Addressing the restoration of degraded land in Europe using waste materials: a brownfield site in Wales

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Abstract

This paper describes experiments that demonstrate the effects and potential for remediation of a former steelworks site in Wales polluted with volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs). Under controlled conditions, vegetated soil columns of differing depths were exposed to solvents from beneath. Solvent vapour affected both shoot and root growth and soil microbial activity; effects varied with distance from the solvent source. In the field, compost was produced from organic wastes co-composted in-vessel with contaminated soil and the fate of PAHs followed over time. The reduction in PAH concentrations varied both with the presence of organic material during composting and with compost constituents after land spreading.

Keywords: compost; organic pollutants; dissipation; plant biomass; microbial activity.

1. INTRODUCTION

The restoration of historic, contaminated brownfield sites is an important part of the regeneration strategy for urban areas where undeveloped land is scarce. Orphaned sites are problematic because the polluter is often no longer legally responsible for their decontamination and it falls to government agencies to render these sites safe and to block source-receptor pathways. It is common practice to cover contaminated land with inert material as this is an inexpensive option, compared with remediation. In the case of volatile pollutants, however, vapour phase vertical movement through open structured soils may limit the effectiveness of this approach. VOCs readily partition to the gas phase and the contamination plume can migrate to the soil surface resulting in emissions to the atmosphere, creating the potential for human exposure [1]. The concomitant effects of VOCs migrating upwards on vegetation and the soil microbial biomass has received scant attention in the literature and yet it is important to quantify these effects in the context of best practice options for contaminated land.

Polycyclic aromatic hydrocarbons (PAHs) are common organic pollutants and occur naturally in fossil fuels e.g. coal and oil, or as a result of incomplete combustion of organic material. PAHs are classified as carcinogenic compounds. The concept of treating PAH-contaminated soil by co-composting or by mixing with finished compost has been well reviewed [2]. Composting soils contaminated with hazardous materials is still an emerging *ex situ* bio treatment [3]. The recycling of organic waste materials is becoming increasingly important not only because of emerging legislation to deal with Europe's ever-increasing production of wastes, but because it can also be viewed as a major tool for improving Europe's degraded soils. By adding value to wastes through composting, the treated product resulting from the composting process becomes a valuable resource for tackling land degradation and addresses many of the concerns highlighted in the EU Thematic Strategy for Soil Protection. The EU Life-Environment funded project TWIRLS – Treating Waste for Restoring Land Sustainability – has demonstrated safe in-vessel compost production using different organic and mineral waste streams. Recent work has shown that materials such as green waste, tertiary-treated biosolids and de-inking paper fibre can be mixed to produce composts that are both safe and fit for purpose [4, 5].

This paper reports on the potential for compost or composting to enhance the dissipation of pollutants from PAH-contaminated soil at a former steelworks site in Wales and examines the upward vertical flow of VOCs through soil and the effect this may have on plants and soil microbial activity.

2. MATERIALS AND METHODS

2.1 Dissipation of PAHs during composting

Parts of the former Shotton steelworks site (53°14' N, 3°2' W) were considered highly contaminated and hazardous prior to the decision to cover it with an inert sand cover taken from the nearby Deeside Estuary during the mid 1990s [6]. The cover was reported to be 4 m deep (anecdotal) but a recent rapid site assessment found it to be variable and less than 0.5 m in places. The experimental site was located in a zone where elevated levels of VOCs were found; the maximum total sum concentration of benzene, toluene, methylbenzene and xylenes (BTEX) was 500 $\mu g\ kg^{-1}$ and maximum total sum concentration of the US EPA priority 16PAHs was 34,000 $\mu g\ kg^{-1}$. Soil arising from the construction of a bentonite wall in year 2000 had been formed into static piles on the surface of the zone and provided us with a source of moderately contaminated soil to use in the composting trial.

Contaminated soil was either composted on its own or with organic wastes (Table 1). Compost was produced using EcoPOD® in-vessel aerobic composting vessels (Ag-Bag International Ltd, Warrenton, OR, USA) which are ideal for in situ work as all components of the system are mobile and suitable for up to 1000m^3 compost production on an 80-day cycle. Wastes were weighed and thoroughly mixed using a vertical auger cattle feed mixer wagon (Biga Twin Eco, Peecon, Etten-Leur, The Netherlands) and loaded into the EcoPOD® vessel (LDPE; 1.5 m diameter) by hydraulic ram, along with perforated plastic aeration pipe to deliver forced aeration. The aeration regime was controlled by means of a timed fan running at a flow rate of approximately 140 $dm^3\ min^{-1}$ for two months. Radio-linked temperature probes (Tinytag, Gemini Data Loggers UK Ltd., Chichester, Portsmouth) were inserted into each pod to log temperature data to an on-site computer.

