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Fate of pharmaceuticals and pesticides in fly larvae composting

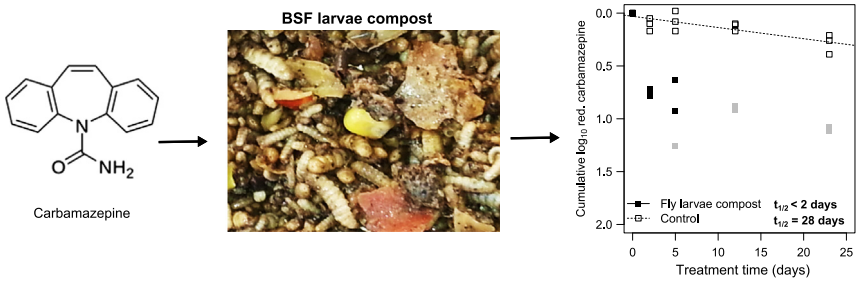
C. Lalander [a,*], J. Senecal [a], M. Gros Calvo [a], L. Ahrens [b], S. Josefsson [b], K. Wiberg [b], B. Vinnerås [a]

[a] Department of Energy and Technology, Swedish University of Agricultural Sciences
[b] Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences

HIGHLIGHTS

- Degradation of pharmaceuticals and pesticides in fly larvae composting (FLC).
- Half-life considerably shorter in FLC than in control with no larvae.
- Half-life of carbamazepine was less than two days in FLC.
- No bioaccumulation in larvae detected.
- FLC could impede the spreading of pharmaceuticals and pesticide in the environment.

GRAPHICAL ABSTRACT



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ABSTRACT

A novel and efficient organic waste management strategy currently gaining great attention is fly larvae composting. High resource recovery efficiency can be achieved in this closed-looped system, but pharmaceuticals and pesticides in waste could potentially accumulate in every loop of the treatment system and spread to the environment. This study evaluated the fate of three pharmaceuticals (carbamazepine, roxithromycin, trimethoprim) and two pesticides (azoxystrobin, propiconazole) in a fly larvae composting system and in a control treatment with no larvae. It was found that the half-life of all five substances was shorter in the fly larvae compost (<10% of control) and no bioaccumulation was detected in the larvae. Fly larvae composting could thus impede the spread of pharmaceuticals and pesticides into the environment.

1. Introduction

Inadequate or absent organic waste management can result in severe negative environmental and anthropogenic impacts. Pathogens present in organic wastes, *e.g. Salmonella* spp., can contribute to the spread of diseases (Hoornweg and Bhada-Tata, 2012), while the nutrients in organic waste can cause eutrophication if leached into water bodies. However, if applied to arable land, these nutrients can instead be of value, *e.g.* in the production of agricultural crops (Diacono and Montemurro, 2010). Capturing the nutrients found in organic wastes could greatly alleviate the environmental burden, by avoiding the negative impact of inadequate treatment and reducing the need for chemical fertilisers (Good and Beatty, 2011). However, the cost of collection and treatment of the organic fraction in the waste is not covered by the value of the products, which often consist of compost and/or biogas (Diener et al., 2011b). Thus unless organic waste management is subsidised, use of the waste even for production of biogas is not

* Corresponding author.
E-mail address: cecilia.lalander@slu.se (C. Lalander).

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economically viable (Gebrezgabher et al., 2010). Another pressing issue globally is the increased consumption of livestock products, which has led to ever greater demand for animal feed (Steinfeld et al., 2006). The recent high demand for protein feedstuffs has encouraged many unsustainable practices, such as soy bean production on virgin land and overfishing of wild fish stocks (Alder et al., 2008; Elizabeth et al., 2010). In recent years, fly larvae composting has gained considerable attention as an alternative organic waste management strategy (Čičková et al., 2015). In this composting technology, fly larvae, *e.g.* of the black soldier fly (BSF, *Hermetia illucens*) convert organic waste into two valuable products: animal feed protein in the form of fly larvae (Stamer, 2015) and a treatment residue that can be used as organic fertiliser (Kováčik et al., 2010). The larvae of BSF are high in protein (40%) and fat (30%) (St-Hilaire et al., 2007). Fly larvae composting is an efficient and quick composting method for organic waste, with degradation rates of up to 70% on a dry matter (dm) basis being demonstrated (Diener et al., 2011a). The greatest advantage of fly larvae composting from a waste management perspective is the generation of valuable products, which brings about a shift in the organic waste value chain, enabling the treatment to bear its own costs (Diener, 2010).

The hazards associated with release of toxic substances (*e.g.* pesticides) in the environment have been known for many decades and have resulted in the banning of many chemical agents and strict measures to avoid unnecessary spreading. It is well known that if toxic pollutants enter surface water bodies, there is a risk of these or their metabolites reaching end-points where they can cause adverse effects on ecological systems (Köhler and Triebskorn, 2013). However, environmental pollution by pharmaceuticals only started to be recognised as a serious concern during the past decade. The pharmaceutical industry is constantly developing new therapeutic agents, many of which are persistent, resulting in biologically active pharmaceuticals being found intact in the aquatic environment and posing a risk of unintentional exposure in humans and wildlife (Brodin et al., 2013; Khetan and Collins, 2007). In closed-loop systems, such as fly larvae composting systems, the fate of pharmaceuticals and pesticides is of even greater interest, as these substances could potentially accumulate in every loop of the treatment system. In a previous study, Zhang et al. (2014) examined removal of common antibiotics in a combined fly larvae (*Musca domestica*)/vermicomposting system and observed cumulative removal rates of around 70% for oxytetracycline, chlortetracycline and sulfadiazine, while the reduction in other antibiotics investigated was 30–40%. However, up-to-date knowledge on the fate of pharmaceuticals and pesticides in fly larvae composting systems is limited.

