Photolysis experiments

The photolysis experiments were conducted using an YQX-1 anaerobic photodegradation chamber (Yuejing Company, China). The UV lamps (Phillips ATLD 20W, Model UVA, Phillips, Holland) had a wavelength of 253.7

nm. In all photolysis experiments, three replicates of 5 g of soil samples were evenly spread on Petri dishes and randomly located in the anaerobic photodegradation chamber under the UV lamps. Light-proof Petri dishes also containing 5 g of uniformly spread soil samples were used as controls for the measurement of non-photolytic PAHs loss for all experimental treatments. Nitrogen gas was used to flush oxygen from the chamber for the anaerobic photolysis treatments. Petri dishes containing soil samples were placed on shelves 450 mm below the lamps for UV irradiation. The temperature within the chamber was held constant at 25°C using a heater and fans throughout all experiments. Soil samples were taken at 1 day intervals from the Petri dishes exposed to UV light.

The effect of oxygen on the photolysis was investigated by conducting experiments under anaerobic conditions, where nitrogen gas was used to flush oxygen from the chamber, as well as experiments in air. The number of UV lamps was adjusted to vary the light intensity. Three different levels of light intensity were employed in the range 119–357 $\mu\text{W}/\text{cm}^2$. The effect of soil moisture on the degradation of PAHs was also examined. The soil samples on the Petri dishes were treated with a fine spray of sterilized water to give the desired degree (by weight) of moisture. The moisture levels in the soil samples were 0, 10% and 20%, respectively.

1.4 PAH extraction procedure

Concentrations of PAHs in the irradiated and non-irradiated soil samples were obtained by the following method: the soil samples were transferred into 100 mL Teflon tubes and mixed 20.0 mL dichloromethane. Each sample was extracted for 2 hr in an ultrasonic bath in which the water temperature was maintained lower than 35°C . The mixtures were then centrifuged at 4000 r/min for 5 min to separate the supernatant from the soil samples. An aliquot of 0.500 mL of extract was passed through a glass column containing 0.9 g of silica gel wetted with hexane. The column was eluted with 1 mL hexane:dichloromethane (1:1, V/V) four times. The combined eluate was completely dried under a gentle stream of nitrogen. The solid residue was re-dissolved in 1.00 mL of methanol for HPLC analysis. The extraction

Table 1 Main elements and components of the experimental soil

Element	Concentration (mg/kg)	Element	Concentration (mg/kg)	Component	Concentration (%)
As	8.95	Ni	26.70	SiO_2	68.22
Ba	676.65	P	630.30	Al_2O_3	13.41
Ce	123.00	Pb	37.00	Fe_2O_3	4.19
Cl	49.55	Rb	109.75	MgO	1.13
Co	11.70	Sr	189.75	CaO	1.16
Cr	67.65	Th	13.00	Na_2O	1.78
Cu	36.70	Ti	4459.40	K_2O	2.43
Ga	17.90	Zn	70.45		
La	65.90	Zr	276.95		
Mn	577.70	V	76.35		

efficiencies of phenanthrene, pyrene and benzo[a]pyrene were $92.1\% \pm 2.3\%$, $90.4\% \pm 3.2\%$, and $86.7\% \pm 2.8\%$, respectively.

1.5 Analytical methods

Quantification of PAHs in methanol solutions was conducted using an Agilent model 1200 HPLC with a variable wavelength detector set at 254 nm (Agilent Technologies, USA). The mobile phase used was methanol:water (90:10, V/V) at a flow rate of 0.7 mL/min. Injection volume was 10 μ L. Three replicates were performed for each analysis.

The molar absorption spectra of phenanthrene, pyrene, and benzo[a]pyrene were measured by U-3100 UV-Vis spectrophotometer (Hitachi, Japan).

1.6 Statistical evaluation

All experiments were performed in triplicate. Data were statistically analyzed using SPSS 13.0 for windows (SPSS Inc., Chicago, Illinois). The Paired-Samples *T* test was used to evaluate the statistical significance of differences amongst mean values.

