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## Quantitative risk assessment of antimicrobials in biosolids applied on agricultural land and potential translocation into food

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### ABSTRACT

The use of biosolids as a fertiliser may be an indirect route for contaminants into the food chain. One of the main concerns regarding the spreading of biosolids on agricultural land is the potential uptake of contaminants into plants which may bio-transfer into grazing animals that supply the food chain directly (e.g. meat and milk) and hence are subsequently consumed. The aim of this project was to create a quantitative risk assessment model to estimate the fate and translocation of triclosan (TCS) and triclocarban (TCC) into the feed (grass) and food chain with subsequent human exposure. The model's results indicate that TCS and TCC have low potential to transfer into milk and beef following the ingestion of contaminated grass by dairy cows. Mean estimated TCS and TCC residues in milk and beef show that TCC had the greatest concentration (mean values of  $7.77 \times 10^{-6} \text{ mg kg}^{-1}$  in milk and  $1.36 \times 10^{-4} \text{ mg kg}^{-1}$  in beef). Human exposure results show that TCC was greater for milk consumption in infants (1–4 years) (mean value  $1.14 \times 10^{-7} \text{ mg kg}^{-1} \text{ bw d}^{-1}$ ) and for beef consumption by teens (12–17 years) (mean value  $6.87 \times 10^{-8} \text{ mg kg}^{-1} \text{ bw d}^{-1}$ ). Concentrations of TCS and TCC were well below the estimated acceptable daily intake (ADI). Human health risk was estimated by evaluation of the hazard quotient (HQ), which used the NOAEL as a toxicity endpoint, combined with milk and beef human exposure estimates. HQ results show that all values were  $< 0.01$  (no existing risk). A sensitivity analysis revealed that the  $K_{ow}$  and initial concentration in biosolids as the parameters of greatest importance (correlation coefficients 0.91 and 0.19, respectively). This highlights the importance of physio-chemical properties of the compounds and their detection in biosolids post wastewater treatment along with their persistence in soil following application. This model is a valuable tool in which to ascertain the potential transfer of contaminants in the environment into animal forage with knock on consequences for exposure through the human food chain.

### 1. Introduction

The two most important farming sectors in Ireland are the milk and meat sectors, accounting for approximately 69% of agricultural output (DAFM, 2016). The value of overall beef exports from Ireland was 2.27 billion in 2014 (EC, 2016), while dairy exports have grown to 3.1 billion in 2014 (IDIA, 2016). There are currently 6.96 million cattle in Ireland according to the June 2015 livestock survey (Bord bia, 2016). The total land area of Ireland is 6.9 million hectares of which 4.5 million hectares is used for agriculture (DAFM, 2016). Eighty one percent of the agricultural area is devoted to pasture, hay and grass silage (3.6 million hectares) and 11% to rough grazing (Bord bia, 2016). Under the 'Code of Good Practice for the use of biosolids in agriculture' (Fehily Timoney & Company, 1999) it states that there are constraints on grazing following application of biosolids to agricultural land. 'Cattle

should not be turned out onto pasture that has been fertilised with biosolids until 3-6 weeks after the date of application'. The interval between application and commencement of grazing will depend on the level of incorporation of biosolids into the soil (Fehily Timoney & Company, 1999). In Ireland, 53,543 tonnes dry solids (tds) of biosolids are generated each year, of which 98% is disposed to agricultural land. It has been predicted that this figure will grow to 96,442 tds/annum by 2040 (Irish Water, 2015).

One of the main concerns for human health regarding the spreading of biosolids on agricultural land is the potential uptake of contaminants into plants which may bio-transfer into grazing animals that are subsequently consumed by humans. Studies have shown that conventional wastewater treatment does not fully eliminate contaminants such as pharmaceuticals (i.e. beta-blockers, carbamazepine, paracetamol and diclofenac) (Clarke & Cummins, 2014; Harris, Cormican, & Cummins,

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2012; Igos et al., 2012; Jelic, Gros, Petrovic, Ginebreda, & Barcelo, 2012); therefore contaminants may still be present in the treated sludge. Biosolids are rich in organic matter and may contain up to 38% organic carbon on a dry mass basis. Therefore, repeated application of biosolids may greatly increase a soil's organic carbon content (OC), leading to enhanced sorption or reduced chemical bioavailability (Fu, Wu, Ye, Ernst, & Gan, 2016). On one hand, biosolid application is a direct point source for contaminants into the environment (Clarke, Healy, Fenton, & Cummins, 2016), on the other hand, increased organic matter may inhibit plant uptake due to reduced bioavailability. A number of studies have demonstrated the uptake of contaminants into plants (Boxall et al., 2006; Carter et al., 2014; Holling, Bailey, Heuvel, & Kinney, 2012; Prosser & Sibley, 2015; Sabourin et al., 2012).

The main route of human exposure to many highly lipophilic contaminants is through ingestion of contaminated agricultural products such as beef and milk (USEPA, 2005). Livestock can ingest contaminants from soil by grazing and/or feeding on harvested forage. In countries where animals can graze all year round, average soil ingestion has been estimated as 4.5% of the dry matter intake for sheep and 6% for cattle when pasture was the only feed source (Duarte-Davidson & Jones, 1996). While there have been many models developed to predict animal uptake, including relating bio-transfer concentrations (BCF) in livestock to physio-chemical properties (Rodrigues, Pereira, Duarte, & Romkens, 2012; Travis & Arms, 1988), there are significant knowledge gaps with regards to actual contaminant concentrations in livestock following direct ingestion of grass from biosolid amended agricultural land. Lupton, Dearfield, Johnson, Wagner, and Huwe (2015) conducted a study to determine plasma and tissue depletion kinetics in cattle. The cattle (2 steers and 4 heifers) were dosed with perfluorooctane sulfate (PFOS) at  $0.098 \text{ mg kg}^{-1}$  weight and  $9.1 \text{ mg kg}^{-1}$ , respectively. Plasma depletion half-lives for steers and heifers were  $120 \pm 4.1$  and  $106 \pm 23.1$  days, respectively. Specific tissue depletion half-lives ranged from 36 to 385 days for intraperitoneal fat, back fat, muscle, liver, bone, and kidney. The results of the experiment showed that PFOS in beef cattle had a sufficiently long depletion half-life to permit accumulation in edible tissues.

The proportion of an organic or inorganic contaminant taken up by plant roots and its translocation route within the plant depends on its physio-chemical properties (Goldstein, Shenker, & Chefetz, 2014), the plant's physical characteristics and soil properties (Taylor-Smith, 2015). The  $\log K_{ow}$  or  $\log$  of the octanol water partitioning coefficient represents a compound's propensity to partition into either polar or non-polar mediums (Fent, Weston, & Caminada, 2006). Highly lipophilic contaminants characterised by high octanol water partitioning coefficients ( $K_{ow} > 3 \log$  unit) or low water solubilities, have a high tendency to be absorbed by plant roots from water (Li, Sheng, Chiou, & Xu, 2005). For example, Carter et al. (2014) attributed the uptake of pharmaceuticals and a personal care product into radishes and ryegrass to the physio-chemical properties of the contaminants, including Henry's Law constant, water solubility and octanol water partition coefficient. Wu, Spongberg, Witter, Fang, and Czajkowski (2010) also demonstrated how the  $K_{ow}$  predictions of contaminant behaviour in plants correlated with the bioconcentration factor of the contaminants. Wild and Jones (1992) categorised non-ionised organic contaminants with  $\log K_{ow} > 4$  as having a high potential for retention in plant roots. Thus, the octanol water partition coefficient ( $K_{ow}$ ) has been suggested as a reliable indicator of uptake behaviour (Goldstein et al., 2014). Lipophilic organic contaminants possess a greater tendency to partition into plant root lipids than hydrophilic contaminants (Duarte-Davidson & Jones, 1996). Chemicals in soil enter plants primarily through the root system and the degree of uptake from soil into root tissues appears to be proportional to the octanol-water partition coefficient ( $K_{ow}$ ).

Since the 1960's antimicrobials triclosan (TCS) and triclocarban (TCC) have been in use as antibacterial agents in many products such as

toothpaste, soaps, creams, etc. (Xia et al., 2010). Toxicological reports have shown that TCC has the potential to disrupt excitation coupling in skeletal and cardiac muscles in humans (Clarke, Peyton, Healy, Fenton, & Cummins, 2016). Studies suggest that TCS and TCC may persist in the sludge post wastewater treatment. The US Environmental Protection Agency (2009) conducted a study on 84 WWTPs to analyse the sludge. TCC was detected in 100% of the samples at a concentration range of  $0.187\text{--}441 \text{ mg kg}^{-1}$ , TCS was detected 94% of the time with a concentration range of  $0.430\text{--}133 \text{ mg kg}^{-1}$ . Heidler, Sapkota, and Halden (2006) reported removal efficiencies of TCS and TCC in digested sludge were 98% and 97%, respectively. Ogunyoku and Young (2014) studied removal efficiencies of conventional wastewater treatment on TCS and TCC. Results show that TCS was more rapidly removed than TCC, indicating that TCS was more readily bio-transformed than TCC.