Table 1. Feedstock composition of co-composting experiment.

Code	Composition	% by dry wt
CS	Contaminated soil	100
GW+BS	Greenwaste + Biosolids	80+20
PP+BS	Paper fibre + Biosolids	40+60
CS+GW+BS	Contaminated soil + Greenwaste + Biosolids	20+64+16
CS+PP+BS	Contaminated soil + Paper fibre + Biosolids	20+32+48
CS+GW+PP+BS	Contaminated soil + Greenwaste + Paper fibre + Biosolids	20+28+28+24

The EcoPOD® contents received forced aeration for 80 days then the immature compost underwent a maturation phase (no forced aeration) of a further four months *in situ*. At this point, composts were removed from the pods and spread immediately onto a designated area in a fully replicated, randomised split plot design. In addition to the six composts in Table 1, untreated contaminated soil was also spread and a control plot with no additions made up the eighth treatment. Treatments were spread to a depth of 7.5 cm over an approximate total area of 6000 m^2. Composts were then incorporated into the top 7.5 cm of *in situ* sand, to give 50:50 sand-compost mixes, to a depth of 15 cm, using a power harrow with 4 m spread.

Activities were carried out under Exemptions to Paragraphs 12 and 9 of the Waste Management License Act (1994) and in addition, planning consent was required.

Three levels of vegetation establishment treatment were superimposed randomly at the split plot level. Poplar trees were planted immediately after land spreading (Feb. 2006) followed by the seeding of mesotrophic grassland species (Apr. 2006). The third split plot was unseeded and allowed us to take account of natural plant regeneration.

PAHs were measured in contaminated soil taken from the static pile and prior to mixing with organic feedstocks (Jun. 2005), immediately after mixing and placement in the Ecopods (Jun. 2005), at pod opening after compost maturation (Jan. 2006), at land spreading with incorporation (Feb. 2006) and 9 months after land spreading (Nov. 2006).

PAHs in soil were analysed by Hewlett Packard 6890 Gas Chromatograph system using a Hewlett Packard 5973 Mass Selective Detector after soxhlet extraction (ALcontrol Geochem Analytical Services, Chester, U.K.). Analysis was conducted under MCERTS (European and International Standard BS EN ISO/IEC 17025:2000) accreditation.

2.2 Solvent vapour flow through soil

To simulate a highly contaminated subsoil environment we placed a reservoir of organic solvent (toluene, chloroform) or uncontaminated water under columns of a Eutric cambisol soil (previously unexposed to VOCs) of differing depths (10, 25, 50 and 75 cm) and monitored vapour flow through the soil over a 6 week period. Columns were packed to a bulk density of 1.2 g cm^{-3} and in triplicate. As a bioassay, five wheat pre-germinated seedlings were planted in each column which was then kept sufficiently moist throughout to ensure water did not limit plant growth. Columns were placed in a 20°C greenhouse with natural light during Sep. and Oct. 2006. Solvent reservoirs were checked daily and topped up as necessary. Plant root and shoot growth were measured as well as soil microbial activity at the end of the experiment.

3. RESULTS AND DISCUSSION

3.1 Dissipation of PAHs during composting

The thermophilic phase of composting attained the requisite temperature of 65°C (data not shown) for at least 7 days [7], even with the inclusion of 20% by dry weight mineral, contaminated soil. Contaminated soil that was composted alone reached a maximum temperature of 40°C.

All composted treatments resulted in some dissipation of USEPA 16PAH. Composted contaminated soil (CS) and CS+GW+BS exhibited the highest percentage removal of PAHs of 38% (Fig.1). The cumulative effect of composting and land spreading increased the dissipation of PAHs in composted CS and CS+PP+BS, with the greatest removal of 64% exhibited by composted CS. CS+PP+BS showed the greatest dissipation in absolute terms of 49,000 μg 16PAH kg^{-1} soil. Finally, there was little change in PAH concentration evidenced in uncomposted contaminated soil spread directly to land.

The addition of the organic mixes PP+BS and GW+PP+BS to contaminated soil appeared to reduce the dissipation of PAHs compared with contaminated soil alone or mixed with GW+BS (Fig. 1). This suggests paper fibre may have been responsible for the occlusion of PAHs. Once land spread, however, the CS+PP+BS exhibited continued dissipation of PAHs, unlike the other two organic mixes. We have previously found that paper fibre has greater carbon (C) availability than greenwaste [4] which results in a longer thermophilic phase during composting, leading us to speculate that only after available C is depleted do specialist microbial communities capable of mineralizing PAHs develop.

