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Research Paper

Influence of compost and biochar on microbial communities and the sorption/degradation of PAHs and NSO-substituted PAHs in contaminated soils

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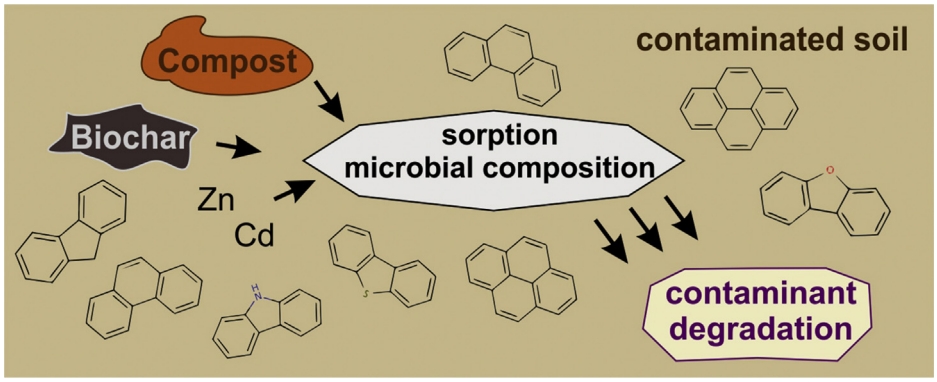
HIGHLIGHTS

- PAH and NSO-PAH sorption in soil increased with both compost and biochar addition.
- Microbial diversity increased with added compost, but did not change with additional biochar.
- Contaminant degradation increased with compost and decreased with biochar addition.
- The combined use of compost and biochar can be used to stabilize contaminated soils.

GRAPHICAL ABSTRACT



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ABSTRACT

Diffusely contaminated soils often remain untreated as classical remediation approaches would be disproportionately expensive. Adding compost can accelerate the biodegradation of organic contaminants and adding biochar can immobilize contaminants through sorption. The combined use of compost and biochar to reduce polycyclic aromatic hydrocarbon (PAH) and NSO-substituted PAH contamination has, however, not previously been systematically investigated. We have therefore investigated the processes involved (i) through sorption batch experiments, (ii) by monitoring changes in bacterial, fungal and archaeal communities using denaturing gradient gel electrophoresis, and (iii) through degradation experiments with fluorene, phenanthrene, pyrene, carbazole, dibenzothiophene, and dibenzofuran. Sorption coefficients for organic contaminants in soils increased tenfold following 10% compost addition and up to a hundredfold with further addition of 5% biochar. The rate of PAH and NSO-PAH degradation increased up to twofold following compost addition despite increased sorption, probably due to the introduction of additional microbial species into the autochthonous soil communities. In contrast, degradation of

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PAHs and NSO-PAHs in soil-compost-biochar mixtures slowed down up to tenfold due to the additional sorption, although some degradation still occurred. The combined use of biochar and compost may therefore provide a strategy for immobilizing PAHs and NSO-PAHs and facilitating degradation of remaining accessible contaminant fractions.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) consist of two or more fused aromatic benzene rings. They can derive from diffuse sources such as oil, coal, tar, or incomplete combustion (either natural or anthropogenic), and are therefore among the most abundant contaminants in soil and sediment [1,2]. Risk assessments for PAH contamination are typically limited to analysis of the 16 compounds defined by the US EPA, commonly referred to as the 16 US EPA PAHs. However, additional compounds, including NSO-substituted PAHs, may also be relevant to risk assessments because of their stability, mobility, and ecotoxicity [3]. For example, the toxicities of the structurally equivalent N-substituted carbazole, S-substituted dibenzothiophene (DBT), and O-substituted dibenzofuran (DBF) are similar to those of the unsubstituted fluorene to both insects and plants [4,5].

Highly contaminated hot spots in soils and sediments are commonly remediated by excavation or other laborious in-situ and ex-situ techniques. However, surrounding areas with low levels of diffuse contamination sometimes remain unremediated as their complete excavation, removal and treatment would be disproportionately expensive. Intermixing compost has previously been proposed as an alternative approach to the remediation of such low-contaminated sites that would accelerate the biodegradation of PAHs by introducing additional nutrients and PAH-degrading microbial communities [1]. Previous research has shown that selected bacteria, fungi, and archaea are able to degrade PAHs via hydroxylation or oxidation pathways [6–8]. Adding compost can increase both soil bioactivity and diversity in the microbial community, as well as potentially increasing the overall soil fertility, thereby contributing to the ecological restoration and stabilization of contaminated sites. A number of studies have shown that the addition of compost can accelerate degradation of the 16 US EPA PAHs in soils [9–11]. However, the degradation of NSO-substituted PAHs remains poorly investigated.

