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TWIRLS – Treating Wastes for Restoring Land Sustainability: approaches to the remediation of contaminated land

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Abstract 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 no longer responsible and it falls to government agencies to render these sites safe and to block source-receptor pathways. Commonly, contaminated land is covered with inert material as this is an inexpensive option compared with remediation. In the case of volatile pollutants, however, vapour phase movement may limit its effectiveness.

This paper describes field-scale and greenhouse experiments that demonstrate the remediation of a former steelworks site in N. Wales polluted with volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs). In the greenhouse, soil columns were exposed to a range of solvents from beneath. Solvent vapour reduced shoot and root growth and microbial mineralization potential, with aromatic and chlorinated solvents showing greatest effect.

In the field, compost was produced from organic wastes co-composted with contaminated soil and the fate of PAHs followed over time. The reduction in PAH concentrations varied with the presence of organic material during composting and compost constituents after landspreading.

1 Introduction The main objective of the TWIRLS project is to demonstrate the effective re-use of industry and municipal wastes to produce soil-forming materials for restoring degraded land. Work tasks include the *in situ* remediation of polluted brownfield sites, re-creation of acid heathland on slate quarry waste, native pine forest establishment at an opencast schist quarry, soil organic matter improvement of degraded arable land and the restoration of colliery shale to pasture. Success has been achieved through the close working partnerships we developed with land owners and statutory authorities.

This paper reports on the potential for compost or composting to enhance the dissipation of pollutants from PAH-contaminated soil at the former Shotton steelworks site at Deeside, North Wales and examines the vertical flow of VOCs through soil and the effect this had on plants and soil microbial activity. 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 composting or with compost has been well reviewed by Semple et al. (2001).

Organic solvents are also common contaminants in soil and groundwater at post-industrial sites. Frequently the contamination is located below the rooting zone in soil possibly indicating that surface contamination may not be an issue.

2 Methods

2.1 Dissipation of PAHs during composting Parts of the Shotton site (OS SJ 305 728) was considered highly contaminated and hazardous prior to the decision to cap it with an inert sand cover taken from the Deeside Estuary during the mid 1990s (Smith Grant LLP, 2002). The cover was reported to be 4 m deep (anecdotal) but our rapid site assessment found it to be variable and less than 0.5 m in places. The TWIRLS demonstration trial was located in Zone 2 where we found elevated levels of VOCs (max. BTEX sum 500 µg kg^{-1}) and PAHs (max. 16PAH sum 34,000 µg kg^{-1}). Soil arisings from the construction of a bentonite wall in year 2000 had been formed into static biopiles on the surface of Zone 2 and provided us with a source of moderately contaminated soil to use in the composting trial.

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 trial.

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	Contaminatedsoil+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 four months in the Ecopods. At this point, composts were removed from the Ecopods 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 top 7.5 cm sand of Zone 2, 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 landspreading (2/06) followed by the seeding of mesotrophic grassland seeds (4/06). The third split plot was unseeded and would allow us to take account of natural plant regeneration.

PAHs were measured in contaminated soil taken from the biopiles and prior to mixing with organic feedstocks (6/05), immediately after mixing and placement in the Ecopods (6/05), at pod opening after compost maturation (1/06), at landspreading with incorporation (2/06) and 9 months after landspreading (11/06).

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). 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 (e.g. free-phase benzene at Shotton) we placed a reservoir of organic solvent (hexane, heptane, pentane, xylene, toluene, dichloroethane, chloroform, carbon tetrachloride) under soil columns and monitored vapour flow through the soil over a 14 day period. Simultaneously, we measured plant germination and growth and soil microbial activity.

3 Results

3.1 Dissipation of PAHs during composting The thermophilic phase of composting attained the requisite temperature of 65^{o}C (data not shown) for at least 7 days (British Standard for compost, PAS 100: 2005), even with the inclusion of 20% by dry weight mineral, contaminated soil. Contaminated soil that was composted alone reached an average max. of 40 ^{0}C.

All composted treatments resulted in some dissipation of Total PAH16. 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 landspreading 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 PAH16 concentration of 49,000 µg kg^{-1} soil. Finally, there was little change in PAH concentration evidenced in uncomposted contaminated soil spread directly to land.

