



Effect of surfactant application practices on the vertical transport potential of hydrophobic pesticides in agrosystems

Jeanne Dollinger ^{a,*}, Veronika J. Schacht ^{b,c}, Caroline Gaus ^b, Sharon Grant ^b

^a UMR ECOSYS, INRA-AgroParisTech, Campus de Grignon, F-78850 Thiverval-Grignon, France

^b QAEHS, The University of Queensland, 39 Kessels Road, Coopers Plains QLD 4108, Australia

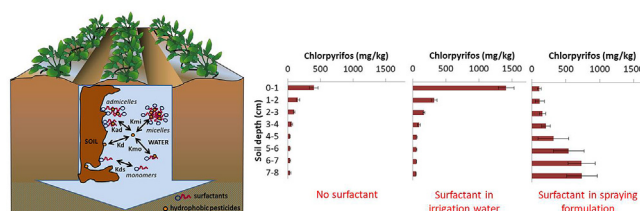
^c Recetox, Masaryk University, Kamenice 753/5, 625 00 Brno, Czech Republic



HIGHLIGHTS

- Surfactants modify hydrophobic pesticide mobility in soils.
- We designed a methodology to study surfactant facilitated transport of pesticides.
- Single irrigation event with waste water containing TritonX100 reduced chlorpyrifos leaching.
- TritonX100 in spraying formulation increase chlorpyrifos mobility in soil.

GRAPHICAL ABSTRACT



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ABSTRACT

Surfactants have the potential to modify the environmental behavior of hydrophobic pesticides leading to an enhanced or reduced mobility risk. This risk is often overlooked in registration procedures due to a lack of suitable methodologies to quantify the transport potential of pesticides with surfactants. In this study we present a novel methodology designed to study the surfactant facilitated transport of pesticides under controlled equilibrium and dynamic hydrologic conditions. Using this methodology, we investigated the risk of chlorpyrifos enhanced mobility for two common surfactant application practices in agrosystems: pesticide spraying and irrigation with waste water. With the dynamic experiments we showed that a single irrigation event with artificial reclaimed water containing the nonionic surfactant Triton X100 at a concentration of 15 mg/L reduced the leaching of chlorpyrifos by 20% while the presence of the same surfactant in the chlorpyrifos spraying formulation reduced the leaching amount by 60%. However, in the first case 90% of the chlorpyrifos fraction remaining in soil was retained in the upper 3 cm while in the second case, 72% was transported to the bottom layers. The presence of Triton X100 in irrigation water or spraying formulation retards the leaching of chlorpyrifos but enhances its downward transport.

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1. Introduction

Hydrophobic pesticides are widely considered immobile in soil due to their very low water solubility and high affinity for organic

carbon (Arias-Estévez et al., 2008). However, a significant number of hydrophobic pesticides have been detected worldwide in groundwater and remote ecosystems (Arias-Estévez et al., 2008; Elliott et al., 2000; Gebremariam et al., 2012). These findings of enhanced unintentional mobility in soil may be explained by preferential water flow or transport with soil colloids (Arias-Estévez et al., 2008; Elliott et al., 2000; Persson et al., 2008). Furthermore, pesticide transport in association with surfactants, i.e.

* Corresponding author.

E-mail address: jeanne.dollinger@inra.fr (J. Dollinger).

surfactant facilitated transport, has also been suggested to contribute to the leaching and off-site transport behavior of hydrophobic pesticides (Lee et al., 2000; Müller et al., 2007).

Surfactant monomers are composed of a hydrophobic tail and a hydrophilic head (Ying, 2006). Above a specific concentration, the critical micelle concentration (CMC), the monomers aggregate into micelles. These water soluble surfactant monomers and micelles contain hydrophobic micro-environments providing an efficient partitioning site for hydrophobic organic compounds increasing their apparent water solubility (Krogh et al., 2003; Ying, 2006; Zhang et al., 2009). Despite the recognition of this solubility enhancement process as a potentially significant unintentional transport pathway for hydrophobic pesticides, and the ubiquitous presence of surfactants in agrosystems, the risk associated with surfactant use has been overlooked to date.

There are two major sources of surfactants in agrosystems. Firstly, surfactants (predominantly alkylphenol ethoxylates (APEO)) are added to commercial pesticide formulations at up to 20% (w/v) or are used as spray adjuvants to facilitate pesticide amendments (Krogh et al., 2003). Secondly, the common agricultural practice of waste water irrigation may also contribute to a substantial amount of surfactants in agrosystems. Surfactant concentrations in reclaimed water have been reported to reach up to 54 mg/L (Fernández-Gálvez and Mingorance, 2010; Krogh et al., 2003) made up of different types of surfactants differing by the composition of the hydrophobic tail and the ionization of the hydrophilic head. For nonionic surfactants like APEO, the most commonly used surfactant type for agricultural, industrial and household applications, the concentration in reclaimed water was reported to vary between 0.75 and 11.3 ± 3.9 mg/L (Fernández-Gálvez and Mingorance, 2010; Hernández Leal et al., 2011).

