



Formation of environmentally persistent free radicals during the transformation of anthracene in different soils: Roles of soil characteristics and ambient conditions

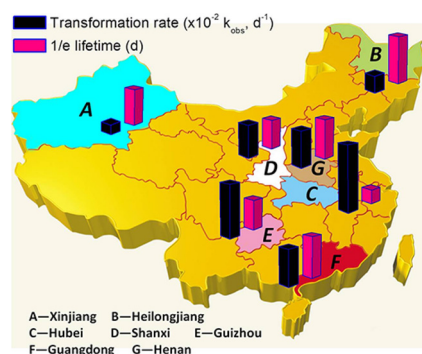


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GRAPHICAL ABSTRACT



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ABSTRACT

Transformation of PAHs, i.e., anthracene, and production of environmentally persistent free radicals (EPFRs) on seven types of representative soils were investigated, focusing on the influences of soil characteristics and ambient conditions on these reactions. The transformation rate of anthracene exhibits the order of red earth > yellow earth > latosol ~ fluvo-aquic soil > brown earth > chernozem > calcic brown soil, which is positively correlated with Fe content in soils. Afterwards, batch reactions on pure representatives of soil minerals, including Fe₂O₃, Fe₃O₄, FeOOH, and MnO₂, demonstrate that anthracene is prone to mineral-promoted transformation. The presence of higher amount of organic carbon lower the transformation rate of anthracene, whereas the formed EPFRs can be stabilized for a longer time. Subsequent experiments associated with the influences of environmental conditions on mineral-promoted reactions suggest that both anthracene transformation and EPFRs generation readily occur under dry condition. Light irradiation not only promotes the formation of EPFRs, but also greatly accelerates the decay of EPFRs and the 1/e lifetime decreases from 5 to 20 d in dark to approximately 1 d. Meanwhile, the anoxic condition is favorable for the persistence of EPFRs. The obtained results suggested the potential environmental risks association with EPFRs in PAHs-contaminated soils.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), mostly discharged by anthropogenic activities such as exploitation and utilization of fossil fuels, have been widely detected in natural environment [1,2]. Particularly, soil and sediment are considered as the crucial sinks and potential reemission sources of PAHs [3]. At superfund sites associated with former coke production, coal mining, and gas works, the total PAH concentrations range from hundreds to thousands of mg kg^{-1} [4–6]. Due to their persistence, bioaccumulation, and toxicity, increased human health risk was caused by the exposure via ingestion of soil or/and inhalation of floating dust directly [7–9]. Therefore, the eco-toxicological effects of PAH-contaminated soils have been paid more attentions among the public, decision-makers and scientists [10].

Detoxification and ecological impacts of PAHs correlate with their natural attenuation processes including biotic and abiotic degradation [11,12]. Biological degradation has long been recognized as a dominant way for their disappearance in soil [13,14]. However, relatively high concentration of PAHs in contaminated sites exhibits toxicity on biomass, and thus preventing or slowing their bio-transformation [15]. Recently, lines of evidence shows that the mineral-promoted reaction, referred as abiotic transformation, can also represent a critical determinant for the fate of PAHs in soil [16–18]. As an example, clay minerals can induce chemical oxidation of PAHs via single-electron-transfer process in soil [18]. Meanwhile, the presence of metal oxides enhances the photodegradation of PAHs on upper layers [19,20]. Oxidation or polymerization of PAHs always occurs during their interaction with minerals containing Mn, Fe, and other transition metals [21–23]. The mineral-mediated transformation might involve in the electron transfer from organic compounds to electron-deficient sites, inducing the production of intermediate radicals [24,25]. Generally, free organic radicals are highly reactive and readily react with available

electron-accepting species, such as O_2 , H_2O , and parent compounds, resulting in final products by coupling, deprotonation, or abstraction termination [21,26]. Under certain conditions, however, the formed free radicals can be stabilized on mineral surfaces, generating novel, sometimes unexpected intermediate products, i.e., environmentally persistent free radicals (EPFRs), which have been considered as emerging contaminants due to their detrimental effects on human health [27,28].

Recently, the PAH-type EPFRs were observed in soil samples collected from former coking sites [29]. The spin densities in those samples correlated with the level of PAHs, transition metals, and organic carbons [29]. Several studies have been carried out to explore the role of metal oxide/ions in the PAHs transformation on mineral surfaces [28,30]. Progress has been made to understand the potential mechanism of EPFRs formation during the interaction between PAHs and individual metal/silica mineral surfaces [28]. Comparatively, only limited studies have been performed examining the PAHs transformation and EPFRs formation on whole soil with multiple metal oxide and mineral mixtures. In addition, the effect of ambient conditions on the production of free organic radicals in PAHs-contaminated soil is still poorly understood, and this information is vital to evaluate the potential risks for PAH-polluted soils with various properties.

In this study, the potential formation of EPFRs was examined during the abiotic transformation of PAHs in sterilized seven model soils obtained around China representing a broad spectrum of soil properties. Anthracene, one of the most widespread PAHs in natural phases, was selected as the target pollutant since our previous study revealed that EPFRs were readily generated during the abiotic transformation of anthracene on mineral surfaces [28]. The correlations between EPFRs production and soil properties were measured to seek the parameters influencing PAHs transformation. Role of environmental factors on the persistence of EPFRs was comprehended by tracing the evolution of



Fig. 1. Profile of the sampling sites.

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