2 Result and discussion

2.1 Anaerobic photolysis

The degradation of three PAHs under anaerobic UV irradiation measured at 1 day intervals over 5 days is shown in Fig. 1. It was observed that the degradation of all three PAHs increased along with increasing irradiation time.

The photolysis of the three PAHs on soil surfaces under anaerobic conditions is the result of photoreaction. A successful photoreaction requires that the emission spectrum of the light source should overlap the absorption spectra of the target compounds (Miller and Olejnik, 2001). The molar absorption spectra of phenanthrene, pyrene, and benzo[a]pyrene are shown in Fig. 2. Their structural differences are reflected in the absorption characteristics

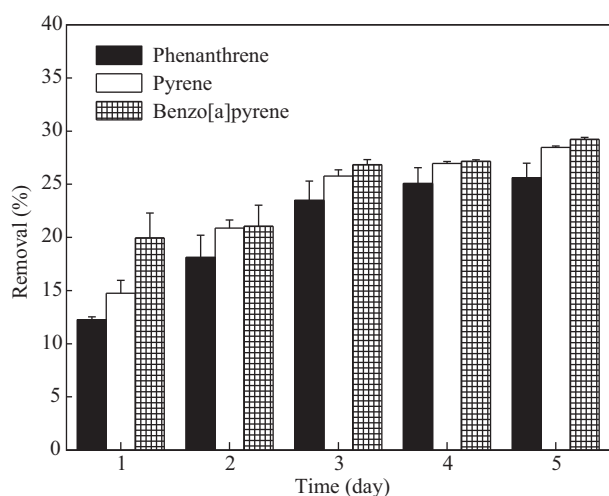


Fig. 1 Photolysis of the three PAHs on natural soil surfaces under anaerobic conditions.

of each compound. All three compounds absorb light at wavelengths ranging from 210 to 400 nm. The absorption spectra of the three compounds consist of several bands of various intensities. The phenanthrene absorption peaks are at 268, 299, 330 and 352 nm, pyrene absorption peaks at 220, 242, 268, 310, 329, 347 and 374 nm, and benzo[a]pyrene absorption peaks at 220, 259, 269, 329, 350 and 380 nm. Hence, the three PAHs have the potential to undergo photoreaction with any UV source having a wavelength about 253.7 nm (Shemer and Linden, 2007).

At all time intervals, the degradation of the three PAHs under anaerobic conditions was in the sequence benzo[a]pyrene > pyrene > phenanthrene. According to photochemical theory, the photoreaction when the PAHs molecules absorb energy, causing their excitation, and the excited molecules then undergo chemical changes. Thus, the photolysis of PAHs is highly dependent on light absorption. As shown in Fig. 2, the phenanthrene absorption peaks at more than 253.7 nm have less intensity than those of pyrene and benzo[a]pyrene, and the benzo[a]pyrene absorption peaks at more than 253.7 nm have stronger light absorbance than those of phenanthrene and pyrene. The results indicate that the degradation degree of PAHs on soil surfaces is related to their molar absorption spectra on soil surfaces. Hence, the more absorption peaks and stronger light absorbance at wavelengths greater than 253.7 nm the PAHs have, the more easily the PAHs molecules degrade.

2.2 Oxygen effect

A comparison of the degradation rates of phenanthrene, pyrene and benzo[a]pyrene in the absence and presence of oxygen within 5 days is shown in Fig. 3. It was observed that the degradation of the three PAHs in both treatments all increased along with increasing irradiation time. The three PAHs undergo photoreaction in addition to photooxidation in the presence of oxygen, and therefore the degradation rate of the three PAHs in the absence of oxygen was consistently lower than in the presence of oxygen. The results indicated that the oxygen enhanced the degradation of the three PAHs on soil surfaces.