Once introduced to the environment, TCS and TCC sorb to soils and sediment and are not predicted to readily degrade (Aryal & Reinhold, 2011). Ying, Yu, and Kookana (2007) reported that TCS degraded faster than TCC by microbial processes in the soil under aerobic conditions. The half-life in air is estimated to be  $1 \text{ d}^{-1}$  for TCS and  $0.75 \text{ d}^{-1}$  for TCC (PBT profiler 2012). Ying et al. (2007) reported half-lives in air of  $0.66 \text{ d}^{-1}$  for TCS and  $0.75 \text{ d}^{-1}$  for TCC. Volatilisation is not expected to be a significant removal mechanism for TCC and TCS (Ogunyoku & Young, 2014). Sorption ( $K_d$ ) and persistence (measured as half-life,  $T_{1/2}$ ) are considered the two primary variables controlling the availability and hence offsite transport potential of contaminants in soil (Fu et al., 2016). To describe the distribution of a chemical in soil, the soil-water partition coefficient ( $K_d$ ) is a suitable measure. The  $K_d$  is generally proportional to the hydrophobicity of the compound and the amount of soil organic matter. The experimental half-life of TCS and TCC in soil ranges from 87 to 231 and 18–58 days, respectively in aerobic soils with longer half-lives in anaerobic soils (Ying et al., 2007). TCS and TCC are both hydrophobic with  $\log K_{ow}$  values of 4.76 and 4.90, respectively (Dhillon et al., 2015). Hence, accumulation of these compounds has been observed in plants (Aryal & Reinhold, 2011; Wu, Spongberg, Witter, & Sridhar, 2012), animals (Coogan & Point, 2008; Higgins et al., 2011; Kinney et al., 2008), humans (Allmyr, Adolfsson-Erici, McLachlan, & Sandborgh-Englund, 2006) and the potential of TCS and TCC as endocrine disruptors are also shown (Chen et al., 2008; Hinthner, Bromba, Wulff, & Helbing, 2011). Wu et al. (2012) demonstrated that after 60 days growth, TCS and TCC had accumulated and translocated into above ground parts of the soybean plant following application of biosolids and reclaimed waste water. Prosser and Sibley (2015) reported the uptake of TCS and TCC in the edible portions of green pepper, carrots, cucumber, tomato, radish and lettuce plants grown in biosolid amended land. TCS was only detected in cucumber and radish up to  $5.2 \text{ ng/g dw}$ , while TCC was detected in carrot, green pepper, tomato and cucumber up to  $5.7 \text{ ng/g dw}$ . However, it was estimated that vegetable consumption represents  $< 0.5\%$  of the acceptable daily intake of TCS and TCC. Aryal and Reinhold (2011) measured concentrations of TCS at approximately 20 and  $40 \text{ } \mu\text{g/g dw}$  in the root and 8 and  $5 \text{ } \mu\text{g/g dw}$  in the stem of pumpkin and zucchini plants, respectively despite a low concentration of TCS ( $0.18 \text{ } \mu\text{g/g}$ ), however liquid biosolids were applied prior to seeding and 8 weeks after seeding accounting for the high levels in the plants.

In this study, the aim was to develop a quantitative risk assessment model to estimate the fate and translocation of antimicrobials (triclosan and triclocarban) into biosolid receiving agricultural soils with transfer into grass and subsequent potential transfer into the food production chain (beef and grass) and potential human consumption/exposure (Fig. 1). The focus of this study is on the primary produce milk and beef only, the accumulation of contaminants in secondary products (e.g. cheese, processed meat products) is not considered and outside the scope of the current study.

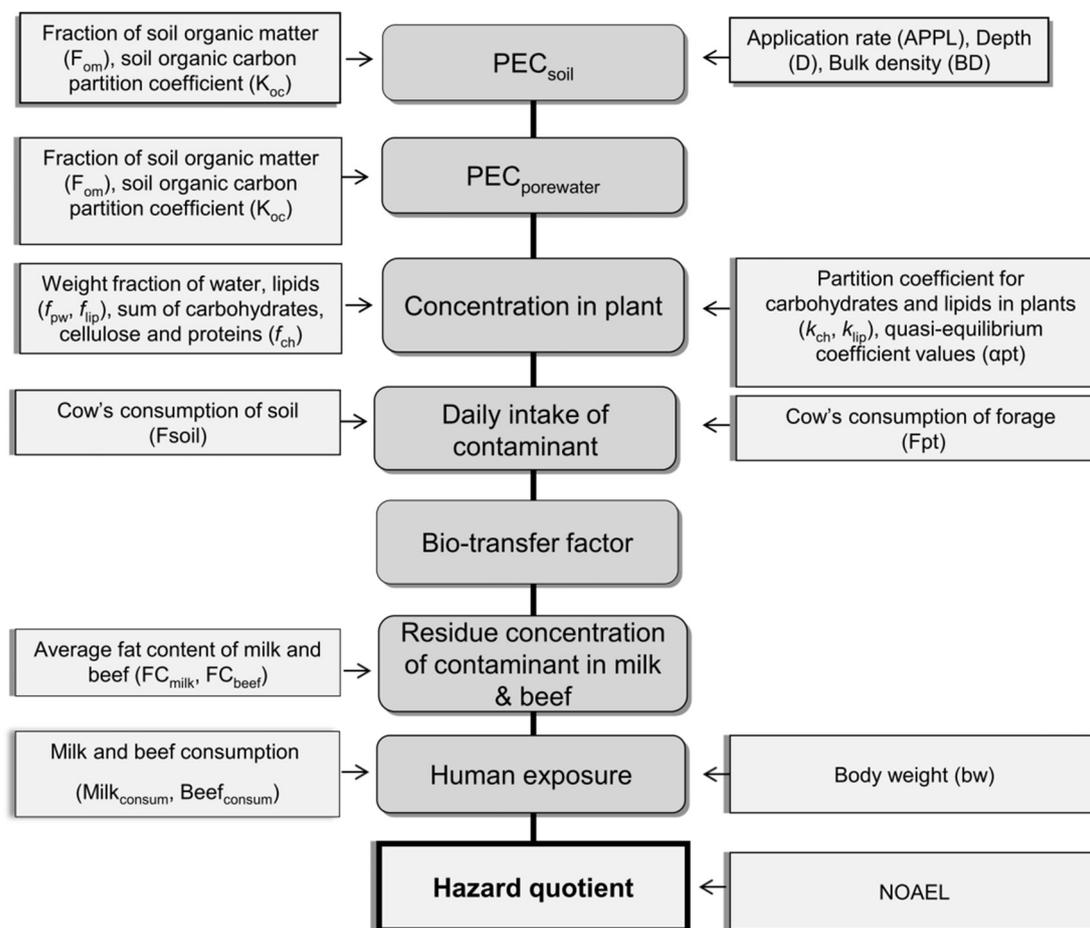


Fig. 1. Flow diagram of inputs and outputs for the quantitative plant uptake and translocation into the food chain model.

2. Materials and methods

2.1. Soil-to-plant transfer model

The  $PEC_{soil}$  was estimated by developing a distribution of contaminant exposure based on the variability and uncertainty of the predicted environmental concentrations in biosolids. The concentration in the soil ( $C_{soil}$ ;  $mg\ kg^{-1}$ ) immediately following a single biosolid application was calculated based on the concentration of the contaminant in the biosolids, application rate, crop intersection, mixing depth of soil and the soil bulk density following biosolids application according to the guidelines of the European Chemicals Bureau's Technical Guidance Document on Risk Assessment Part II (EC, 2003a).

$$C_{soil} = (C_{sludge} \times APPL \times (1 - f_{int}/100))/(D \times BD) \quad (1)$$

where:

$C_{sludge}$  is the concentration of the contaminant of interest in biosolids ( $mg\ kg^{-1}$ ).

APPL is the application rate of biosolids on agricultural land for one application ( $kg\ m^{-2}$ ).

$f_{int}$  is the fraction intercepted by the crop (-).

D is the depth (m)

BD is the soil bulk density ( $kg\ m^{-3}$ ).

The peer reviewed literature was searched for Irish and European organic contaminant concentrations in biosolids (Table 1) and probabilistic distributions were fitted to characterise uncertainty/variability in the level of TCS and TCC in biosolids (Table 2). Uncertainty regarding the application rate was represented using a triangular distribution (minimum 300; mean 330; and maximum  $520\ g\ m^{-2}$ ) (Table 3). The application rate of biosolids was retrieved from Lucid,

Table 1  
Concentrations of TCS and TCC in biosolids ( $\mu g\ kg^{-1}$ ).