Where the PAHs in contaminated soil were not occluded by organic matter during composting, forced aeration appeared to be effective as a means of dissipation. We postulate that these losses were largely through volatilisation and that microbial mineralization will play an increasingly important role with time. In support of this, dissipation of PAHs has been found to be biphasic, a preliminary short period of rapid loss and a subsequent longer period of slower loss [8]. If so, organic amendments may be expected to directly influence mineralization through nutrient input and indirectly, through enhanced plant rhizosphere development. Indeed, Parrish et al. [9] found a positive relationship between plant biomass development and PAH biodegradation.

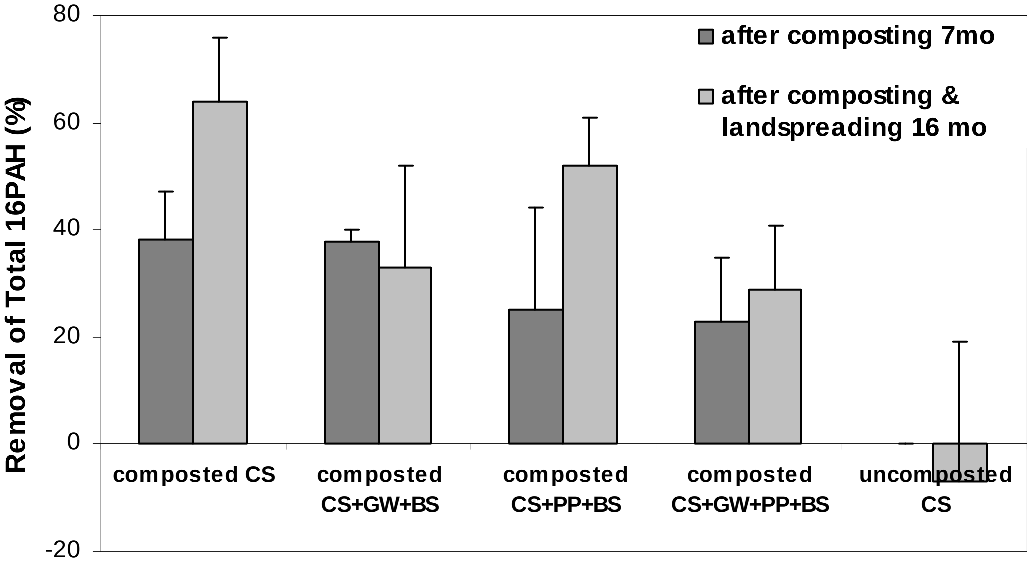

Figure 1. Effect of composting and of cumulative composting and land spreading on the percentage removal of Total 16PAH in contaminated soil (CS) undergoing different treatments. Values represent means ± SEM (*n* = 6). Abbreviations as in Table 1.

3.2 Solvent vapour flow through soil

There was a strong effect of solvent vapour on individual wheat plant shoot biomass (Fig. 2a), total shoot biomass and root length (data not shown) and microbial mineralization potential (Fig. 2b) in soils closest (≤25 cm) to the solvent source. Chloroform is known to disrupt biological function by lysing cell membranes and our data shows that it had a more pronounced effect on shoot biomass at the 10 cm distance than toluene. Solvent vapour flow tended to reduce shoot biomass for all core depths, though this was not statistically significant in the 25 and 50 cm cores. In the medium- to longer- term, acclimation by the soil microbial biomass would lead to contaminant biodegradation, particularly in the presence of plants [10]. Phytoremediation is growing in popularity as a clean-up technology and our results suggest that it is important to assess upward solvent vapour flow where this approach is planned. Although phytoremediation may not enhance the biodegradation of VOCs in the short term, the plants have been shown to play a role in retarding volatile contaminant transport to the atmosphere by reducing surface soil volatilisation [11].