Although potentially significant, the off-site transport risk for pesticides with surfactants is not assessed during pesticide registration procedures. This may be due to the difficulty of studying this specific pathway in-situ or in the lab because of the simultaneous action of other preferential transfer pathways such as colloidal transport and preferential flow. Few methodologies are available to assess the role of surfactants on the mobility of hydrophobic compounds. Existing methods are mostly focused on surfactants' use during soil washing for decontamination purposes, for which excessive amounts of surfactants are intentionally applied. Moreover, these studies are mainly based on equilibrium batch methods. Other studies investigate the effect of waste water irrigation on the mobility of organic pollutants in the field or in the lab under both dynamic and equilibrium conditions (Chefetz et al., 2008; Peña et al., 2011). However, the experimental designs implemented in those studies don't enable the separate assessment of the different preferential pathways. Thus, methods adapted to assess the risk of unintentional surfactant facilitated pesticide transport in both equilibrium and dynamic conditions are lacking to date.

The aims of this study were thus i) to adapt batch and column technics to assess the risk of unintentional surfactant facilitated transfer of hydrophobic pesticides in equilibrium and dynamic conditions and ii) investigate the risk of surfactant facilitated pesticide transfer induced by two common agricultural practices, pesticide spraying and irrigation with reclaimed water. We used chlorpyrifos (CPF) and Triton X100 (TX100) as model compounds. CPF is one of the most widely used organophosphorus pesticides being the main active ingredient of about 400 commercial mixtures. An estimated 25 million kilograms of active ingredient are sprayed globally every year for pest control on a large variety of crops (Gebremariam et al., 2012). Typically, this hydrophobic insecticide is assumed immobile in soils based on its physico-chemical properties (EPHC, 2003; Gebremariam et al., 2012). However, CPF has been detected worldwide in groundwater,

drinking water wells and remote ecosystems (Gebremariam et al., 2012). APEOs such as TX100 are very likely to be associated with hydrophobic pesticides in soils as this surfactant type in particular is a major component of commercial pesticide formulations. Furthermore, TX100 is recommended as a spray adjuvant for a variety of insecticides, including CPF (Krogh et al., 2003; O'Connell, 2011). An artificial soil matrix was designed to eliminate other pesticide transport processes and enable assessment of the surfactant facilitated transfer risk. For this, constituents of soils playing a key role in hydrophobic pesticide and surfactant mobility/retention i.e. clay minerals and humic acid, were immobilized onto silica sand and further mixed in a proportion representative of soils in typical Australian agrosystems and other Vertisols around the world.

2. Material and methods

2.1. Model compounds & chemicals

CPF, the selected model pesticide for this study was purchased from AccuStandard (New Haven, USA). The model APEO, TX100, was purchased from Sigma Aldrich (Sydney, Australia). The physico-chemical properties of both model compounds are listed in Table 1.

For the preparation of the artificial soil matrix, silica sand (50–70 mesh), humic acid, N-(3-dimethylaminopropyl)-N'-ethylcarbodiimide hydrochloride (EDC) and Polyvinyl alcohol (PVA) were purchased from Sigma Aldrich. Two clays, montmorillonite and illite, were obtained from the Source Clay Minerals Repository (University of Missouri). Pure deionized water was prepared using a Millipore water purification system (Merck Millipore, Kilsyth, Australia). The solvents were of analytical grade; toluene (99.99% purity), methanol (98% purity) and dichloromethane (99.8% purity) and purchased from Merck.

2.2. Soil matrix design and validation

The OC and clay content in the artificial soil matrix were represented by humic acid, montmorillonite and illite. The importance of these materials in sorption processes of hydrophobic pesticides and surfactants, in particular CPF and TX100, onto soil has been demonstrated extensively (Amirianshoja et al., 2013; Gebremariam et al., 2012; Laha et al., 2009; Rodríguez-Cruz et al., 2005; Valverde García et al., 1992; Zhu et al., 2003). Humic acids are often used to represent soil organic matter in sorption studies of hydrophobic compounds (Joo et al., 2012; Tanaka et al., 2005; Wan et al., 2011; Zhang et al., 2009). Together with fulvic acids, humic acids make up the predominant fractions of soil organic matter. Their distinction is operational rather than structural. Although sharing many characteristics, humic acids are less polar than fulvic acids and thus assumed to drive the sorption processes of hydrophobic compounds (Wang and Keller, 2009). Humic acid also seems to represent fairly well soil organic matter for TX100 sorption assessment as the partition coefficients of TX100 to soil organic matter and humic acid are reported to be similar (Zhang et al., 2009). Montmorillonite and illite were selected as they are the most representative of the clay minerals constituting Queensland's Vertisols (Chinn and Pillai, 2008).

The coating of silica sand with humic acid was performed with the EDC incubation method followed by end-capping of the free amine groups as described by Koopal et al. (1998). Silica sand was also coated with montmorillonite or illite. PVA was used to bind the clay minerals to silica sand following the optimized method described by Jerez et al. (2006). The efficiency and homogeneity of the coatings were checked with scanning electron microscope (see

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