Concentration in biosolids ( $\mu g\ kg^{-1}$ )		
Triclosan	<sup>a</sup> 1840	<sup>a</sup> (Clarke & Smith, 2011), <sup>b</sup> (Davis, Klosterhaus, & Stapleton, 2012), <sup>c</sup> (Walters, McClellan, & Halden, 2010), <sup>d</sup> (Chu and Metcalfe, 2007)
	2830	
	3210	
	5993	
	<sup>b</sup> 4370	
	1429	
	11,843	
	12,876	
	1265	
	<sup>c</sup> 7860	
	<sup>d</sup> 9080	
	11,550	
	1490	
	1110	
1510		
17,950		
Triclocarban	<sup>a</sup> 5970	<sup>a</sup> (Chu & Metcalfe, 2007), <sup>b</sup> (Snyder, O'Connor, & McAvoy, 2010), <sup>c</sup> (McClellan & Halden, 2010), <sup>d</sup> (Walters et al., 2010), <sup>e</sup> (Cha & Cupples, 2010)
	3050	
	5490	
	4920	
	3300	
	3490	
	3700	
	4780	
	<sup>b</sup> 19000	
	<sup>c</sup> 36000	
	<sup>d</sup> 2715	
<sup>e</sup> 4510		
7085		

**Table 2**  
Model inputs, distributions and outputs for PEC<sub>soil</sub>.

Stage	Symbols	Description	Model/distribution	Units
PEC <sub>soil</sub>	C <sub>sludge</sub>	Concentration in biosolids	Uniform or triangular (contaminant specific, Table 1)	mg kg <sup>-1</sup>
	APPL	Application rate	Triangular (300,330, 520)	kg m <sup>-2</sup>
	<i>fint</i>	Crop intersection	Triangular (0, 10, 20)	–
	D	Depth	0.1	m
	BD	Bulk density	Uniform (min 800, max 1000)	kg m <sup>-3</sup>
Output	C <sub>soil</sub>	(C <sub>sludge</sub> × APPL × (1 – <i>fint</i> × 100) / (D × BD)	Uniform (contaminant specific, Table 3)	mg kg <sup>-1</sup>
	DT <sub>50</sub>	Half-life in soil	Uniform (min 21, max 42)	d <sup>-1</sup>
	k	Dissipation rate constant	Ln (2)/DT <sub>50 soil</sub>	d <sup>-1</sup>
	t	Time to graze	Uniform (min 21, max 42)	d <sup>-1</sup>
Output	PEC <sub>soil</sub>	Concentration of contaminant in soil following dissipation	C <sub>soil</sub> × e <sup>-kt</sup>	mg kg <sup>-1</sup>
	C <sub>applied</sub>	Concentration of contaminant applied on grass	C <sub>sludge</sub> × APPL × <i>fint</i>	mg m <sup>-2</sup>
	DT <sub>50</sub>	Half-life in air	Uniform (contaminant specific, Table 3)	d <sup>-1</sup>
	K	Dissipation rate constant	Ln (2)/DT <sub>50 air</sub>	–
	P <sub>d</sub>	Plant density	1.8	kg m <sup>-2</sup>
Output	C <sub>plant</sub>	Concentration on plant following dissipation	C <sub>applied</sub> × e <sup>-kt</sup> /P <sub>d</sub>	mg kg <sup>-1</sup>

Fenton, and Healy (2013). It was assumed that the biosolids were spread on grassland.

The mixing depth (0.1 m) was obtained from the EU Technical Guidance Document on Risk Assessment part II (EC, 2003a) and is representative of grassland as grassland is not traditionally ploughed. The BD of soil (800–1000 kg/m<sup>3</sup>) (uniform distribution) were obtained from [Vero, Antille, Lator, and Holden \(2014\)](#), which is a typical range for the upper 100 mm of the soil profile of Irish grasslands. It was assumed that the soil was poorly drained with OM and BD ranges as stated. The fraction intercepted by the crop was based on tabular interception fractions values as proposed by [Linders, Mensink, Stephenson, Wauchope, and Racke \(2000\)](#) which were based on field experiments found in the literature. The authors adopted the approach that interception fraction plus the soil deposition fraction is unity (*fint* + *Fsoil* = 1). The study focuses on interception rather than retention. Conceptually, it is assumed that both interception and deposition on soil are instantaneous processes. Crop interception (*fint*) was estimated to be a triangular distribution (minimum 0, most likely 10 and maximum 20%) assuming a worst-case scenario.

The degradation kinetics in soil was described using a first order reaction model. The half-life (DT<sub>50</sub>) of each contaminant in soil was obtained from the peer reviewed literature and shown in [Table 3](#). To account for uncertainty and variability in the data, TCS and TCC were assigned a uniform distribution. The dissipation rate constant ‘k’ was obtained by Eq. (2):

$$k = \text{Ln}(2)/\text{DT}_{50\text{soil}} \quad (2)$$

The actual concentration in soil following dissipation was estimated using Eq. (3).

$$\text{PEC}_{\text{soil}} = \text{C}_{\text{soil}} \times e^{-kt} \quad (3)$$

where PEC<sub>soil</sub> is the concentration remaining in soil following dissipation. ‘t’ is the time the contaminant has in the soil prior to grazing. The Code of Good Practice for Application of Biosolids on Agricultural Land states that cattle may not be turned out onto grassland until at least 3–6 weeks following biosolid application. Therefore the time ‘t’ (in days) was assigned a uniform distribution (min 21, max 42) to allow for constraints in allowing cattle to graze.

While concentrations of TCS and TCC may leach through the soil or adhere to sludge post biosolid application, biosolids may also remain on the grass or sward of grass and be consumed by grazing cattle. To account for potential consumption of applied biosolids on grass, the concentration of TCS and TCC in biosolids, the application rate and the percentage of crop intersected (*fint*) were multiplied to give the concentration of contaminant on the grass swards.

$$\text{C}_{\text{applied}} = \text{C}_{\text{sludge}} \times \text{APPL} \times \textit{fint} \quad (4)$$

where C<sub>applied</sub> (mg m<sup>2</sup>) is the concentration of biosolid remaining on the grass following biosolid application.

When biosolids are applied to agricultural land, the field dissipation of the contaminants contained within the biosolids is likely to be

**Table 3**  
Properties of triclosan and triclocarban.

Contaminant	Distribution	Min	Mean	Max	References
<b>Triclosan</b>					
Log K <sub>oc</sub> (L kg <sup>-1</sup> )	Triangular <sup>a</sup>	2.7	4.0	4.7	<sup>a</sup> Barron et al., 2009, Agyin-Birikorang, Miller, & O'Connor, 2010, Chen et al., 2011, Gasperi et al., 2014, WFD, 2012
C <sub>sludge</sub> (mg kg <sup>-1</sup> )	Triangular <sup>b</sup>	1110	7298	19,676	<sup>b</sup> Chu & Metcalfe, 2007, Clarke & Smith, 2011, Walters et al., 2010, Cha & Cupples, 2010, Davis et al., 2012
K <sub>ow</sub>	Triangular <sup>c</sup>	4.38	4.66	4.8	<sup>c</sup> Coogan, 2007, Wu, Spongberg, & Witter, 2009, Chen et al., 2011, Rudel et al., 2013, Banihashemi & Droste, 2014
DT <sub>50</sub> soil	Uniform <sup>d</sup>	87		231	<sup>d</sup> Wu et al., 2009, ChempSpider, 2015.
DT <sub>50</sub> air	Uniform <sup>e</sup>	0.66		1	<sup>e</sup> USEPA, 2013, Ying et al., 2007
Henry's Law Constant	Uniform <sup>f</sup>	1.3 × 10 <sup>-3</sup>		5.2 × 10 <sup>-4</sup>	<sup>f</sup> Thompson, Griffin, Stuetz, & Cartmell, 2005
<b>Triclocarban</b>					
Log K <sub>oc</sub> (L kg <sup>-1</sup> )	Triangular <sup>f</sup>	3.59	4.06	4.9	<sup>f</sup> Ying et al., 2007, King, 2010, Cha & Cupples, 2010, ChempSpider, 2015.
C <sub>sludge</sub> (mg kg <sup>-1</sup> )	Triangular <sup>g</sup>	2715	14,756	38,839	<sup>g</sup> Chu & Metcalfe, 2007, Snyder et al., 2010, Walters et al., 2010, McClellan & Halden, 2010, Cha & Cupples, 2010.
K <sub>ow</sub>	Uniform <sup>h</sup>	2.7		7.1	<sup>h</sup> Wu et al., 2009, Agyin-Birikorang et al., 2010, Oehha, 2010
DT <sub>50</sub> soil	Uniform <sup>i</sup>	18		120	<sup>i</sup> Ying et al., 2007, Walters et al., 2010
DT <sub>50</sub> air	Uniform <sup>j</sup>	0.5		0.75	<sup>j</sup> USEPA, 2013, Ying et al., 2007
Henry's Law Constant	Uniform <sup>k</sup>	3.6 × 10 <sup>-5</sup>		8.3 × 10 <sup>-6</sup>	<sup>k</sup> ChempSpider, 2015

influenced by environmental conditions. Variations in temperature and available moisture are likely to play an important role in the dissipation of contaminants (Langdon, Warne, Smernik, Shareef, & Kookana, 2012). The half-life in air for both TCS and TCC was obtained from the PBT Profiler (USEPA, 2013) and Ying et al. (2007) (Table 3). To account for variability and uncertainty in the data, a uniform distribution (min 0.66, max 1) for TCS and (min 0.5, max 0.75) for TCC was assigned. A first order exponential decay model was used to calculate the dissipation of the contaminants on the swards of grass.

The same time 't' was used as above. It was assumed that 3 weeks had passed since the land spreading of biosolids on agricultural grasslands. The amount of fresh grass in kg per m<sup>2</sup> (P<sub>d</sub>) was obtained from Agrinet Farm Management Software (2015) and it was estimated that there was 18,000 kg of fresh grass per Ha. The overall concentration of contaminant on grass was calculated according to Eq. (5).

$$C_{\text{plant}} = C_{\text{applied}} \times e^{-kt}/P_d \quad (5)$$

where  $C_{\text{plant}}$  (mg kg<sup>-1</sup>) is the concentration remaining on the plant following dissipation.  $P_d$  is the plant density (kg m<sup>2</sup>).