4. CONCLUSIONS

Our work has shown that the simple act of covering land or groundwater contaminated with VOCs may not be sufficient to interrupt the source-receptor pathway and that vapour phase solvent flow can be harmful to both soil microbial and plant biomass in extreme situations. Plants are beneficial

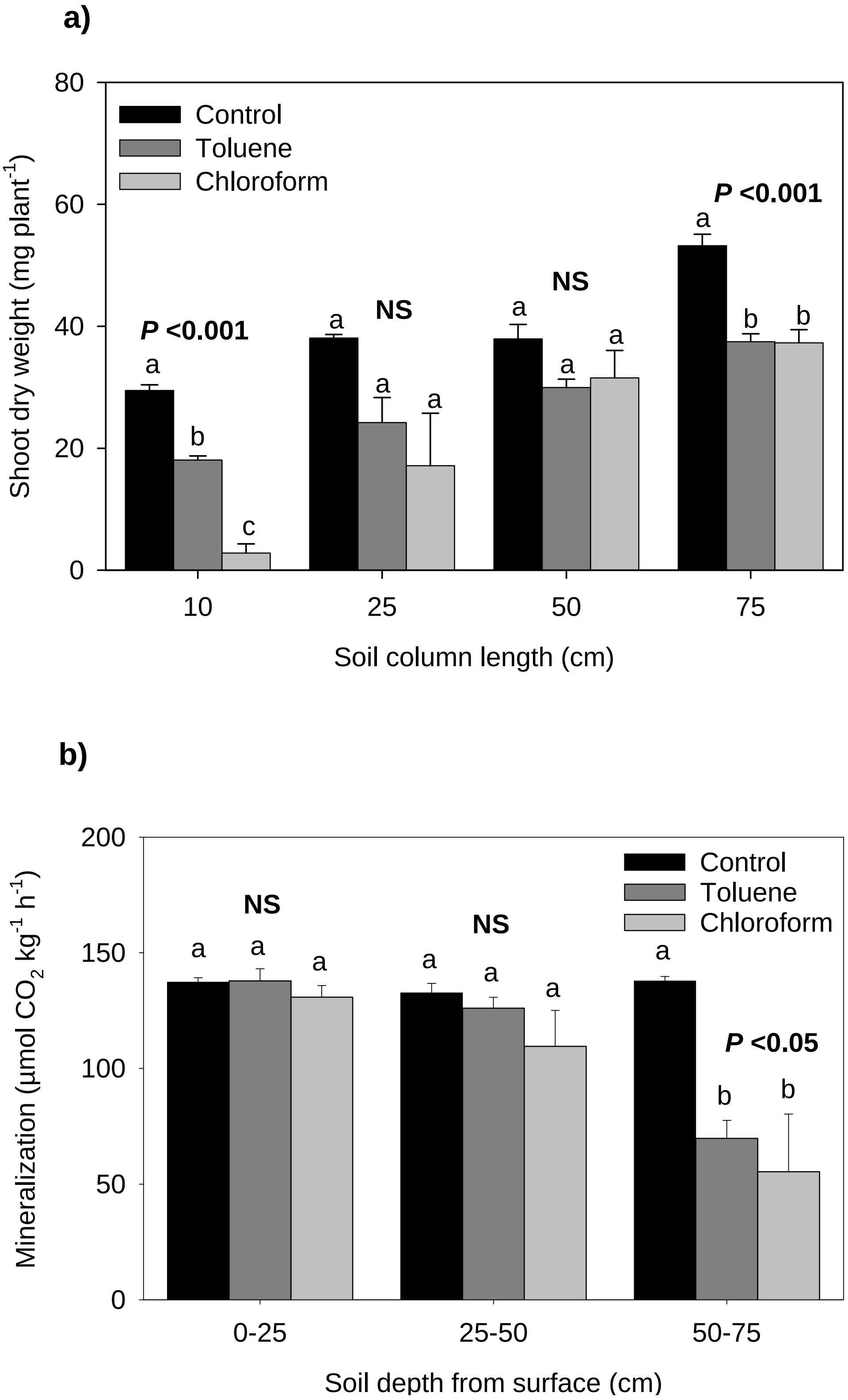

Figure 2. Effect of vapour phase solvent flow through soil on a) plant shoot mass in different soil column depths from the solvent source and b) microbial mineralization of ^{14}C-glucose in the top, middle and bottom 25 cm-sections of 75 cm deep soil cores. Values represent means ± SEM (n=3). Different letters indicate statistically significant differences between treatments and refer to within soil depth comparisons but not between depth comparisons.

to VOC dissipation (volatilization, sequestration and biodegradation) and hence it is important to monitor the effect of upward vapour phase movement on the soil ecosystem.

The dissipation of aged PAHs in soil, as opposed to PAH-spiked soil, remains a poorly understood and complex process. We have shown that on-site remediation can be partly achieved by subjecting contaminated soil to forced aeration and that the EcoPOD® in-vessel system is ideal for delivering this. The benefits after the land spreading of co-composted contaminated soil plus organic materials depended on the feedstocks used and further work will evaluate how the feedstocks influence microbial PAH-mineralization capability.

Acknowledgements

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