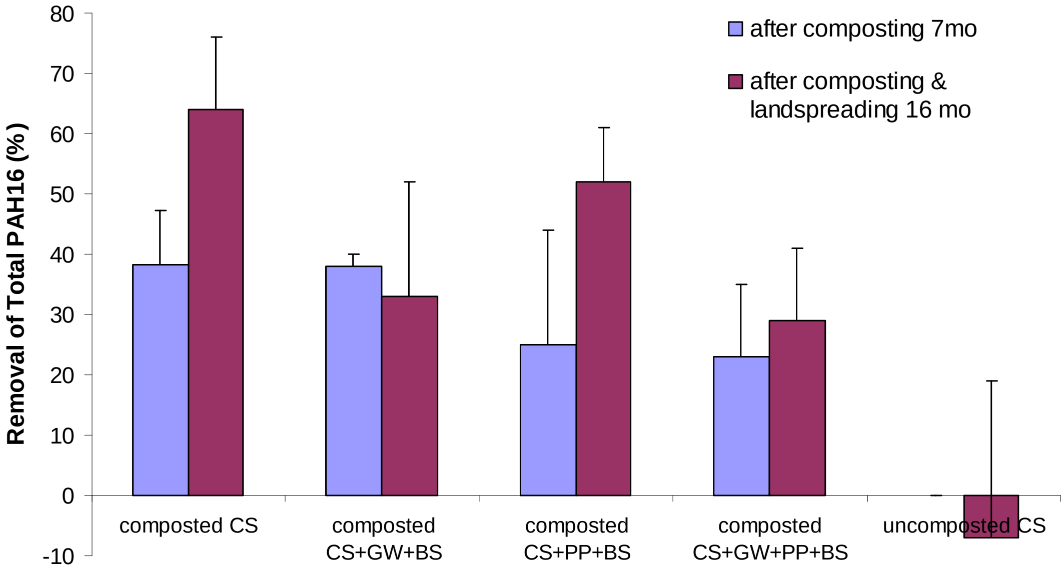

Figure 1. Effect of composting and of cumulative composting and landspreading on the percentage removal of Total PAH16 in contaminated soil (CS) undergoing different treatments. Values represent means ± SEM (n = 6). Abbreviations as in Table 1.

3.2 Solvent vapour flow through soil Solvent vapour phase flow through the soil significantly affected seed germination and plant growth in all treatments in comparison to the control (Fig. 2). Root growth tended to be more affected than shoot growth. Soil microbial activity was similarly affected. The toxicity of the solvents to both plants and microbes followed the series: heptane < hexane< pentane < xylene < toluene < dichloromethane < chloroform = carbon tetrachloride. Toxicity was inversely related to the octanol-water partition coefficient of the solvent ($r^2 = 0.90$) and its Henry's Law constant ($r^2 = 0.95$).

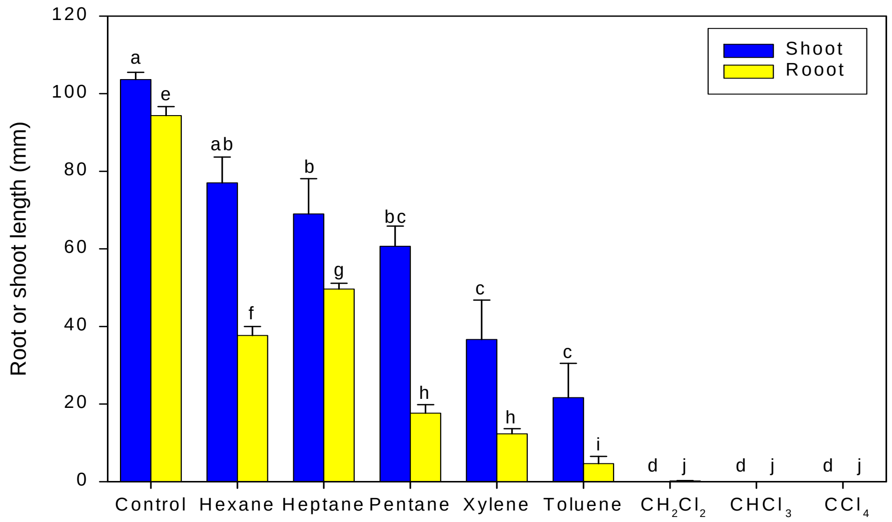

Figure 2. Effect of vapour phase solvent flow through soil on plant shoot and root length. Values represent means ± SEM (n = 3). Different letters indicate statistically significant differences between treatments at the $P < 0.05$ level.

4 Discussion and Conclusions

4.1 Dissipation of PAHs during composting 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 landspread, however, the CS+PP+BS exhibited continued dissipation of PAHs, unlike the other two organic mixes. We had previously found that paper fibre has greater carbon (C) availability than greenwaste (Williamson et al., 2006) which results in a longer thermophilic phase during composting. We hypothesise that only after available C is depleted do specialist microbial communities capable of mineralising 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. If so, organic amendments may be expected to directly influence mineralization through nutrient input and indirectly, through plant rhizosphere development. The next phase of work will be to demonstrate mineralization capability by enumerating phenanthrene (a prevalent PAH in this soil) degrader colony forming units using agar overlay plates and respirometry using ^{14}C-phenanthrene. In addition, a pot experiment is running concurrently which mirrors the landspreading treatments in the field study.

4.2 Solvent vapour flow through soil We hypothesise that organic solvent vapour flow through the soil inhibits plant growth by, (a) displacing oxygen from the soil so inducing hypoxia, and (b) by dissolving in the soil solution and disrupting biological membrane functioning.

In conclusion, 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. The dissipation of aged PAHs in soil, as opposed to PAH-spiked soil, remains a poorly understood 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 landspreading 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.

5. Acknowledgements

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6. References

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