In the present study the model approach developed by Chitescu, Nicolau, Romkens, and Van der Fels-Klerx (2014) and Chiou, Sheng, and Manes (2001). While the model has been modified for Irish conditions (e.g. application rates, bulk density, cow's consumption of forage and human consumption rates), there is potential to use the model universally. The effective concentration of contaminants available for plant uptake is the concentration of the contaminant in soil interstitial (pore) water. Soil composition influences the concentration of the contaminant in pore water, by its fraction of organic matter ( $F_{oc}$ ) (Chitescu et al., 2014). The model is expressed as:

$$PEC_{\text{porewater}} = PEC_{\text{soil}}/(F_{oc} \times K_{oc}) \quad (6)$$

where  $PEC_{\text{porewater}}$  is the contaminant concentration in the pore water (mg kg<sup>-1</sup>).  $F_{oc}$  is the fraction of organic matter content ( $F_{oc}$ ) in the soil; and  $K_{oc}$  is the soil organic carbon-water partitioning coefficient of the contaminants (L kg<sup>-1</sup>) (contaminant specific). Triangular distributions were used to model  $K_{oc}$  uncertainty (Table 3). The fraction of organic matter content in the soil was obtained from peer reviewed literature for  $F_{oc}$  in soil (2%, 5% and 7%) (Chalew & Halden, 2009). To account for variability and uncertainty in the data, a uniform distribution (min 2%, max 7%) was assigned (Table 4). To convert the units from mg L<sup>-1</sup> to mg kg<sup>-1</sup>, the density of water was assumed.

To calculate the concentration of contaminant in the whole plant, a partition-limited model for the passive uptake of contaminants from the external water to the plant, taking explicit account of the contaminant level in the external water (Chiou et al., 2001).

$$C_{\text{pt}} = \alpha_{\text{pt}} \times PEC_{\text{porewater}} \times [f_{\text{pw}} + f_{\text{ch}} \times K_{\text{ch}} + f_{\text{lip}} \times K_{\text{lip}}] \quad (7)$$

where  $C_{\text{pt}}$  is the concentration of the contaminant in the plant on a fresh weight base (mg kg<sup>-1</sup>);  $f_{\text{pw}}$ ,  $f_{\text{lip}}$  and  $f_{\text{ch}}$  are the weight fraction of,

respectively, water, lipids and the sum of carbohydrates, cellulose, and proteins in the plant;  $K_{\text{lip}}$  is the partition coefficient for the lipids fraction of the plant assumed to be equal to the log  $K_{ow}$ ;  $K_{\text{ch}}$  is the partition coefficient for the carbohydrate fraction of the plant, available according to  $K_{ow}$ . The symbol  $\alpha_{\text{pt}}$  is the quasi-equilibrium factor, defined as the ratio of the respective concentration of the contaminant in plant water and external water. Thus,  $\alpha_{\text{pt}} = 1$  denotes the state of equilibrium.  $\alpha_{\text{pt}} < 1$  is a measure of the approach to equilibrium (Chiou et al., 2001). The quasi-equilibrium coefficient values are based on the overall hydrophilic to lipophilic trend of the solutes in that more water soluble compounds have  $\alpha_{\text{pt}}$  values close to 1 and that the  $\alpha_{\text{pt}}$  values for lipophilic contaminants (high  $K_{ow}$  values) are  $< 1$  (Chiou et al., 2001). Therefore a value of 0.1 was assigned for both contaminants.

The weight composition of grass is comparable to ryegrass shoots. The  $f_{\text{pw}}$  (water content) was valued at 88.8%,  $f_{\text{lip}}$  (lipid content) 0.97% and  $f_{\text{ch}}$  (carbohydrate content) was 10.2% according to (Chitescu et al., 2014; Li et al., 2005).  $K_{\text{lip}}$  data were obtained from the peer reviewed literature and a uniform distribution was assigned to account for uncertainty in the data (Table 4). The partition coefficient for the carbohydrate fraction of the plant was determined from Hung, Lin, and Chiou (2010) and relates to contaminants with a log  $K_{ow}$  between 3.30 and 5.18. The calculation is based on the partitioning of five polyaromatic hydrocarbons with carbohydrates.  $K_{\text{ch}}$  was calculated according to:

$$\text{Log}K_{\text{ch}} = 1.23 \text{ log}K_{ow} - 2.42 \quad (8)$$

## 2.2. Plant to animal transfer model

The daily intake (DI) (mg d<sup>-1</sup>) of TCS and TCC in cows was calculated according to Eq. (9).

$$DI = PEC_{\text{soil}} \times F_{\text{soil}} + C_{\text{pt}} \times F_{\text{pt}} + C_{\text{plant}} \times F_{\text{pt}} \quad (9)$$

where  $PEC_{\text{soil}}$  (mg kg<sup>-1</sup>), is the concentration of contaminant in soil,  $F_{\text{soil}}$ , (kg<sup>-1</sup> d<sup>-1</sup>) is the cow's consumption of soil  $C_{\text{pt}}$  is the concentration of the contaminant in the plant a fresh weight basis (mg kg<sup>-1</sup>) and  $F_{\text{pt}}$ , (kg<sup>-1</sup> d<sup>-1</sup>) is the cow's consumption of the forage. Daily intake of a contaminant by a cow is proportional to the amount of forage ingested and the degree of contamination of the particular forage (Chitescu et al., 2014). Chitescu et al. (2014) proposed a value of 0.1 kg d<sup>-1</sup>, for cows consumption of soil, while Duarte-Davidson and Jones (1996) proposed that a cow consumes 0.9 kg d<sup>-1</sup> of soil. To account for the uncertainty, a uniform distribution (min 0.1, max 0.9) was assigned (Table 5). The cow's consumption of forage is between 12 and 18 kg d<sup>-1</sup> dry matter (Mc Gilloway & Mayne, 1996), and it was assumed that dairy and beef cows consumed the same amount. Therefore a uniform distribution was also assigned to account for variability and uncertainty. This model also takes into account the consumption of the contaminant that remained on the swards of grass following biosolid

**Table 4**  
Model inputs, distributions and outputs for  $PEC_{\text{porewater}}$  and concentration of contaminant in external water.

$PEC_{\text{porewater}}$				
	$F_{oc}$	Fraction of soil organic matter	Uniform (min 2, max 7)	%
	$K_{oc}$	Organic carbon-soil sorption coefficient	Triangular (contaminant specific, Table 3)	L kg <sup>-1</sup>
Concentration of contaminant in external water				
Output	$PEC_{\text{porewater}}$	Concentration of contaminant in external water	$PEC_{\text{soil}} / (F_{oc} \times K_{oc})$	mg kg <sup>-1</sup>
	$\alpha_{\text{pt}}$	Quasi-equilibrium factor	0.1	-
	$f_{\text{pw}}$	Weight fraction of water	89	%
	$f_{\text{ch}}$	Sum of carbohydrates, cellulose and proteins in plant	10.2	%
	$K_{ow}$	Octanol-water Partition coefficient	Triangular (contaminant specific, Table 3)	log
	$K_{ch}$	Partition coefficient for carbohydrate fraction of the plant	$1.23 \times K_{ow} - 2.42$	-
	$f_{\text{lip}}$	Weight fraction of lipids	97	%
	$k_{\text{lip}}$	Partition coefficient for lipid fraction of plant	$K_{ow}$	log
Output	$C_{\text{pt}}$	Concentration of contaminant in plant	$C_{\text{pt}} = \alpha_{\text{pt}} \times PEC_{\text{porewater}} \times [f_{\text{pw}} + f_{\text{ch}} \times K_{\text{ch}} + f_{\text{lip}} \times K_{\text{lip}}]$	mg kg <sup>-1</sup>

**Table 5**  
Model inputs, distributions and outputs for daily intake rate.

Daily intake rate				
	$F_{soil}$	Cow's consumption of soil	Uniform (min 0.1, max 0.9)	$kg\ d^{-1}$
	$F_{pt}$	Cow's consumption of forage	Uniform (min 12, max 18)	$kg\ d^{-1}$
Output	DI	Daily intake rate	$PEC_{soil} \times F_{soil} + C_{pt} \times F_{pt} + C_{plant} \times F_{pt}$	$mg\ d^{-1}$

application and dissipation rates.

### 2.3. The bio-transfer factor

Models that predict chemical transfer into beef and milk due to cattle ingestion of contaminated vegetation (e.g. silage or forage) often use a bio-transfer factor (BTF). The BTF is the ratio of the concentration in either beef or milk to the chemical intake rate in mass of chemical per day (USEPA, 2005). Travis and Arms (1988) developed a linear regression analysis of the log BTF for meat/milk and log  $K_{ow}$ . They compiled data from a review of literature resources to derive BTF's for a series of approximately 40 chemicals bio-transfer factors for organic chemicals in beef and milk are directly proportional to the octanol-water partition coefficient. Application of the equation requires that the user knows the log  $K_{ow}$  of the contaminant to estimate a BTF. Eqs. (10) and (11) show the BTF's for chemical in beef and milk, respectively as follows:

$$\text{LogBTFb [mg kg}^{-1}\text{/mg d}^{-1}] = -7.735 + 1.033 \times \log k_{ow} \quad (10)$$

$$\text{LogBTFm [mg kg}^{-1}\text{/mg d}^{-1}] = -8.056 + 0.992 \times \log K_{ow} \quad (11)$$

where measured concentrations of contaminants in beef or milk fat are converted to a fresh meat or whole milk basis.

