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Bioremediation of polychlorinated-*p*-dioxins/dibenzofurans contaminated soil using simulated compost-amended landfill reactors under hypoxic conditions

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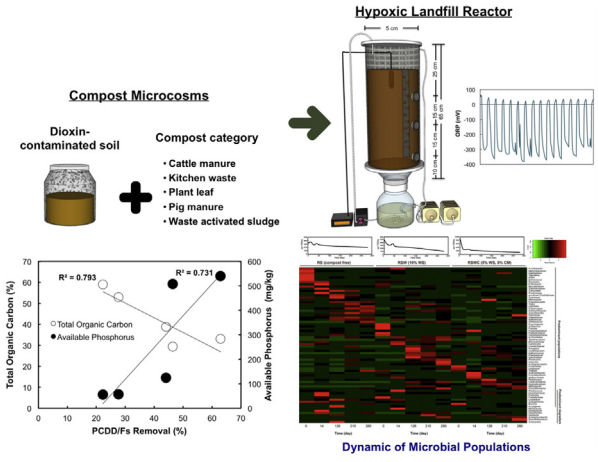
HIGHLIGHTS

- We developed a new hypoxic reactor system for remediating PCDD/Fs.
- We demonstrated effects of compost on the degradation of PCDD/Fs.
- We uncovered microbial compositions and dynamics during the degradation of PCDD/Fs.

GRAPHICAL ABSTRACT



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ABSTRACT

Compost-amended landfill reactors were developed to reduce polychlorinated-*p*-dioxins and dibenzofurans (PCDD/Fs) in contaminated soils. By periodically recirculating leachate and suppling oxygen, the online monitoring of the oxidation reduction potential confirmed that the reactors were maintained under hypoxic conditions, with redox levels constantly fluctuating between −400 and +80 mV. The subsequent reactor operation demonstrated that PCDD/F degradation in soil could be facilitated by amending compost originating from the cow manure and waste sludge and that the degradation might be affected by the availability of easily degradable substrates in the soil and compost. The pyrosequencing analysis of V4/V5 regions of bacterial 16S rRNA genes suggested that species richness of the soil microbial community was increased by a factor of 1.37–1.61. Although the bacterial community varied with the compost origin and changed markedly during reactor operation, it was dominated by *Alphaproteobacteria*, *Gammaproteobacteria*, *Actinobacteria*, and *Firmicutes*. The aerotolerant anaerobic *Sedimentibacter* and *Propionibacterium* spp., and the uncultured *Chloroflexi* group could be temporarily induced to a high abundance by amending the cow manure compost; the bacterial growths were associated with the rapid degradation of PCDD/Fs. Overall, the novel bioremediation method for PCDD/F-contaminated soils using hypoxic conditions was effective, simple, energy saving, and thus easily practicable.

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1. Introduction

Polychlorinated-*p*-dioxins and dibenzofurans (PCDD/Fs) are a group of toxic and persistent pollutants [1]. Over the recent decades, industrial activities involving the use of chlorinated pesticides, herbicides, and wood preservatives have led to the dispersion of large amounts of PCDD/Fs in the environment, with Agent Orange used in Vietnam War being the well-known examples [2–4]. Similarly, a pentachlorophenol (PCP) manufacturing plant at An-Shun of Tainan city in Taiwan was among the largest annual producers of PCP in East Asia during 1965–1979 [5]. After the plant closure, more than 40 ha of soil and sediment in An-Shun and its surroundings were contaminated with PCP and PCDD/Fs, the impurities formed during the manufacturing process [6,7]. As PCP was the precursor, the formed congeners were mostly octachlorinated, such as octachlorodibenzodioxin (OCDD), and octachlorodibenzofuran (OCDF) [8,9]. The levels of PCDD/Fs in An-Shun soil, analyzed as the international toxicity equivalent quotient (I-TEQ) was in the order of hundreds of mg I-TEQ/kg soil in the hot-spot area. The concentrations were extremely high compared with the regulation standard (1 μg I-TEQ/kg). Various physical, chemical, and biological methods have been developed to remove the PCDD/F toxicants [7]. Among these, the microbial degradation approach is considered ideal for field practice because of such advantages as environmental friendliness and cost effectiveness and because it can be implemented alone or as a complementary to other methods [10].

