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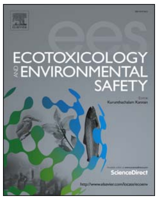

# Antimicrobial compounds (triclosan and triclocarban) in sewage sludges, and their presence in runoff following land application

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## ABSTRACT

The reuse of treated municipal sewage ('biosolids') on land is an effective method to divert waste away from landfill and to use an alternative, low cost method of fertilisation. While legislation has mainly focused on the control of nutrient and metal application rates to land, other potentially harmful emerging contaminants (ECs) may be present in biosolids. Up to 80% of municipal sewage sludge is reused in agriculture in Ireland, which is currently the highest rate of reuse in Europe. However, unlike other countries, no study has been conducted on the presence of ECs across a range of wastewater treatment plants (WWTPs) in this country. This study evaluated the concentrations of two ECs in sewage sludge, the antimicrobials triclosan (TCS) and triclocarban (TCC), and their presence in surface runoff following land application in controlled rainfall simulation studies. In 16 WWTPs, concentrations of TCS and TCC were 0.61 and 0.08 μg $g^{-1}$, which is at the lower end of concentrations measured in other countries. The concentrations in runoff post land application were also mainly below the limits of detection (90 ng $L^{-1}$ for TCS, 6 ng $L^{-1}$ for TCC), indicating that runoff is not a significant pathway of entry into the environment.

## 1. Introduction

The reuse of treated municipal sewage sludge ("biosolids") in agriculture provides the necessary nutrients and micronutrients essential for plant and crop growth (Latare et al., 2014; Liu et al., 2015). Biosolids may be used as a soil conditioner, improving its physical (e.g. water holding capacity; Cele and Maboeta, 2016) and chemical properties (e.g. soil test phosphorus; Shu et al., 2016). Their use also addresses European Union (EU) policy on sustainability and recycling of resources (COM, 2014a).

There are several issues associated with the reuse of municipal sewage sludge in agriculture (Peyton et al., 2016). While many of these are issues of perception (Robinson et al., 2012), there is considerable concern, which is scientifically based, regarding a number of substances that may be present in biosolids. There are concerns regarding pharmaceutical and personal care products (PPCPs), antimicrobial compounds, and other endocrine-disrupting compounds and synthetic compounds in biosolids (Clarke and Cummins, 2014) and the associated risk of contamination of soil, and surface and groundwater (Hanief et al., 2015; Fu et al., 2016). Toxic metals in sludge may accumulate in the soil and crops and enter the food chain following continuous applications to land (Stietiya and Wang, 2011; Latare et al., 2014; García-Santiago et al., 2016). Organic and inorganic contaminants may be lost along surface runoff and leaching pathways following land application (Gottschall et al., 2012; Peyton et al., 2016). Furthermore, there is a risk of emission and transport of bioaerosols containing manure pathogens following land application of biosolids (Brooks et al., 2005; Jahne et al., 2015). These concerns are confounded by the fact that although EU legislation controls the application of biosolids to land by setting limit values for nutrients and metals (EEC, 1986), no safety guidelines currently exist for PPCPs or many emerging contaminants (ECs).

Wastewater treatment plants (WWTPs) cannot fully remove PPCPs or other organic or synthetic compounds from wastewater, the removal of which is affected by treatment technique and operating conditions (Narumiya et al., 2013). Removal pathways include sorption onto

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sludge (Ternes et al., 2004) and biodegradation/biotransformation (Verlicchi et al., 2012). Despite this, several compounds have been measured in digested sewage sludge (Walters et al., 2010; Verlicchi and Zambello, 2015). Therefore, when biosolids are spread on land, there is a risk of indirect exposure to humans through several pathways, including the food chain (consumption of crops, meat, dairy products and drinking water), surface runoff, and leaching to land drainage systems or groundwater used for abstraction by water treatment plants. Clarke et al. (2016) developed a quantitative risk ranking model for human exposure to 16 organic contaminants following biosolids application to land. They found that while nonylphenols had the highest risk, the antimicrobials, triclosan (TCS) and triclocarban (TCC), were considered more of an evolving risk, as these contaminants are emerging and have only recently been restricted within the US (US-FDA, 2015a, 2015b) and EU (COM, 2014b). In addition, both compounds are commonly the most abundant contaminants in biosolids (McClellan and Halden, 2010) and both are listed in the top contaminants of concern worldwide (von der Ohe, 2012; Verlicchi and Zambello, 2015). Triclosan, a broadspectrum bacteriostat and fungicide, and TCC, a fungicide and bacteriostat, are known toxins for humans and have been linked to inhibition of muscle function (Cherednichenko et al., 2012), resistance to antibiotics used in human medicine (Yazdankhah et al., 2006), and ecotoxicity in the environment such as the inhibition and killing of algae, crustaceans and fish (Chalew and Halden, 2009). On account of this, these compounds are the main focus of the current study.