TCS and TCC residue concentrations in beef and milk are calculated by:

$$C_{m/b} = \text{BTF (b, m)} \times \text{DI} \times \text{FC}_{(\text{milk, beef})} \quad (12)$$

where  $C_m$  and  $C_b$  is the TCS and TCC residue concentrations in beef and milk ( $mg\ d^{-1}$ );  $FC_{milk}$  and  $FC_{beef}$  is the average fat content of milk and beef. The average fat content of milk ( $FC_{milk}$ ) as reported by the Irish Cooperative Organisation Society (ICOS) (2009) is 3.7%. Chitescu et al. (2014) used a value of 4%. To account for uncertainty in the data, a uniform distribution was assigned (Table 6). The average fat content in beef ( $FC_{beef}$ ) tissue can range widely from 7.5% to over 27% (Hendriks, Smitkova, & Huijbregts, 2007). A uniform distribution was used to account for uncertainty. It was assumed that the antimicrobials did not have time to interfere with the cow's rumen flora as milking and slaughter of the cows took place within a day of eating the contaminated grass.

### 2.4. Human exposure

The amount of contaminant that may be ingested by humans through drinking milk and eating beef meat each day was estimated by:

**Table 6**  
Bio-transfer factor and residue for milk and beef.

Bio-transfer factor				
	BTFb	Bio-transfer factor beef	$\text{Log BTF} = -7735 + 1.033 \log k_{ow}$	$[\text{mg kg}^{-1}\text{/mg d}^{-1}]$
	BTFm	Bio-transfer factor milk	$\text{Log BTF} = -8.056 + 0.992 \log K_{ow}$	$[\text{mg kg}^{-1}\text{/mg d}^{-1}]$
Residue in milk and beef				
	$FC_{milk}$	Average fat content of milk	Uniform (min 3.7, max 4)	%
	$FC_{beef}$	Average fat content of beef	Uniform (min 7.5, max 27)	%
Output	$C_{milk}$	residue in milk	$\text{BTFm} \times \text{DI} \times \text{FC}_{milk}$	$mg\ d^{-1}$
Output	$C_{beef}$	residue in beef	$\text{BTFb} \times \text{DI} \times \text{FC}_{beef}$	$mg\ d^{-1}$

$$\text{HE} = C_{\text{milk/beef}} \times M_c / \text{bw} \quad (13)$$

where HE is human exposure ( $mg\ kg^{-1}\ bw\ d^{-1}$ );  $M_c$  is the consumption of milk or beef a day, and bw is the body weight of the individual. The consumption of milk and beef was based on several studies conducted by The Irish Universities Nutrition Alliance (IUNA). The National pre-school Nutrition Survey investigated the habitual food and drink consumption, health and lifestyle characteristics and assessed the body weight status in 500 pre-school children aged 1–4 years and living in the Republic of Ireland between 2010 and 2011. The National Children's Food Survey (2003–2004) assessed the consumption and body weights of 594 children aged 5–12 years (IUNA, 2005). The National Teens' Food Survey (2005–2006) investigated habitual food and drink consumption and health and lifestyle characteristics in 441 teenagers aged 13–17 years from the Republic of Ireland. The National Adult Nutrition Survey (2011) assessed the consumption and body weights of 1500 Irish consumers (IUNA, 2011). A log normal distribution was used to model the uncertainty regarding the intake of milk and beef. A summary of all human exposure model inputs are provided in Tables 7 and 8.

### 2.5. Acceptable daily intake

The acceptable daily intake (ADI) procedure has been used to calculate permissible chronic exposure levels for humans based on non-carcinogenic effects. The ADI is the amount of contaminant a human can be exposed to each day over a long time (usually lifetime) without suffering harmful effects. It is determined by applying safety factors (to account for uncertainty in the data) to the highest dose in human or animal studies which has been demonstrated not to cause adverse effects (NOAEL) (EC, 2003b). In determining the ADI the no observed adverse effects level (NOAEL) is divided by a safety factor in order to provide a margin of safety for allowable human exposure. A safety factor of 300 was applied in accordance with the European Commission Health and Consumer Protection Directorate-General (2005) and is composed of three factors; 10 is for intra-species variation, 10 is for inter-species variation and 3 is for a limited database of studies (Prosser & Sibley, 2015).

$$\text{ADI} = \text{NOAEL} / 10 \times 10 \times 3 \quad (14)$$

A NOAEL value of  $25\ mg\ kg^{-1}\ bw\ d^{-1}$  for TCS and TCC were obtained from Prosser and Sibley (2015) and was based on a sub chronic 90 day oral toxicity study with mice and a 2 year oral toxicity study with rats, respectively. A NOAEL of  $50\ mg\ kg^{-1}\ bw\ d^{-1}$  for TCC was obtained from the USEPA (2008) and was based on the reproductive

**Table 7**

Mean consumption and standard deviation of milk and beef for individual age groups.

Age group	Pre-school (1–4 yr) n = 500	Children (5–12 yr) n = 594	Teens (13–17 yr) n = 441	Adults (18–65 yr) n = 1274	Elderly (> 65) n = 226
Body weight (kg)	15.2 ± 1.95	33 ± 11.3	59.8 ± 11	78 ± 16.5	74.6 ± 13.9
Milk consumption (g d <sup>-1</sup> )	220 ± 193	9 ± 13	10 ± 14	13 ± 19	17 ± 19
Beef consumption (g d <sup>-1</sup> )	2 ± 5	5 ± 11	30 ± 44	19 ± 31	16 ± 27

toxicity of Sprague-Dawley rats over an 80 day period. The Scientific Committee on Consumer Products (SCCP) (EC, 2008) proposes a NOAEL of 12 mg kg<sup>-1</sup> bw d<sup>-1</sup> for TCS based on rat haematotoxicity studies as the critical effect level against which human exposure to TCS is compared. Rodricks, Swenberg, Borzelleca, Marronpot, and Shipp (2010) considered over 50 health endpoints and has developed a lower bound benchmark dose level of 47 mg kg<sup>-1</sup> bw d<sup>-1</sup>. To account for variability and uncertainty in the data, a uniform distribution was assigned for both TCS (min 12, max 47 mg kg<sup>-1</sup> bw d<sup>-1</sup>) and TCC (min 25, max 50 mg kg<sup>-1</sup> bw d<sup>-1</sup>) (Table 8).

Risk characterisation was quantified for potential non-carcinogenic risks, reflected for the hazard quotient (HQ) – the ratio of the potential exposure to a substance and the level at which no adverse effects are expected (the threshold toxicity reference value (RfD)). A HQ value < 0.01 indicates no existing risk, 0.1–1.0 risk is low, 1.1–10 risk is moderate and > 10 risk is high (Lemly, 1996). The reference dose value (RfD) (mg kg<sup>-1</sup> bw d<sup>-1</sup>) was calculated according to;

$$RfD = NOAEL/UF \times MF \quad (15)$$

where UF is one or more uncertainty factors and MF is a modifying factor based on professional judgement. Because the NOAEL is based on animals and of subchronic duration, the USEPA (2015) recommend a UF of 1000 and an MF of 0.8. The HQ values were calculated by dividing the exposure levels by the reference dose (RfD). The HQ for non-carcinogenic risk was calculated according to;

$$HQ = HE/RfD \quad (16)$$

## 2.6. Sensitivity analysis

Sensitivity analysis assesses how the model predictions are dependent on variability and uncertainty in the model's inputs. The input parameters were assembled in a spreadsheet in Microsoft Excel 2010 with the add-on package @Risk (version 6.0, Palisade Corporation, New York, USA), and the simulation was performed using Monte Carlo sampling with 10,000 iterations.

## 3. Results and discussion

The environmental fate of the antimicrobials triclosan and

**Table 8**

Model inputs, distributions and outputs for human exposure and hazard quotient.

Human exposure				
	bw	Body weight	Normal (Table 7)	kg
	Milk <sub>consum</sub>	Milk consumption	Lognormal (Table 7)	kg d <sup>-1</sup>
Output	HE <sub>milk</sub>	Human exposure milk	C <sub>milk</sub> × Milk <sub>consum</sub> / bw	mg kg <sup>-1</sup> bw d <sup>-1</sup>
	Beef <sub>consum</sub>	Beef consumption	Lognormal (Table 7)	kg d <sup>-1</sup>
Output	HE <sub>beef</sub>	Human exposure beef	C <sub>beef</sub> × beef <sub>consum</sub> / bw	mg kg <sup>-1</sup> bw d <sup>-1</sup>
	NOAEL	No observed adverse effects level	(TCS - min 12, max 47) (TCC - min 25, max 50)	mg kg <sup>-1</sup> bw d <sup>-1</sup>
	SF	Safety factor	10 × 10 × 3	–
	ADI	Acceptable daily intake	NOAEL/safety factor	mg kg <sup>-1</sup> bw d <sup>-1</sup>
	UF	Uncertainty factor	1000	–
	MF	Modifying factor	0.8	–
	RfD	Reference dose	NOAEL/UF × MF	mg kg <sup>-1</sup> bw d <sup>-1</sup>
Output	HQ	Hazard quotient	HE/RfD	–