The pseudo first-order rate equation (Niu et al., 2003):

$$-\ln \frac{C_t}{C_0} = kt + c \quad (1)$$

where, C_0 (mg/kg) and C_t (mg/kg) are the PAHs concentrations at irradiation time zero and t (day), respectively, and k is the rate constant, can be applied to the results of the current study. Linear relationships were observed between $-\ln(C_t/C_0)$ and time, which demonstrates that photolysis of the three PAHs on soil surfaces in the presence of oxygen follows pseudo first-order kinetics as shown in Fig. 4. Half-lives ($t_{1/2}$) were calculated using Eq. (2), which is derived from Eq. (1) by replacing C_t with $C_{1/2}$:

$$t_{1/2} = \frac{\ln 2}{k} = \frac{0.693}{k} \quad (2)$$

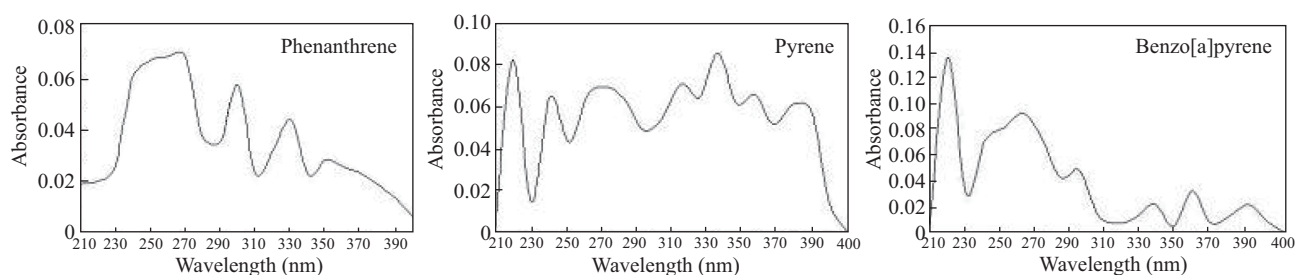


Fig. 2 Molar absorption spectra of phenanthrene, pyrene, and benzo[a]pyrene on soil surfaces.

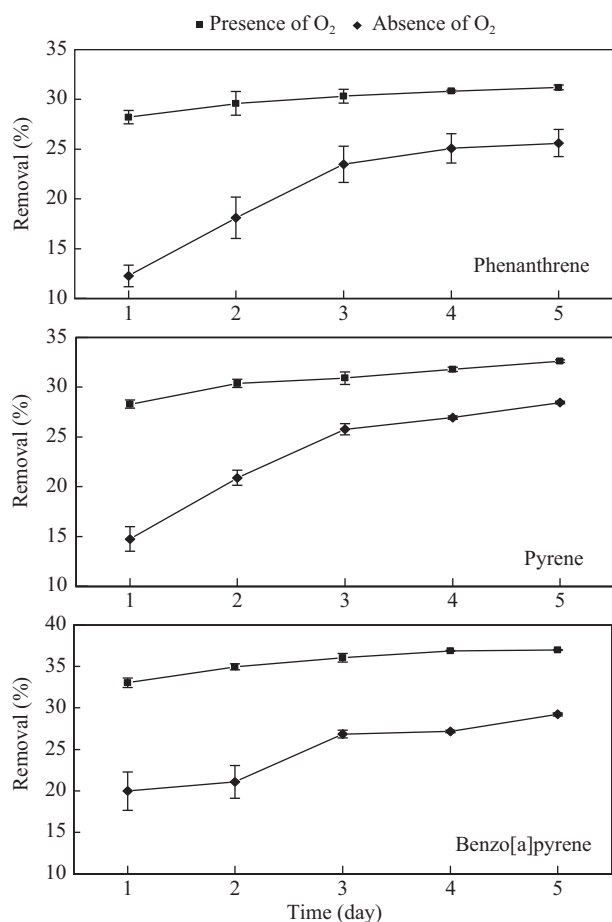


Fig. 3 Photolysis of phenanthrene, pyrene, and benzo[a]pyrene under UV irradiation in the presence and absence of oxygen.

