



Study of processes influencing bioavailability of pesticides in wood-soil systems: Effect of different factors



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ARTICLE INFO

Keywords:

Oak
Pine
Wood
Soils
Sorption
Desorption
Bioavailability
Alachlor
Linuron
Metalaxyl

ABSTRACT

Lignocellulosic wastes and by-products containing lignin are now available in large amounts from forestry and industrial activities, and could be promising organic materials for the biosorption of pesticides by soils in order to reduce point-source pollution. Adding these materials to soil requires understanding the process of pesticide sorption-desorption by wood-soils, as sorption capacity could increase, with changes in pesticide bioavailability and final fate. The objective of this work was to study the effect that pine and oak wood added to soils had on the sorption/desorption of the pesticides linuron, alachlor, and metalaxyl. Experiments were conducted with two sandy loam and sandy clay soils each amended with two wood doses (5% and 50%) after different incubation times (0, 5 and 12 months). A low wood dose (5%) had no significant impact on the sorption (K_f) of alachlor, but K_f increased for linuron (up to 5.4–1.7 times) and metalaxyl (up to 4.4 and 8.6 times) in all wood-soil systems. The results were not significantly different after different incubation times. The desorption results indicated that wood decreases the sorption irreversibility of alachlor, and increases that of linuron and metalaxyl, with a varying effect of the wood-soil incubation time. The addition of a high wood dose to soil (50%) was more significant for increasing the sorption of all the pesticides, and the sorbed amounts remaining after desorption (> 49% for linuron, > 33% for alachlor and > 6% for metalaxyl), although there was no apparent discrimination between the two types of woods. The role of the nature of the organic carbon (K_{oc} values) for sorption was evidenced for alachlor and metalaxyl, but not for linuron. These outcomes are of interest for extending wood application to soil as a barrier for avoiding environmental risk by point-source pollution due to the use and management of pesticides in farming systems.

1. Introduction

Pesticides are used in modern farming to increase food production. However, surface and ground water contamination has been detected in recent years, possibly due to inadequate pesticide management (Herrero-Hernández et al., 2013; Papadakis et al., 2015; Ccanccapa et al., 2016). Point-source pollution caused by accidental spillages when filling a tank or cleaning spraying equipment has been identified as a major contamination risk (Castillo et al., 2008; Karanasios et al., 2012). To reduce this pollution, the use of natural organic materials or modified inorganic materials as barriers for enhancing the retention of pesticides by soils has been attracting growing attention for some years now (Rodríguez-Cruz et al., 2007a; Marín-Benito et al., 2012a; Tran

et al., 2015; Álvarez-Martín et al., 2016a; Centofanti et al., 2016), because organic matter (OM) plays an important role in the sorption of these compounds. These techniques enhance the initial immobilization of pollutants in the barrier, and their subsequent chemical, physical and biological transformation (De Wilde et al., 2009).

The biodegradation of pesticides after the addition of organic materials to soil may occur because the application of an additional source of OM, and sometimes microorganisms, accelerates the degradation of pesticides (Kravariti et al., 2010; García-Delgado et al., 2015; Pinto et al., 2016). Most of these organic materials or biomasses can in fact allow the development of fungi producing extracellular ligninolytic enzymes that promote pesticide degradation (Castillo and Torstenson, 2007), preventing their transport to surface and ground waters

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(De Wilde et al., 2009). However, by decreasing the bioavailability of pesticides owing to their increased sorption capacity, the addition of an organic residue to soil may also decrease pesticide degradation (Briceño et al., 2007; Kravariti et al., 2010; Álvarez-Martín et al., 2016b). Accordingly, there is increased interest in evaluating the sorption-desorption capacity of pesticides by the biomixtures of soil-organic residue as the principal component in the barrier system, which is the main element controlling immobilization and degradation efficacy, and therefore pesticide dissipation (Karanasios et al., 2012).

One type of highly promising organic materials for biosorption are lignocellulosic wastes and by-products containing lignin, available in large amounts from forestry and industrial activities (Karanasios et al., 2010, 2012; Pintor et al., 2012; Abdolali et al., 2014; Muter et al., 2014). The literature has covered the use of bark, cork and woodchip residues as biomaterials or low-cost sorbents for point-source pollution barriers, or for creating sorbent zones in the soil to increase the sorption of organic pollutants and delay their mobility. There are studies on hydrophobic compounds such as toluene, benzene and o-xylene (MacKay and Gschwend, 2000), polynuclear aromatic hydrocarbons (Boving and Zhang, 2004), persistent organic pollutants (Olivella et al., 2011, 2013) and pesticides (Brás et al., 1999; Domingues et al., 2007; Rodríguez-Cruz et al., 2007b; Boudesocque et al., 2008; Esra Ilhan et al., 2012; Olivella et al., 2015; Mandal and Singh, 2016).

Besides lignin, wood contains cellulose and hemicellulose as its main components. Rodríguez-Cruz et al. (2009) have reported that lignin, the hydrophobic component of wood, has a high sorption capacity towards hydrophobic organic compounds and/or pesticides, which can be retained by different interaction mechanisms. The authors studied the sorption-desorption and interaction mechanisms of three non-ionic pesticides (linuron, alachlor and metalaxyl) and two ionic ones (paraquat and dicamba) by three commercial lignins (hydrophobic macromolecule) and cellulose (hydrophilic macromolecule) as wood components. The use of wood residues as a soil amendment and their potential to immobilize pesticides, and hence their potential to be used in technologies for preventing soil and water pollution, has not been widely reported. Some studies provide information on the properties of different woods and pesticides involved in the immobilization process (Rodríguez-Cruz et al., 2007b) or the effect of fixed wood barriers on the retention of pesticides under saturated flow conditions (Rodríguez-Cruz et al., 2011). However, a further scale-up of such studies requires biomixing these biomasses with soils of different properties at both bench and pilot scales. Gathering information about this performance involves investigating the sorption capacity of pesticides by biomass-soil mixtures and their potential for desorption as an indicator of the bioavailability of the sorbed compounds. These processes depend on the characteristics of wood-soil systems and on the properties of pesticides (Chaplain et al., 2011).