The bioremediation of PCDD/Fs requires diverse microbial degradation processes together with the activity of specialized microbial communities. Reductive dehalogenation and oxidative degradation, mediated by specific anaerobic and aerobic microbial populations, respectively, have been proposed as mechanisms involved in the transformation of PCDD/Fs [11,12]. So far, numerous bacterial genera under the phyla *Actinobacteria*, *Proteobacteria*, and *Firmicutes* with various ring-hydroxylating dioxygenases have been reported to degrade PCDD/Fs aerobically in pure cultures [12], whereas only members of the genus *Dehalococcoides* and their relatives under the phylum *Chloroflexi* were known to dechlorinate the PCDD/Fs under obligate anaerobic conditions [13]. Several bacterial groups, such as *Clostridium* and *Alcaligenes*, require a primary substrate for growth and cometabolism of PCDD/Fs and chlorinated aromatics [14,15]. Metabolic or cometabolic dechlorination of highly (octa-, hepta-) chlorinated DD/Fs is predicted to occur in the degradation pathway upstream, prior to the oxidative ring hydroxylation and mineralization in the tricarboxylic acid cycle; separately operated anaerobic dechlorination and aerobic decomposition have been proposed to facilitate these processes [16–18]. However, the rate of anaerobically reductive dechlorination is usually too low for site application [16,19]. Several recent studies have demonstrated that "degraders" grown at various redox levels coexist at the semiaerobic or semianaerobic conditions (herein referring to the oxygen-deficient (<2 mg/L) hypoxic conditions [20]) and that their collaborative interactions can rapidly degrade PCDD/Fs [21,22].

Sanitary landfills, which are bioreactors wherein various microbiological degradation processes can occur simultaneously, have been widely used for stabilizing organic solid waste with the soil matrix in full-scale applications. This approach typically confines the decomposable organic materials to an isolated area and minimizes the risks of exposure, allowing soil microorganisms to progressively convert the organic content into stable products. To optimize the landfill reactors, compost materials are added to improve soil matrix functions such as humidity, ventilation, and resistance to compaction, facilitating biostimulation and bioaugmentation by providing nutrients and microorganisms to replenish the soil microbial community [23]. The efficiency of xenobiotic biodegradation and carbon mineralization can be enhanced using a compost [24], and the efficacy of these processes depends on the type and origin of the compost [25]. Although landfills amended with compost are potentially a promising biotechnology [24,26], the feasibility and operational parameters of landfill bioremediation for PCDD/F$_S$, the effects of compost on landfill performance, and the succession of microbial communities have not yet been studied.

In this study, we developed a hypoxic landfill reactor system amended with a compost for remediating PCDD/F-contaminated soils. Consistent with our previous study, a hypoxic environment stimulated the growth of facultative anaerobic and aerobic microorganisms for the simultaneous dechlorination and oxidative decomposition of chlorinated congeners [22]. The hypoxic condition inside the landfill reactor was realized by maintaining a specific redox range through periodic leachate circulation and aeration. The effects of composts from various origins were first investigated. PCDD/F degradations in the landfill reactors with and without the selected compost were assessed. The abundance, diversity, and dynamics of the microbial community were characterized using PCR-based polyphasic molecular tools to clarify the changes in the microbial community structures associated with the operation of hypoxic landfill reactors using compost for the bioremediation of PCDD/F-contaminated soil.

2. Materials and methods

2.1. Contaminated soil

PCDD/F-contaminated soil was obtained from the site of an earlier PCP plant in An-Shun located in northwestern Tainan, Taiwan. The soil was collected from a depth of 10–30 cm and sieved through a 10–60-mesh (0.25–2 mm) sieve. Observable biological debris was removed. The soil sample was homogenized thoroughly and stored at ambient temperature. The soil used in this study was slightly alkaline (pH, 7.5–8.5), salty (chloride, 1.42 g/kg), and characterized by a sandy loam texture. Highly chlorinated PCDD/F congeners, namely octa- and hepta-chlorinated DD/Fs, contributed to >90% of the total toxicity equivalency [22].

2.2. Compost materials and microcosms

Composts of various origins—namely cattle manure (CM) and plant leaf (PL), pig manure (PM), kitchen waste (KW), and waste sludge (WS) from a pulp and paper wastewater treatment plant—were used in the microcosm experiments. All the composts were passed through a 20-mesh sieve. Chemical properties of the composts (pH_{water}, moisture, total organic carbon (TOC), total Kjeldahl nitrogen (TKN) and available phosphorus) were analyzed according to the methods reported preciously [27]. To prepare the microcosms, soil and compost (1:1, w/w) were thoroughly blended in a rotary shaker for 2 days, and 120 g of this mixture was introduced into 250-mL serum bottles. The water content in the microcosms was adjusted to 70% using Milli-Q water. Each treatment for the compost-amended soil and the soil without compost (control treatment) was prepared in duplicate. The serum bottles were capped with cotton stoppers, which allowed oxygen supplementation during dark incubation in an orbital shaker (200 rpm) at room temperature for 30 days.

2.3. Landfill bioreactor setup and operation

Three laboratory-scale column reactors were established for evaluating bioremediation. Fig. 1 shows the reactor configuration. The glass column was connected to a 2-L bottle underneath. For sampling, three ports were constructed at intervals of 15 cm. The