The results are listed in **Table 2**.

According to the rate constants and half-lives, the degradation of the three PAHs under aerobic conditions also follows the sequence benzo[a]pyrene > pyrene > phenanthrene. The degradation is the result of the direct photolysis and photooxidation. There are two reasons: Firstly, photolysis of the three PAHs has already been discussed, and the degree of degradation of the three PAHs is related to their absorption spectra on soil surfaces. Secondly, the oxidation-half-wave potential ($E_{1/2}$) illustrates the difficulty of photooxidation of the three PAHs. The lower the $E_{1/2}$, the more easily the PAH can be oxidized. The half wave potentials are 1.27, 1.50 and 1.54 for benzo[a]pyrene, phenanthrene and pyrene respectively;

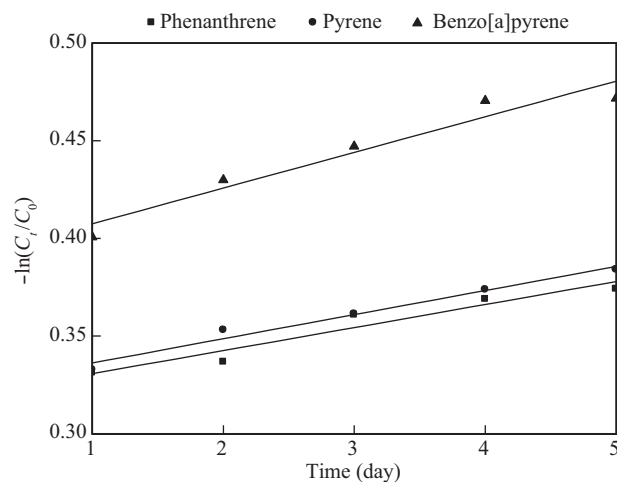


Fig. 4 Degradation kinetics of PAHs in presence of oxygen under UV irradiation.

there is a trend for degradation to increase with decreasing $E_{1/2}$ (Pan et al., 1999).

2.3 Irradiation intensity effect

The effect of irradiation intensity in the range of 119–357 $\mu\text{W}/\text{cm}^2$ on the degradation of phenanthrene, pyrene, benzo[a]pyrene was examined over 5 days, as shown in **Fig. 5**. It was observed that the degradation of the three PAHs all increased with increasing irradiation time for all three treatments. The results indicated that the irradiation intensity enhanced the degradation of the three PAHs on soil surfaces.

In general, when using UV irradiation the degradation gradually increases with increasing irradiation intensity. The reason may be as follows: First, theoretically, at a given depth z , the rate of photoreaction is assumed to obey a first-order rate law (Balmer, 2000):

$$\frac{dC(z)}{dt} = -k_p(z)C(z) \quad (3)$$

where, $C(z)$ (mg/kg) is the concentration of the compound

Table 2 Kinetic parameters for PAH photodegradation in the presence of oxygen

Compound	k (day^{-1})	$t_{1/2}$ (day)	R^2
Phenanthrene	0.0118	58.73	0.9338
Pyrene	0.0123	56.34	0.9766
Benzo[a]pyrene	0.0183	37.87	0.9333

