Environmental Pollution 210 (2016) 182–191

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Organic amendments for risk mitigation of organochlorine pesticide residues in old orchard soils

Tiziana Centofanti [a, 1], Laura L. McConnell [b, *], Rufus L. Chaney [b], Nelson W. Beyer [c], Natasha A. Andrade [a], Cathleen J. Hapeman [b], Alba Torrents [a], Anh Nguyen [b], Marya O. Anderson [a], Jeffrey M. Novak [d], Dana Jackson [b]

[a] *Department of Civil and Environmental Engineering, University of Maryland College Park, 1173 Glenn L. Martin Hall, College Park, MD 20742, USA*
[b] *U.S. Department of Agriculture, Agricultural Research Service, Henry A. Wallace Beltsville Agricultural Research Center, 10300 Baltimore Ave., Beltsville, MD 20705, USA*
[c] *U.S. Geological Survey, Patuxent Wildlife Research Center, 10300 Baltimore Ave., Bldg. 308, BARC-East, Beltsville, MD 20705, USA*
[d] *United States Department of Agriculture, Agricultural Research Service, Coastal Plains Soil, Water, and Plant Research Center, 2611 West Lucas St., Florence, SC, USA*

ARTICLE INFO

Article history:
Received 27 August 2015
Received in revised form
21 November 2015
Accepted 23 November 2015
Available online xxx

Keywords:
Bioavailability
DDT
Dieldrin
Soil
Earthworms

ABSTRACT

Performance of compost and biochar amendments for *in situ* risk mitigation of aged DDT, DDE and dieldrin residues in an old orchard soil was examined. The change in bioavailability of pesticide residues to *Lumbricus terrestris* L. relative to the unamended control soil was assessed using 4-L soil microcosms with and without plant cover in a 48-day experiment. The use of aged dairy manure compost and biosolids compost was found to be effective, especially in the planted treatments, at lowering the bioavailability factor (BAF) by 18–39%; however, BAF results for DDT in the unplanted soil treatments were unaffected or increased. The pine chip biochar utilized in this experiment was ineffective at lower the BAF of pesticides in the soil. The US EPA Soil Screening Level approach was used with our measured values. Addition of 10% of the aged dairy manure compost reduced the average hazard quotient values to below 1.0 for DDT + DDE and dieldrin. Results indicate this sustainable approach is appropriate to minimize risks to wildlife in areas of marginal organochlorine pesticide contamination. Application of this remediation approach has potential for use internationally in areas where historical pesticide contamination of soils remains a threat to wildlife populations.

1. Introduction

In the United States and other countries, it is common to find former orchard sites with organochlorine pesticide (OCP) contamination. Historical applications of OCPs have left a continuing risk to local ecosystems as the contaminants persist in soils and present a distinct challenge for land managers and regulators. Current remediation methods for removing contamination vary from physical to chemical processes and have significant costs (Semple et al., 2001). Common remediation measures for contaminated sites include physical remediation, such as extraction, thermal desorption (FRTR, 2015), chemical remediation such as soil washing (Atteia et al., 2013; Sudharshan et al., 2012), and biological approaches such as bioremediation (Odukkathil and Vasudevan, 2013) and composting (Semple et al., 2001). Most physical and chemical techniques involve removing or processing a large amount of contaminated soil which is costly and time consuming. Removal costs have been estimated to range from approximately $40,000 to over $1million per acre (Hood, 2006; Gavrilescu, 2005). For large areas of marginal contamination, economical approaches are needed that can be used to effectively reduce risks while meeting the requirements of State and Federal regulations to protect human and ecological health.

Bioavailability and bioaccessibility as opposed to total concentration are critical factors to be considered when evaluating the

---

This paper has been recommended for acceptance by Jay Gan...

[*] Corresponding author. Present address: Bayer CropScience, 2 T.W. Alexander Dr., Research Triangle Park, NC 27709, USA.

E-mail addresses: tiziana.centofanti@gmail.com (T. Centofanti), laura.mcconnell@bayer.com (L.L. McConnell).

[1] Present address: Center for Irrigation Technology, California State University Fresno, Fresno, CA 93740, USA.

http://dx.doi.org/10.1016/j.envpol.2015.11.039
0269-7491/© 2015 Elsevier Ltd. All rights reserved.

risks posed by OCPs (Alexander, 2000; Reid et al., 2000; NRC, 2003; Semple et al., 2013). In the case of contaminated soils, human and ecological risks can be mitigated by reducing bioavailability. For example, aldrin and dieldrin uptake by cucumbers was reduced with the addition of activated charcoal to agricultural soils (Hilber et al., 2009; Saito et al., 2011). Activated carbon was also effective at reducing the bioavailability of polychlorinated biphenyls in soils to pumpkins and earthworms in laboratory and greenhouse studies (Paul and Ghosh, 2011; Langlois et al., 2011). Biochar amendments to soils have been shown to reduce uptake of chlorpyrifos and fipronil by Chinese chives (*Allium tuberosum*) (Yang et al., 2010), and addition of 5% compost to soils contaminated with chlordecone, a highly persistent organochlorine, reduced uptake in radish tubers and cucumbers by 50 and 60%, respectively (Clostre et al., 2014).