Biochar contains a graphene-like aromatic carbon skeleton produced during biomass pyrolysis; it has a high sorption potential for organic contaminants and is thus well suited for the immobilization of PAHs [12,13]. The use of biochar has been proposed for PAH immobilization in diffusely contaminated soils [14–16]. Biochar is also a soil amendment that counteracts the washing out of nutrients, increases the soil's water-holding capacity, raises the pH of acidic soils, and can alter a soil's microbial composition [17]. The mixing of biochar into diffusely contaminated soil can also contribute to the immobilization of heavy metals such as Zn and Cd that can inhibit PAH degradation [1,18].

Combining biochar (to immobilize organic and inorganic contaminants) with compost (to enhance the biodegradation of any remaining accessible organic contaminants) may therefore be a promising approach to use for the remediation of diffusely contaminated soils. Sites that are contaminated with heavy metals that may inhibit the degradation of organic contaminants [1,18] could be especially suitable for this approach. The effect that adding biochar to soil has on contaminant mobility and bioavailability has been compared with that of adding compost by Beesley et al. [19], who showed that both biochar and compost can reduce PAH, Zn and Cd concentrations in the pore water of contaminated soils, as well as phytotoxicity. This is in good agreement with Zeng et al. [20] who found that biochar and compost were both able to reduce the bioavailability and mobility of Cd, Cu, Zn and Pb in a contaminated soil. In addition, Marchal et al. [21] found that biochar reduced the desorbable and degradable phenanthrene fraction in soil. A reduction in microbial PAH degradation following biochar amendment can be expected. However, the effect that amendment with a combination of compost and biochar has on the degradation of PAHs and NSO-PAHs in contaminated soils remains poorly understood.

Our objective was therefore to reduce the gaps in our knowledge by investigating the effects of compost amendment, both with and without biochar on PAH and NSO-PAH degradation in soils. To improve our understanding of discrepancies in degradation behavior we also investigated (i) changes in microbial community structures with respect to bacteria, fungi and archaea, (ii) the sorption of PAHs and NSO-PAHs, and (iii) the effects that heavy metals (Zn and Cd) have on the degradation of PAHs and NSO-PAHs.

2. Materials and methods

2.1. Materials

To enable the systematic investigation of sorption and degradation, fresh soils were recovered from two agricultural field sites in Austria: sandy loam (SL) from Eschenau in Lower Austria and clayey loam (CL) from Kaindorf in Styria (see Table S-1 in the Supporting information). Compost was kindly provided by FK Agrar- und Umweltservice GmbH (Pixendorf, Austria). The materials were sieved to <2 mm in order to facilitate soil mixing and homogenization. To ensure the microbial activity of the fresh soils was not compromised, soils were stored in the dark at 4 °C for no more than two weeks prior to the degradation experiments. For the sorption experiments the materials were also dried overnight at 80 °C, crushed, and sieved to <250 μm to allow precise and representative sample weighing. A biochar produced from *Miscanthus* at a pyrolysis temperature of 550 °C was purchased from the UK Biochar Research Center (see Table S-2 in the supporting information). The biochar was selected from the UK Biochar Research Center Biochars based on its immobilization potential for PAHs, Cd and Zn established in pre-experiments (data not shown). Both sorption and degradation experiments were performed with unamended soils, soils amended with 10% compost, and soils amended with 10% compost and 5% biochar. The amendment rates were selected on the basis of results obtained from degradation and sorption pre-experiments (data not included), together with published information on the immobilization of heavy metals with biochar [22,23]. The characteristics of the soil mixtures were determined in a previous study [24]; they are shortly presented in Section 3.1 of this paper and Table S-1 in the Supporting information. Elemental compositions of the soil mixtures were also measured using an elemental analyzer (CHNS Elementar Vario MACRO). The pore size distribution for the biochar before and after incubation in the soil mixtures was measured using N_2 and CO_2 physisorption, following the protocols described in Sigmund et al. [25].