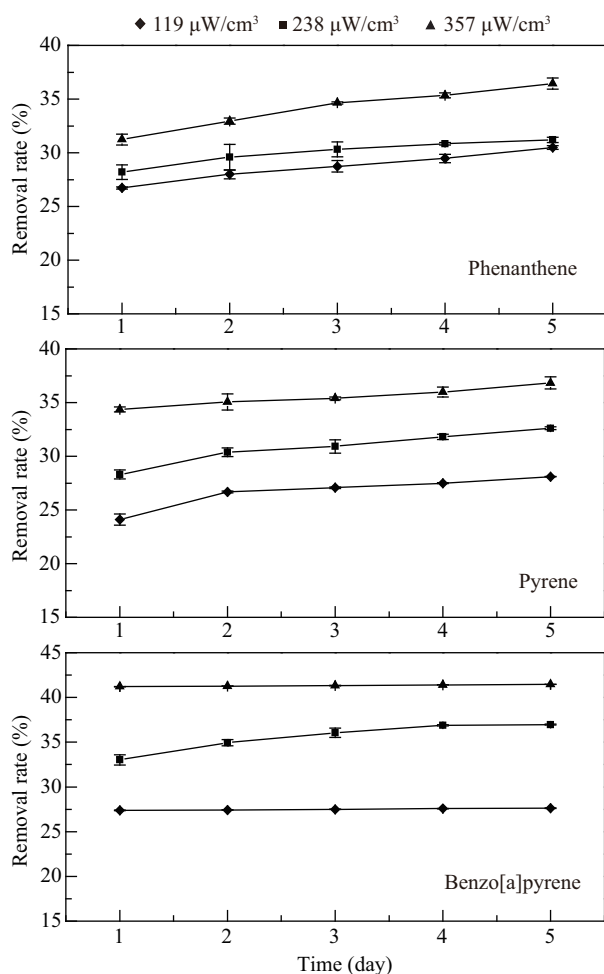


Fig. 5 Effect of irradiation intensity on phenanthrene, pyrene, and benzo[a]pyrene degradation.

expressed as mass of compound per mass solid phase, and $k_p(z)$ is the first-order photolysis rate constant at the depth z (cm^{-1}). In analogy to photoreaction in aqueous solution, k_p^0 may be rationalized by

$$k_p^0 = 2.303 \int [I_0(\lambda)\epsilon(\lambda)\Phi(\lambda)d(\lambda)] \quad (4)$$

where, $I_0(\lambda)$ is the light intensity on the top of soil surface, and $\epsilon(\lambda)$ and $\Phi(\lambda)$ are the molar absorption coefficient and the quantum yield of photolysis of the compound as a function of the wavelength, λ , respectively. Note that both $\epsilon(\lambda)$ and $\Phi(\lambda)$ of a compound adsorbed on soil surfaces may be quite different from the corresponding values determined in aqueous solution. Therefore, prediction of k_p^0 is presently not possible. To relate $k_p(z)$ to k_p^0 , the first-order photolysis rate constant on soil surfaces, it is assumed that the light intensity decreases exponentially with depth and that light is only attenuated by solid (i.e., self-filtering effect is negligible). The attenuation of light at a given wavelength can then be described by a characteristic depth, $z_{0.5}(\lambda)$, over which the light intensity decreases to half. Hence, the determined values for k_p^0 and

$z_{0.5}(\lambda)$ are the weighted average values for the whole range of wavelengths where light is absorbed by the compound. Thus, $k_p(z)$ can be approximated by

$$k_p(z) = k_p^0 e^{-[z/(1.443z_{0.5}(\lambda))]} \quad (5)$$

Hence, the rate of photoreaction is

$$-d[\text{PAH}]/dt = 2.303e^{-[z/(1.443z_{0.5}(\lambda))]} \int [I_0(\lambda)\epsilon(\lambda)\Phi(\lambda)d(\lambda)] \quad (6)$$

When the light is at given wavelength, the $I_0(\lambda)$ is constant, thus

$$-d[\text{PAH}]/dt = 2.303e^{-[z/(1.443z_{0.5}(\lambda))]} I_0(\lambda) \int [\epsilon(\lambda)\Phi(\lambda)d(\lambda)] \quad (7)$$

when the light energy absorbed, $I_0(\lambda)$, is significantly increased due to increasing irradiation intensity. Higher $I_0(\lambda)$ values increase the degradation rates (Eq. (7)). Hence, the photoreaction of PAHs was enhanced by increased irradiation intensity.

Secondly, under the higher irradiation intensity, the concentration of reactive species increases. The enhancement rates of photooxidation were considerably higher because a higher concentration of reactive species can lead to greater degradation of PAHs on soil surfaces.