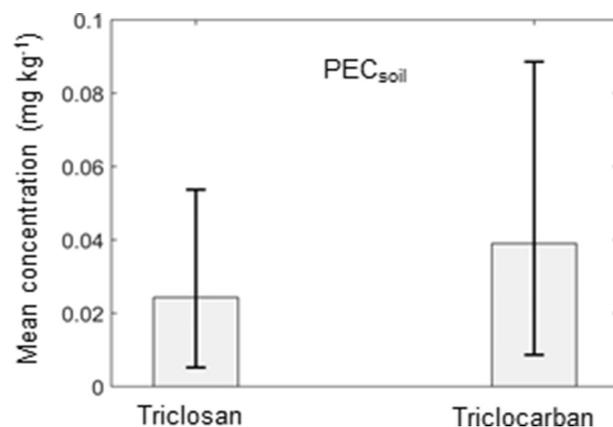


Fig. 2. Mean concentrations of TCS and TCC in soil following a single biosolid application (PEC<sub>soil</sub>) (mg kg<sup>-1</sup>). Error bars denote 5th and 95th percentiles.

triclocarban were modelled from biosolid application to plant uptake and bio-transfer to animal tissue with subsequent human consumption of milk and beef. The model resulted in several output distributions which include the PEC<sub>soil</sub>, Concentration on grass (C<sub>plant</sub>), Concentration in plant tissue (C<sub>pt</sub>), daily intake of contaminant (DI), and subsequent human exposure (HE) through consumption of beef and milk and the acceptable daily dose based on NOAEL values.

The results for the PEC<sub>soil</sub> show that TCC had a greater concentration in biosolids compared to TCS (mean values 3.90 × 10<sup>-2</sup> mg kg<sup>-1</sup>, 5th and 95th percentile values 8.63 × 10<sup>-3</sup> and 8.86 × 10<sup>-2</sup> for TCC and 2.43 × 10<sup>-2</sup> mg kg<sup>-1</sup>, 5th and 95th percentile values 5.19 × 10<sup>-3</sup> and 5.37 × 10<sup>-2</sup> for TCS) (Fig. 2). TCS and TCC have similar chemical properties. Both compounds are polychlorinated aromatic compounds which suggest significant resistance to biodegradation and bio-transformation (Halden & Paull, 2005); however, concentrations detected in biosolids may differ. This is in agreement with previous studies investigating the degradation potential of TCS and TCC which indicate that TCC is more persistent in the environment. Cha and Cupples (2010) reported that TCC was more persistent than TCS based on concentrations measured in the soil and the greater half-life values for TCC in aerobic and anaerobic conditions.

The results for concentration of contaminant remaining on the plant ( $C_{\text{plant}}$ ) show that TCS had the greater concentration remaining (mean value  $9.47 \times 10^{-10} \text{ mg kg}^{-1}$ , 5th and 95th percentile values  $5.54 \times 10^{-18}$  and  $3.13 \times 10^{-9}$ , respectively), compared to TCC (mean value  $7.14 \times 10^{-12} \text{ mg kg}^{-1}$ , 5th and 95th percentile values  $8.28 \times 10^{-28}$  and  $1.69 \times 10^{-11}$ , respectively).

Concentrations in plant tissue were only slightly greater for TCC with mean values  $1.40 \times 10^{-5} \text{ mg kg}^{-1}$ , 5th and 95th percentile values  $1.22 \times 10^{-6}$  and  $4.46 \times 10^{-5}$ , respectively, compared to TCS  $1.23 \times 10^{-5} \text{ mg kg}^{-1}$ , 5th and 95th percentile values  $6.45 \times 10^{-7}$  and  $5.03 \times 10^{-5}$ , respectively. These results are in agreement with a study conducted by García-Santiago et al. (2016) who demonstrated values of  $8.66 \times 10^{-5} \text{ mg kg}^{-1}$  of TCS in plant following a single application of biosolids. Plant uptake of TCS or TCC is a function of many variables which include initial concentration in biosolids, behaviour of contaminant in soil and plant type.

Factors such as the sorption coefficient ( $K_d$ ) and persistence (half-life) dictate availability and transport potential of contaminants in soil. Fu et al. (2016) performed a simple correlation test between  $K_d$  and plant uptake for TCS and TCC. A significant negative relationship was found between plant uptake and  $K_d$  for TCS ( $r^2 = 0.40\text{--}0.65$ ,  $p < 0.05$ ) or TCC ( $r^2 = 0.21\text{--}0.74$ ,  $p < 0.05$ ). This suggests that sorption played a dominant role in the inhibition of biosolids on plant uptake of these contaminants. Similarly, the authors also found that there was a poor relationship between the half-life and plant uptake of TCS ( $r^2 = 0.007\text{--}0.2$ ,  $p < 0.05$ ) or TCC ( $r^2 = 0.007\text{--}0.51$ ,  $p < 0.05$ ), implying that persistence alone did not impact a discernable effect on plant uptake of the contaminants.

The daily intake results of TCS and TCC by cows show that TCC had a greater intake rate, mean values  $1.97 \times 10^{-2} \text{ mg d}^{-1}$ , 5th and 95th percentile values  $2.60 \times 10^{-3}$  and  $5.49 \times 10^{-2}$ , respectively, compared to TCS with  $1.23 \times 10^{-2} \text{ mg d}^{-1}$ , 5th and 95th percentile values  $1.78 \times 10^{-3}$  and  $3.35 \times 10^{-2}$ , respectively (Fig. 3). Variability in soil and feed concentrations were included to account for uncertainty in the data. The concentration of contaminant on the plant was also included. Depending on the grazing season, concentrations of the contaminant in soil may vary. Concentrations of TCC were greater than TCS in the soil, therefore it was expected that there would be greater concentrations of TCC in the consumption of soil. The concentration in the feed (silage or forage) is dominated by uptake factors previously mentioned (sorption and persistence).

Predicted mean residue concentrations of TCS and TCC in beef show that concentrations of TCC were greater than TCS in beef (mean value  $1.47 \times 10^{-4} \text{ mg kg}^{-1}$ , 5th and 95th percentile values  $3.43 \times 10^{-8}$  and  $8.03 \times 10^{-4}$ , respectively and mean value  $2.62 \times 10^{-6} \text{ mg kg}^{-1}$ , 5th and 95th percentile values  $2.97 \times 10^{-7}$  and  $7.91 \times 10^{-6}$ , respectively) (Fig. 4). Mean residue concentrations in milk show that TCC had a higher concentration in milk than TCS (mean value  $8.06 \times 10^{-6} \text{ mg kg}^{-1}$ , 5th and 95th percentile values  $3.04 \times 10^{-9}$  and  $4.43 \times 10^{-5}$ , respectively and mean value  $1.81 \times 10^{-7} \text{ mg kg}^{-1}$ , 5th and 95th percentile values  $2.42 \times 10^{-8}$  and  $5.13 \times 10^{-7}$ , respectively) (Fig. 4). The hydrophilicities of TCC and TCS ( $\log k_{ow}$  4.9

and 4.6, respectively) indicate the potential for bioaccumulation. It has been suggested that compounds with high  $\log K_{ow}$  values and low water solubilities are the contaminants that have the greatest potential to accumulate in animal tissues (Duarte-Davidson & Jones, 1996). Contaminants with a higher half-life  $> 36 \text{ d}^{-1}$  combined with a higher  $\log K_{ow}$  value  $> 4.5$  have been associated with potential animal soil ingestion (Duarte-Davidson & Jones, 1996). Studies have shown that TCC and TCS can bioaccumulate in earthworms (Kinney et al., 2008), TCS in sheep placenta (James, Li, Summerlot, Rowland-Faux, & Wood, 2010) and humans (Adolfsson-Erici, Petterson, Parkkonen, & Sturve, 2002). TCS has been in human breast milk (Allmyr et al., 2006). Bioaccumulation of TCS and TCC occurs in humans but to a much lesser extent for example sheep and earthworms due to well-known detoxification reactions resulting in the rapid elimination of parental TCS and TCC (Halden, 2014).

Fig. 5 shows the modelled results for mean human exposure to TCS and TCC via beef. The teen group show the greatest risk of exposure to TCC levels in beef consumed (mean value  $7.41 \times 10^{-8} \text{ mg kg}^{-1} \text{ bw d}^{-1}$ ).

Fig. 6 shows the modelled results for mean human exposure to TCS and TCC via milk. The infant group show the greatest risk of exposure to TCC levels in milk consumed (mean value  $1.14 \times 10^{-7} \text{ mg kg}^{-1} \text{ bw d}^{-1}$ ).