In the EU there are considerable differences in national policy regarding the reuse of biosolids in agriculture. In some countries, such as Belgium (Brussels and Flanders), Switzerland and Romania, the reuse of biosolids in agriculture is prohibited (Milieu et al., 2013a, 2013b, 2013c), whereas in other countries, such as Ireland, up to 80% of municipal wastewater sludge is reused in agriculture (EPA, 2014; Eurostat, 2016). However, despite this, as the country with the greatest reuse of biosolids on land, no study has examined the concentrations of TCS or TCC in biosolids from WWTPs in Ireland. Such national studies of TCS and TCC have been conducted in the USA, Canada, India and South Korea (Table 1), but currently no extensive study across a range of WWTPs exists in the EU.

Once applied to land, TCS and TCC in biosolids may either accumulate in plants (Mathews et al., 2014); accumulate, biodegrade or biotransform in soil (Wu et al., 2009), or be released in surface runoff during rainfall-runoff or leaching events (Sabourin et al., 2009). The potential for loss via surface runoff or leaching depends on their availability in soil, which is a function of their persistence or half-life (Fu et al., 2016). It has been speculated that the persistence of TCS or TCC in the soil may be enhanced by the organic content of the soil (Fu et al., 2016), soil temperature (which is positively correlated to half-life), the physicochemical properties of the compounds (Wu et al., 2009), and the presence of co-contaminants (Walters et al., 2010), making them potentially more available for loss in surface runoff during rainfall events. Many studies have investigated losses of TCS and TCC in surface runoff from agricultural lands (Table 2), but few, if any, studies have investigated the surface losses from lands which have received sludge applications from the same WWTP having undergone different treatments. Such an experiment may allow the potential for surface water contamination from different sludge treatment methods to be evaluated.

Therefore, the aim of this study was to (1) characterise, for the first time, the TCS and TCC in biosolids from a range of WWTPs in Ireland, and (2) measure the surface runoff of TCS and TCC under successive rainfall simulations at 1, 2 and 15 days after application of two types of biosolids, originating from the same WWTP.

## 2. Methodology

### 2.1. WWTP identification and sample collection

In January and February 2015 (Winter in Ireland), biosolids were collected from 16 WWTPs, which had population equivalents (PEs, i.e. the amount of oxygen demanding substances in wastewater equivalent to the demand of the wastewater produced by a single person) ranging from 2.3 million to 6500. Details of the PE and influent wastewater characteristics of each WWTP are given in Healy et al. (2016a). Most WWTPs received quantities of landfill leachate in low quantities (less than 2% of the influent biochemical oxygen demand (BOD) load), whilst others received industrial, commercial and domestic septic tank sludge comprising up to 30% of the influent BOD load. Anaerobic digestion of sewage sludge was carried out in five WWTPs, thermal drying in eight WWTPs, and lime stabilisation in four WWTPs (one WWTP carried out both anaerobic digestion and thermal drying). Discrete samples (n = 8) of biosolids were collected in clean LDPE containers from each WWTP, and were pulverised in an agate ball mill (Fritsch™ Pulverisette 6 Panetary Mono Mill) with a rotational speed of 500 rpm for 5 min (repeated three times). The metal content of the biosolids are reported in Healy et al. (2016a).

### 2.2. Field study site description and runoff simulations

Treated municipal sewage sludge from the WWTP in which anaerobic digestion and thermal drying was carried out, was used in this study. Raw, untreated sludge from the same WWTP was modified by the authors with calcium oxide following the method outlined by Fehily Timoney and Company (1999). Therefore, the anaerobically digested (AD), thermally dried (TD) and lime stabilised (LS) biosolids used in this study originated from the same WWTP. The biosolids were applied to replicated (n = 3), hydraulically isolated, field-scale micro-plots, each measuring 0.4 m-wide by 0.9 m-long. The slope of each micro-plot ranged from 2.9% to 3.7% and each micro-plot was instrumented with a runoff collection channel, which allowed all surface runoff to be collected over the duration of a rainfall event (Peyton et al., 2016). The site was planted with ryegrass for over twenty years and the soil pH ranged from 5.9 to 6. The soil in all micro-plots was classified as loam and the soil organic matter ranged from 8.1% to 9.0%. Full classification of the plots is detailed in Peyton et al. (2016).

Anaerobically digested, TD and LS biosolids were applied by hand to the surface of each micro-plot at the maximum legal application rate in

**Table 1**
Triclosan and triclocarban concentrations (μg g$^{-1}$ dry weight) in national studies of biosolids produced in municipal wastewater treatment plants.

| Reference | Country | # WWTPs examined | Mean concentration (μg g$^{-1}$) | | Maximum concentration (μg g$^{-1}$) | |
|---|---|---|---|---|---|---|
| | | | Triclosan | Triclocarban | Triclosan | Triclocarban |
| McClellan and Halden (2010) | USA | 94 | 12.6 | 36 | 19.7 | 48.1 |
| Subedi et al. (2015) | India | 5 | 1.2 | 7.0 | | |
| Chu and Metcalfe (2007) | Canada | 4 | 4.2 | 4.3 | | |
| Guerra et al. (2014) | Canada | 6 | 6.8 | 2.9 | 11.0 | 8.9 |
| Subedi et al. (2014) | S. Korea | 40 | | 3.1 | | 6.9 |
| This study | Ireland | 16 | 0.61 | 0.08 | 4.9 | 0.15 |