2.4 Soil moisture effect

The effect of soil moisture in the range of 0–20% (W/W) on the degradation of PAHs was examined over the course of 5 days. The results are shown in **Fig. 6**. The degradation in all three treatments increased with increasing irradiation time. The level of soil moisture clearly influenced the photolysis of the PAHs on soil surfaces.

When the soil was moist, the degradation of the three PAHs increased along with increasing soil moisture. Similar results have also been reported by Hilarides et al. (1994), Shelton et al. (1995), Frank et al. (2002) and Graebing et al. (2003). Firstly, the superoxide radical anion (O_2^-) can react with water (H_2O) to produce hydroxyl radicals ($\text{OH}\cdot$), which can enhance the photolysis of the three PAHs on soil surfaces. Secondly, there is a direct radiation effect; water greatly increases the amount of radiation absorbed in the soil. Thirdly, with less water available, the mobility of the chemicals is affected; there is more PAHs sorption and less diffusion in drier soils.

3 Conclusions

The degradation of the three PAHs on soil surfaces under both aerobic and anaerobic conditions follows the sequence of benzo[a]pyrene > pyrene > phenanthrene, which

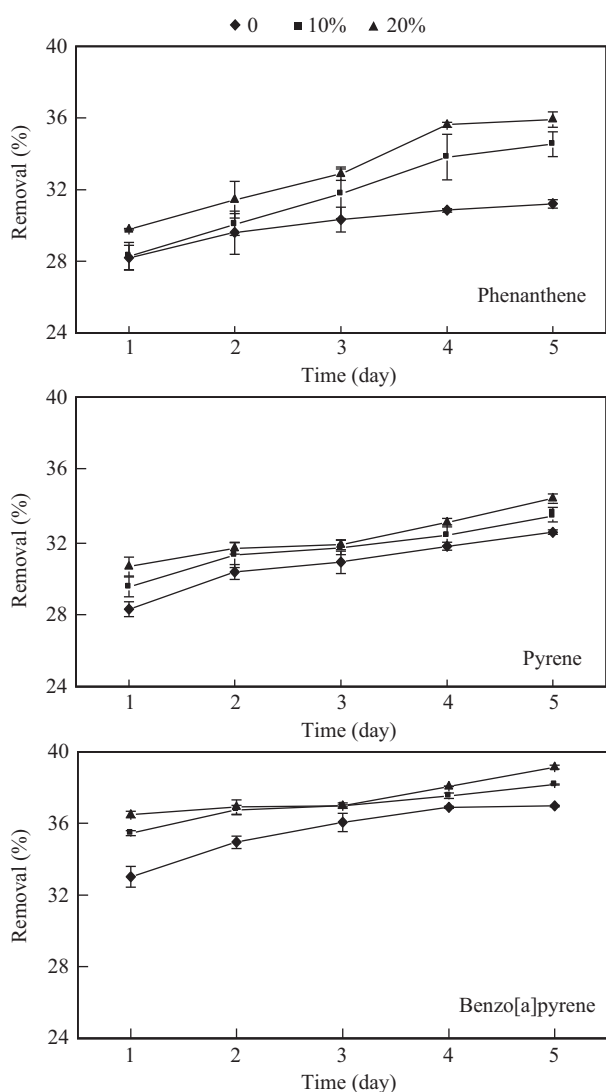


Fig. 6 Effect of soil moisture on phenanthrene, pyrene, and benzo[a]pyrene degradation.

indicates that the degradation of the PAHs is related to the absorption spectra on the soil surfaces and the oxidation-half-wave potentials. The photolysis of the three PAHs on soil surfaces in the presence of oxygen follows first-order kinetics under UV irradiation. The degradation of the three PAHs is affected by oxygen, the irradiation intensity and the soil moisture. Photoreactions and photooxidation would be expected to contribute to the degradation of PAHs on soil surfaces.

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