None of the human exposure values exceeded the ADI (mean ADI values of  $0.058 \text{ mg kg}^{-1} \text{ bw d}^{-1}$  for TCS and  $0.1 \text{ mg kg}^{-1} \text{ bw d}^{-1}$  for TCC, respectively). Prosser et al. (2014) estimated ADI values of  $0.083 \text{ mg kg}^{-1} \text{ bw d}^{-1}$  for TCS and TCC based on a NOAEL value of  $25 \text{ mg kg}^{-1} \text{ bw d}^{-1}$  and an uncertainty factor of 300. Blanset, Zhang, and Robson (2007) estimated the ADI for TCS at  $0.05 \text{ mg kg}^{-1} \text{ bw d}^{-1}$  and concluded that, based on TCS levels typically measured in drinking water, the risk to human health is minimal. The European Union Health and Consumer Protection Directorate-General have set an ADI of  $0.8 \text{ mg kg}^{-1} \text{ bw d}^{-1}$  for TCC based on a 2 year repeated-dose toxicity test in rats. No ADI for TCS has been established yet. Chitescu et al. (2014) showed how 3 pharmaceuticals (sulfamethoxazole, ketoconazole and oxytetracycline) were transferred from contaminated soil through plant uptake and into the dairy food production chain. The results showed that the pharmaceuticals did contaminate the dairy cow's milk and meat due to the ingestion of contaminated grass by the cattle. However, human exposure results were below the ADI for all 3 pharmaceuticals and represented a minor risk. Aryal and Reinhold (2011) demonstrated how  $8.18 \text{ mg kg}^{-1}$  of TCC and  $0.18 \text{ mg kg}^{-1}$  of TCS detected in biosolids applied to land (3.25 dry tons per acre) could accumulate in plants. Detectable concentrations of TCS and TCC in pumpkin and zucchini plants was 2 orders less than exposure from using products (i.e. personal care products) that contained TCS and TCC and 35 times greater than exposure to drinking water.

All HQ results are below the threshold of risk ( $\text{HQ} < 0.01$ ). The results of the HQ show that of all the scenarios considered, TCC in milk and infant exposure had the highest value (mean HQ value  $3.9 \times 10^{-6}$  and 95th percentile value  $1.9 \times 10^{-5}$ ), while, TCC in beef and teen exposure had the highest value (mean HQ value  $2.40 \times 10^{-6}$  and 95th

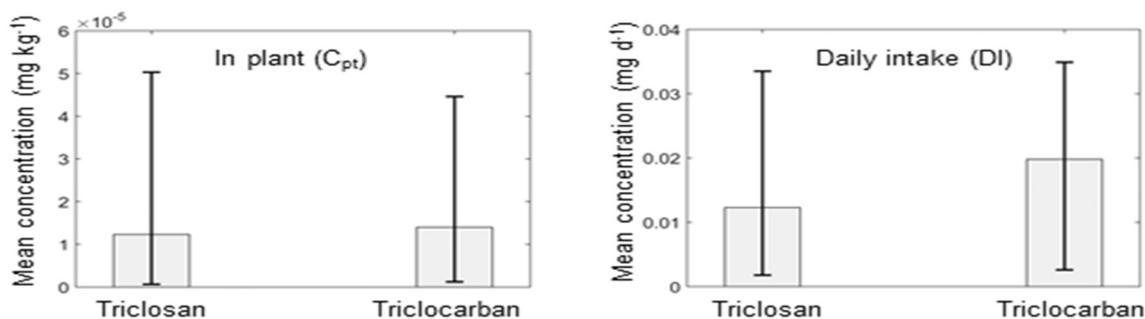


Fig. 3. Mean concentrations of TCS and TCC in plant tissue ( $\text{mg kg}^{-1}$ ) and daily cow intake ( $\text{mg d}^{-1}$ ). Error bars denote 5th and 95th percentiles.

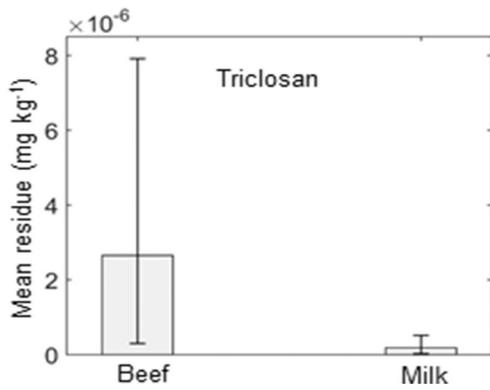


Fig. 4. Mean residue concentrations of TCS and TCC in milk and beef ( $\text{mg kg}^{-1}$ ). Error bars denote 5th and 95th percentiles.

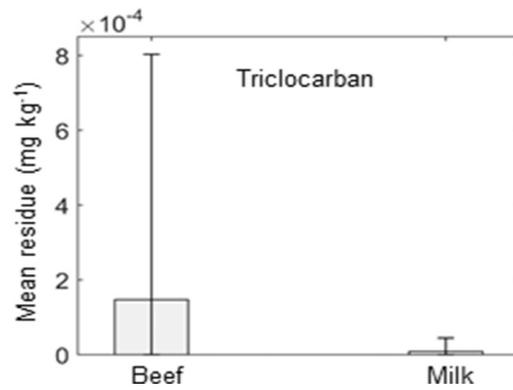


Fig. 5. Mean human exposure of TCS and TCC via beef consumption ( $\text{mg kg}^{-1} \text{bw d}^{-1}$ ).

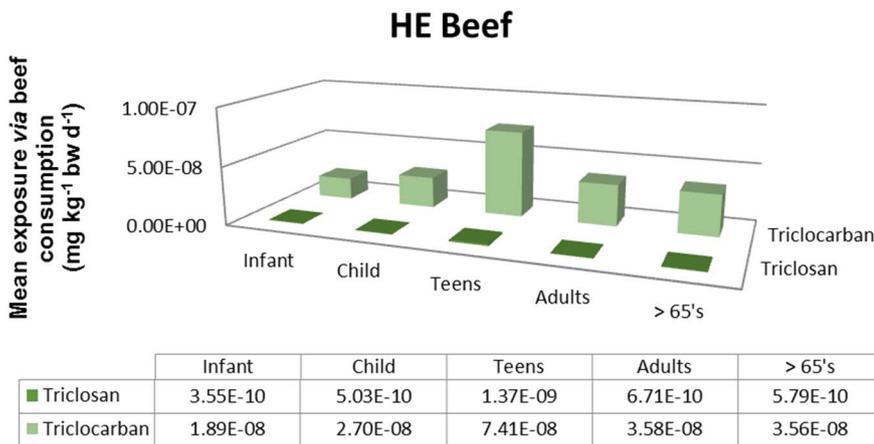
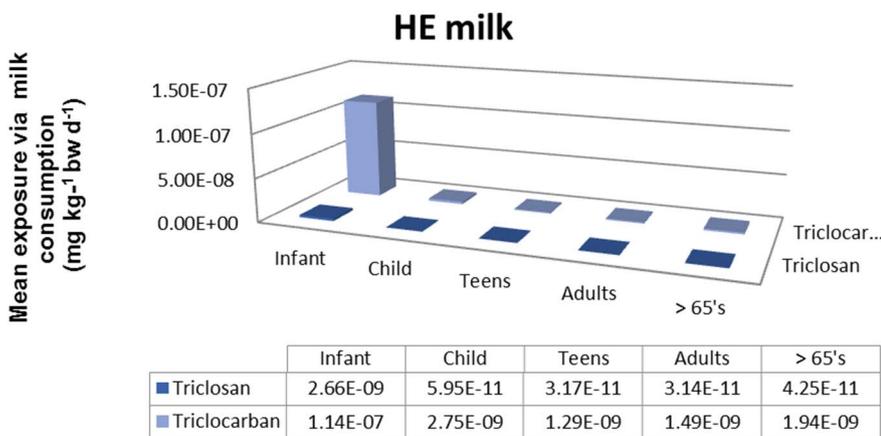


Fig. 6. Mean human exposure of TCS and TCC via milk consumption ( $\text{mg kg}^{-1} \text{bw d}^{-1}$ ).



**Table 9**  
Hazard quotient results for TCS and TCC including 5th and 95th percentiles.

	Infant (1–4 yr)	Child (5–12 yr)	Teen (13–17 yr)	Adult (18–64 yr)	> 65 yr
Triclosan					
Milk consumption	1.3e <sup>-07</sup> (7.3e <sup>-09</sup> , 4.5e <sup>-07</sup> )	3.1e <sup>-09</sup> (7.3e <sup>-11</sup> , 1.0e <sup>-08</sup> )	1.5e <sup>-09</sup> (4.7e <sup>-11</sup> , 5.8e <sup>-09</sup> )	1.5e <sup>-09</sup> (4.4e <sup>-11</sup> , 5.8e <sup>-09</sup> )	2.0e <sup>-09</sup> (8.4e <sup>-11</sup> , 7.9e <sup>-09</sup> )
Beef consumption	1.7e <sup>-08</sup> (1.9e <sup>-10</sup> , 6.9e <sup>-08</sup> )	2.4e <sup>-08</sup> (2.7e <sup>-10</sup> , 9.0e <sup>-08</sup> )	6.8e <sup>-08</sup> (1.7e <sup>-09</sup> , 2.6e <sup>-07</sup> )	3.3e <sup>-08</sup> (7.0e <sup>-10</sup> , 1.3e <sup>-07</sup> )	2.7e <sup>-08</sup> (5.9e <sup>-10</sup> , 1.1e <sup>-07</sup> )
Triclocarban					
Milk consumption	3.9e <sup>-06</sup> (1.1e <sup>-09</sup> , 1.9e <sup>-05</sup> )	8.5e <sup>-08</sup> (1.2e <sup>-11</sup> , 3.5e <sup>-07</sup> )	4.6e <sup>-08</sup> (7.3 <sup>-12</sup> , 2.0e <sup>-07</sup> )	4.7e <sup>-08</sup> (7.4e <sup>-12</sup> , 2.0e <sup>-07</sup> )	6.4e <sup>-08</sup> (1.2e <sup>-11</sup> , 3.0e <sup>-07</sup> )
Beef consumption	6.8e <sup>-07</sup> (3.6e <sup>-11</sup> , 2.3e <sup>-06</sup> )	8.7e <sup>-07</sup> (4.8e <sup>-11</sup> , 3.0e <sup>-06</sup> )	2.4e <sup>-06</sup> (2.5e <sup>-10</sup> , 1.1e <sup>-05</sup> )	1.3e <sup>-06</sup> (1.1e <sup>-10</sup> , 5.0e <sup>-05</sup> )	1.2e <sup>-06</sup> (9.4e <sup>-11</sup> , 4.3e <sup>-06</sup> )

percentile value  $1.1 \times 10^{-5}$  (Table 9).