The objectives of this study were thus to assess: *i*) potential bioaccumulation of selected pharmaceuticals and pesticides in BSF larvae, and *ii*) potential degradation of pharmaceutical and pesticide residues over time in fly larvae composting compared with a control with no larvae. Five biologically active compounds were selected based on their common use and persistence in the environment: the pharmaceuticals carbamazepine, roxithromycin and trimethoprim and the pesticides azoxystrobin and propiconazole.

2. Materials and methods

2.1. Target compounds

The target compounds were: carbamazepine (CAS 298-46-4), roxithromycin (CAS 80214-83-1), trimethoprim (CAS 738-70-5), azoxystrobin (CAS 131860-33-8) and propiconazole (CAS 60207-90-1). Carbamazepine (carboxamide-^{13}C, ^{15}N) and trimethoprim-d_9 were used as mass-labelled internal standards (IS).

2.2. Experimental set-up

Dog food (Purina Pro Plan puppy; 40% dm) was used as the feed (compost) substrate, as it is comparable to food waste in terms of nutrient composition and fat/energy content (Vinnerås et al., 2003). At the start of the experiment, 63 g of substrate were inoculated with the selected pharmaceuticals (carbamazepine 1.8–1.9 mg g^{-1} dm, trimethoprim 5.9–9.9 mg g^{-1} dm, roxithromycin 5.8–5.9 mg g^{-1} dm) and pesticides (azoxystrobin 2.4–4.6 mg g^{-1} dm, propiconazole 3.2–14.1 mg g^{-1} dm). Bioaccumulation in the larvae and degradation in the substrate were investigated in batch systems over a period of 27 days in three different set-ups comprising: *i*) spiked substrate and BSF larvae (spiked BSF; $n = 3$), *ii*) spiked substrate and no BSF larvae (control substrate; $n = 3$) and *iii*) unspiked substrate and BSF larvae (control BSF; $n = 1$) (Fig. 1).

The spiked BSF treatments were fed with uncontaminated substrate on days 2, 5, 7, 9, 12, 14 and 16, at a feeding rate of 100 mg substrate $larva^{-1}$ day^{-1} (Diener et al., 2009). Similar amounts were added to the control treatments, to replicate the dilution effect on the concentration of the target substances. The uncontaminated substrate was mixed into the treatments in the control, while it was added without mixing to the larvae treatments as the movement of the larvae was assumed to be sufficient.

2.3. Sampling

Each larval treatment received 450 10-day-old larvae (average weight 14 mg $larvae^{-1}$) at the start of the experiment. On days 2 and 3, 50 larvae were removed and on day 5 and 12, 35 larvae were removed for analyses of pharmaceuticals and pesticides. The remaining larvae were allowed to develop into prepupae, and in total 165–230 prepupae were collected between day 21 and day 27 as they migrated from the feed. The remaining prepupae were allowed to develop into flies and were collected between day 64 and day 104 as flies. Upon collection, larvae, prepupae and flies were allowed to empty their gut for 24 h and were then stored at −19 °C until analysis. A set of 10 g samples of substrate (treatment residues) were collected from the larvae treatment and control on days 2, 5, 12 and 23 and stored at −19 °C until analysis.

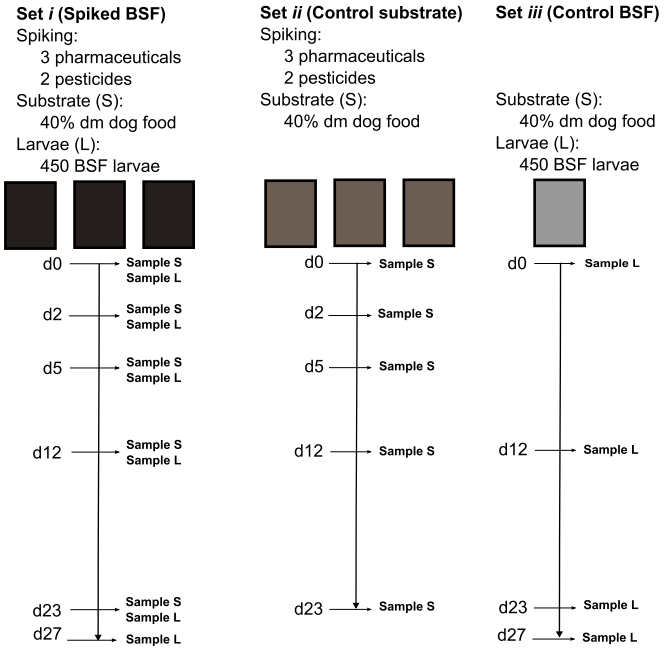

Fig. 1. Schematic representation of the experimental set-up comprising; *i*) spiked BSF treatment, *ii*) spiked substrate control and *iii*) unspiked control BSF; and the timeline of sampling of substrate (Sample S) and of larvae (Sample L) on days 0, 2, 5, 12, 23 and 27.