Regulatory agencies have incorporated the concept of bioavailability of OCPs and other hydrophobic contaminants when developing screening level regulations, such as the EPA Ecological Soil Screening Levels (Eco-SSL) (US EPA, 2007a,b) which can be used for an initial assessment of risks to wildlife from soil borne contaminants at a particular site. Therefore, by reducing the bioavailable portion of contaminants in soil, more total contaminant mass could potentially be left in place thereby reducing the need for more expensive remediation actions (Ehlers and Luthy, 2003; Sorell and McEvoy, 2013). A critical factor in reducing ecological risks from persistent organic pollutant contamination in soils is the mitigation of bioaccumulation of residues by soil invertebrates such as earthworms and the consequent biomagnification in the terrestrial food chain (Armitage and Gobas, 2007).

Previous studies have shown that growing plants on test soils during earthworm exposure to DDT and dieldrin may significantly reduce bioaccumulation (Kelsey and White, 2005; Peters et al., 2007). These studies are limited and nearly all used *Eisenia foetida* in their testing, but consistent in showing that vegetation on a soil reduces earthworm accumulation of these compounds, so the concept seems likely to be a general protection of soil fauna by the presence of growing roots of a crop or stronger sorption in the amended soils. In the case of earthworms, a possible mechanism for this effect in agricultural soils is that earthworms feed on dead roots that contain reduced levels of contaminants.

The performance of compost and biochar amendments for *in situ* risk mitigation of aged DDT and dieldrin residues in old orchard soils was examined in a soil microcosm approach. Four organic amendments of differing total organic carbon content and quality were added to the orchard soil. The change in bioavailability of the pesticide residues was assessed using earthworms, *Lumbricus terrestris* L., in 4-L soil microcosms with the addition of plant cover (orchard grass or rye grass). At the end of the study, the bioaccumulation factor (BAF) of each treatment was determined for 1,1,1-trichloro-2,2-di(4-chlorophenyl)ethane (4,4′-DDT), 1,1-dichloro-2,2-di(4-chlorophenyl) ethylene (4,4′-DDE), and dieldrin. In addition, hazard quotient (HQ) values were determined for the American woodcock and the northern short-tailed shrew based on results of the microcosm experiments using toxicity reference values provided in the EPA Eco-SSL guidance (US EPA, 2007a, 2007b).

To our knowledge this is the first experiment reported in the literature that combines addition of organic amendments and growing plants to determine the reduction in bioaccumulation of DDT, DDE, and dieldrin to earthworms.

This study presents new information on the effects of plant cover in combination with organic amendments on the bioavailability of pesticide residues in soil. It also provides a unique opportunity to investigate the fate and availability of highly aged pesticide residues in agricultural soils and the interaction between these residues and different types of organic matter and agricultural byproducts.

Results of this work will not only provide critical information needed for the remediation of the particular old orchard site under study, but it will also provide valuable information to natural resource and regulatory managers seeking lower cost remediation solutions to mitigate ecological risks using sustainable materials such as compost.

2. Materials and methods

2.1. Description of orchard field site

The soil utilized in this study was collected from a former orchard located at the US Department of Agriculture, Henry A. Wallace Beltsville Agricultural Research Center in Beltsville, Maryland, USA. The site received continuous DDT applications starting sometime in the 1940s until 1972 when DDT was banned (US EPA, 2015). While specific records of dieldrin use on this field do not exist, the presence of dieldrin in the soil indicates it was likely used from the 1950s until it was banned for most uses in 1987 (US EPA, 1998). This Area of Concern (AOC) BARC 19, henceforth also referred to as the “site”, was first identified as a debris filled, erosional gully adjacent to open fields during an Environmental Protection Agency (US EPA) Preliminary Assessment/Site Inspection in 1991, as part of a larger effort to identify Federal facilities with significant environmental impacts that might require them to be listed on the National Priorities List and addressed under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA), commonly referred to as Superfund (http://www.epa.gov/superfund/policy/cercla.htm).

In 2007, an initial bioavailability study was conducted in 40 ha of the former orchard site including the collection of soil, earthworms, and small mammals followed by a risk assessment using the screening level approach described in the EPA Eco-SSL guidance for DDT and dieldrin (US EPA, 2007a,b; USDA, 2007, 2009). An area of 10 ha was determined to have contaminated soils elevated levels above the ecological risk according to the EPA Eco-SSL guidance for DDT and dieldrin. An area of 0.55 ha of highest contamination was excavated and removed from the site, but approximately over 9 ha of less-contaminated vegetated area remain. Details of all site investigations and removal actions at BARC 19 are available at the Beltsville Agricultural Research Center Information Repository (USDA-ARS, 2015).

2.2. Soil collection and organic carbon amendments

A surface soil sample (15-cm depth) of approximately 2 m^2 was collected in March 2011 from the BARC 19 Area of Concern at the USDA-ARS Beltsville Agricultural Research Center, Beltsville, Maryland, USA. The site was selected based on earlier soil survey results (USDA, 2007) and preliminary testing carried out 2–3 months prior to sample collection. The soil was a gradation between Sassafras and Croom sandy loams with an organic carbon (OC) content of 2.56 ± 0.08% ($n = 14$). The USDA taxonomic classification for the Sassafras series is– Fine-Loamy, siliceous, semiactive, mesic Typic Hapludult; and for the Croom series is—Loamy-Skeletal, siliceous, semiactive, mesic Typic Hapludult.

The bulk soil sample was sieved (4 mm) and well mixed and split into sub-samples for mixing with amendments. The soil collected from the field site was homogenized using cross mixing on a clean plastic tarp (Beyer et al., 2013a). This method consisted of spreading out the soil on a plastic tarp and mixing it by pulling half the tarp over the other half and vice versa for two times each side. Between each mixing the soil is spread out again and the soil