Linuron, alachlor and metalaxyl are compounds from urea, chloroacetanilide and alaninate that constitute some of the most important groups of pesticides used in agriculture. They have very different properties in terms of water solubility, hydrophobicity and GUS mobility index (PPDB, 2016). These compounds could be considered models for (i) studying the effect different factors have on sorption-desorption by wood-soil systems, and (ii) extending the information to other pesticides widely used in farming, involving biobed systems supported with wood residues.

The objective of this work was to study the effect that two woods, pine (softwood) and oak (hardwood), added as sawdust to sandy loam and sandy clay soils, had on the immobilization of three pesticides with different characteristics: linuron, alachlor, and metalaxyl. Assays were conducted on the sorption-desorption of such compounds in the two soils amended with two doses of wood and in soil-wood systems after different incubation times (0, 5 and 12 months) in order to i) evaluate such processes under more extended conditions, and ii) better simulate real field scenarios for the retention of pesticides from point sources of pollution.

2. Materials and methods

2.1. Soil and wood samples

Samples of two soils with different properties were collected from the surface layer (0–15 cm depth) of an agricultural field located in Aldarrubia (AL-soil) (41° 0' 29.16" N, 5° 29' 51" W) and in Castellanos de Moriscos (CM-soil) (41° 01' 10.92" N, 5° 35' 33" W), both in the province of Salamanca (Spain). The soils were left overnight at room temperature to reduce moisture content, and then sieved (< 2 mm). The pH was determined in a soil/water suspension (1/2.5 w/v), and particle size distribution was determined using the pipette method. Organic carbon (OC) content was determined by oxidation (Walkley-Black method). AL-soil was a Fluventic Eutrochrepts soil (Soil Survey Staff, 2010) of a sandy loam texture (11.8% clay, 13.6% silt, and 74.5% sand), with 0.51% OC content, a pH of 6.3, and a cation exchange capacity of 4.8 cmol kg⁻¹, whereas CM-soil was a Typic Rhodolxeralfs soil of a sandy clay texture (38.1% clay, 5.8% silt, and 56.1% sand), with 1.04% OC content, a pH of 6.9 and a cation exchange capacity of 20.5 cmol kg⁻¹ (Bremner, 1996). The clay minerals identified by X-ray diffraction indicated the presence of illite and kaolinite (AL-soil), and of montmorillonite, illite and kaolinite (CM-soil). The water holding capacities were 34% and 44% for AL-soil and CM-soil, respectively.

Pine and oak sawdust (< 1 mm) was obtained from a local industry in Salamanca (Spain). The woods' main physicochemical properties have been determined in a previous work (Rodríguez-Cruz et al., 2007b). They have a similar total OC content, 38.5% (oak) – 41.6% (pine), but different soluble organic carbon (DOC), 6.86% for oak and 1.62% for pine, and different lignin contents: 18.2% for oak and 24.4% for pine.

2.2. Chemicals

Alachlor (2-chloro-2',6'-diethyl-N-(methoxymethyl) acetanilide) was supplied by Chem Service (West Chester, USA) (99.5% purity). It is a solid herbicide with a water solubility of 240 µg mL⁻¹ at 20 °C. Linuron (3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea) was supplied by Riëdel de Haën (Hannover, Germany) (> 99% purity). It is a solid herbicide with water solubility of 63.8 µg mL⁻¹ at 20 °C. Metalaxyl (methyl N-(methoxyacetyl)-N-(2,6-xylyl)-DL-alaninate) was supplied by Riëdel de Haën (Hannover, Germany) (> 98% purity). It is a solid fungicide with a water solubility of 8400 µg mL⁻¹ at 22 °C. The log K_{ow} values for these compounds at pH 7 and 20 °C were 3.09 for alachlor, 3.00 for linuron, and 1.75 for metalaxyl. The GUS index values (1.08 for alachlor, 2.34 for linuron, and 2.79 for metalaxyl) (Table S1 in the Supplementary Material, PPDB, 2016) indicate the low or moderate leaching potential of these compounds for moving toward groundwater, although literature data support their frequent presence in it (Herrero-Hernández et al., 2013). The ¹⁴C-labelled pesticides with a specific activity of 2.31 MBq mg⁻¹ and 97.0% purity (alachlor), 4.13 MBq mg⁻¹ and 98% purity (linuron), and 1.37 MBq mg⁻¹ and 97.2% purity (metalaxyl) were supplied by International Isotopes (Munich, Germany).

2.3. Treatment of soils with oak and pine wood

A bulk sample of 2 kg dry weight (dw) of soil was placed in a glass laboratory biobed (40 × 30 × 15 cm), and amended with oak and pine sawdust (< 1 mm) at a dose of 5% w/w, equal to 40 t C ha⁻¹, similar to other organic residues (Rodríguez-Cruz et al., 2012), and ten times higher at 50% w/w, equal to 400 t C ha⁻¹, to examine the effect of the wood dose and its nature. The soils' initial moisture content was adjusted to 40% of their maximum water-holding capacity. A fraction of unamended AL-soil and CM-soil and both soils amended with the low dose (5%) of wood was incubated in a thermostated chamber at 20 °C and 40% of water-holding capacity for 0, 5 and 12 months. Sub-samples

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