Prosser and Sibley (2015) reported that the HQ for triclosan in the root of radish plants (0.91  $\text{mg kg}^{-1}$ ) following amendments with biosolids (total application of 1084–1180  $\text{Mg ha}^{-1}$  over a 16 yr period)

was 0.2 for toddlers. Garcia-Santiago et al. (2016) studied the environmental fate and the risks of persistent cosmetics and pharmaceutical compounds following detection of TCS in sludge (5.89  $\text{mg kg}^{-1}$ ), the bio-transfer to meat and milk, crops, dermal and inhalation with

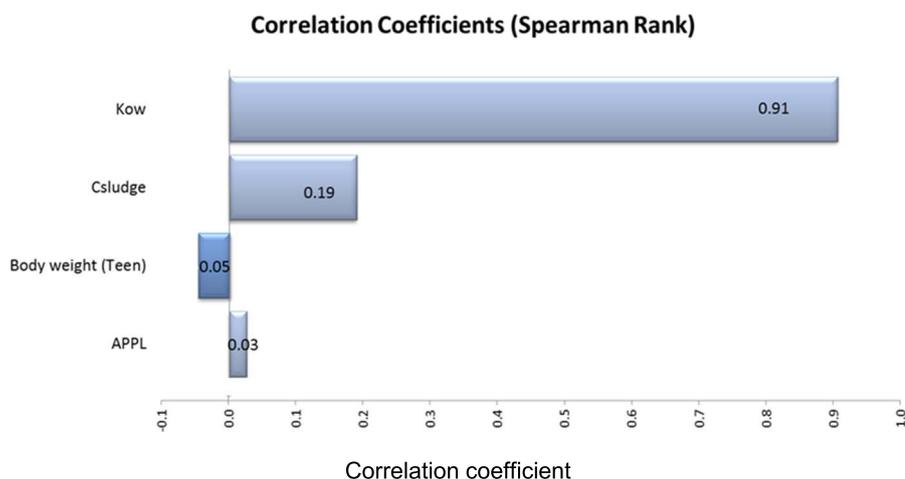


Fig. 7. Model input sensitivity analysis (Spearman Rank correlation coefficient) for TCC.

soil particles and human exposure. The study revealed hazard quotient values of 0.28 for TCS with a 95th percentile of 0.95 for root plant ingestion which could pose a potential hazard to human health. Snyder and O'Connor (2013) performed a two-tiered human health and ecological risk assessment of land applied biosolids-borne TCC. Hazard quotients were calculated to estimate risk for 14 exposure pathways identified in the USEPA Part 503 Biosolids Rule Risk Assessment assuming the 'worst case' scenarios (50 Mg biosolids ha<sup>-1</sup>, one time application, and incorporation to a depth of 15) and '100 year' (5 Mg biosolids ha<sup>-1</sup>, annual applications incorporated to a depth of 15 cm for 100 years). The majority of biosolids-borne TCC exposure pathways resulted in HQ < 1. Two pathways exceeded the HQ, the biosolid to predator pathway and biosolid to aquatic organism pathway. The study concluded that there was an unacceptable risk associated with TCC in land applied biosolids.

A sensitivity analysis based on the rank order correlation coefficient was conducted for TCC as this contaminant had the highest concentration in biosolids right through to consumption. Sensitivity analysis assesses how the model predictions are dependent on variability and uncertainty in the model's inputs. Results revealed that the  $K_{ow}$  was the most important parameter (correlation coefficient value 0.91) that affected the variance in model predictions (Fig. 7). This highlights the potential bioaccumulation of both contaminants. The high log  $K_{ow}$  values of 4.76 and 4.90 for TCS and TCC, respectively, suggest high sorption potential. The other parameter of importance was the initial concentration of the contaminants in sludge ( $C_{sludge}$ ) (correlation coefficient value 0.19) highlights detectable concentrations of TCS and TCC in biosolids post wastewater treatment and their continuum from land application through to the food chain. This is further heightened by the physical-chemical properties of the compounds such as sorption and persistence in sludge. Hence appropriate management of initial concentrations may lower overall human health risk.

#### 4. Conclusion

In this study detectable concentrations of TCS and TCC in biosolids estimated from the peer review literature were evaluated to assess their environmental fate in soil and plants, transfer into animal tissues and translocation into the food chain through the consumption of beef or milk. Introduction of these compounds to the environment is mainly through biosolid spreading as most of the TCS and TCC mass entering the WWTP is attached to the particles in the wastewater and most of the mass outgoing is contained in the biosolids. It is accepted that other routes of exposure may exist, however exposure through primary meat and dairy milk are likely to dominate. The  $PEC_{soil}$  showed that concentrations for TCC were greater than TCS; this was due to the overall concentration in the biosolids and greater half-life. This trend

continued throughout the model, however, it cannot be attributed to the initial concentrations in the biosolids alone, rather factors such as sorption and persistence dictates the behaviour of the contaminant in the soil. Both compounds are highly lipophilic and rarely found in soil solution, are preferably found in roots due to the contact with soil particles. This attribute also results in a higher bioaccumulation in beef and milk. Predicted human exposure to TCS and TCC through beef and milk showed that there was no appreciable risks as all values were well below the ADI. A hazard quotient (HQ) was also calculated and the results showed that there was no appreciable risk as all values were < 0.01. The study showed that infants and teens had the highest level of exposure through milk and beef, respectively, as the data obtained from consumer consumption studies show that these age categories typically consume more milk and beef. Sensitivity analysis showed that the  $K_{ow}$  and the initial concentration of the contaminants in biosolids as being the parameters of importance. Once introduced into the soil, concentrations of TCS and TCC may decrease over time as a result of a variety of dissipation processes. The study does highlight a route into which TCS and TCC may enter the food chain through the spreading of biosolids. The fact that they are highly lipophilic may hinder their progress along the food chain; however, their persistence in soil may introduce other consequences such as resistance to antibiotics. While exposure would appear to be small for humans, more research needs to be conducted to evaluate if the continued use of TCS and TCC may exacerbate the issue of antibiotic resistance, which may be another inadvertent consequence of the use of antimicrobials. Future work should continue the assessment to secondary products e.g. cheese butter and yogurts as these products typically have a higher fat content and are consumed in greater amounts by all age groups.

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#### References

- (OEHTA) (Office of Environmental Health Hazard Assessment) (2010). *Triclocarban. Potential designated chemical. May 24, 2010 meeting of Scientific Guidance Panel (SGP). Biomonitoring California* <http://oehha.ca.gov/multimedia/biomon/pdf/052410Triclocarban.pdf>, Accessed date: May 2016.
- Adolfsson-Erici, M., Petterson, M., Parkkonen, J., & Sturve, J. (2002). Triclosan, a commonly used bactericide found in human milk and in the aquatic environment in Sweden. *Chemosphere*, 46, 1485–1489.
- Agrinet (2015). Farm management software. <http://www.agrinet.ie/>, Accessed date: August 2016.
- Agyin-Birikorang, S., Miller, M., & O'Connor, G. A. (2010). Retention-release characteristics of triclocarban and triclosan in biosolids, soils, and biosolids-amended soils. *Environmental Toxicology and Chemistry*, 29, 1925–1933.

- Allmyr, M., Adolfsson-Erici, M., MCLachlan, M. S., & Sandborgh-Englund, G. (2006). Triclosan in plasma and milk from Swedish nursing mothers and their exposure via personal care products. *Science of the Total Environment*, 372, 87–93.
- Aryal, N., & Reinhold, D. M. (2011). Phytoaccumulation of antimicrobials from biosolids: Impacts on environmental fate and relevance to human exposure. *Water Research*, 45, 5545–5552.
- Banihashemi, B., & Droste, R. L. (2014). Sorption–desorption and biosorption of bisphenol A, triclosan, and 17 $\alpha$ -ethynylestradiol to sewage sludge. *Science of the Total Environment*, 487, 813–821.
- Barron, L., Havel, J., Purcell, M., Szpak, M., Kelleher, B., & Paull, B. (2009). Predicting sorption of pharmaceuticals and personal care products onto soil and digested sludge using artificial neural networks. *Analyst*, 134, 663–670.
- Blanset, D. L., Zhang, J., & Robson, M. G. (2007). Probabilistic estimates of lifetime daily doses from consumption of drinking water containing trace levels of N,N-diethyl-meta-toluamide (DEET), triclosan, or acetaminophen and the associated risk to human health. *Human and Ecological Risk Assessment: An International Journal*, 13, 615–631.
- BORD BIA (Irish Food Board) (2016). Factsheet on the Irish Agriculture and Food and Drink Sector. <http://www.bordbia.ie/industry/buyers/industryinfo/agri/pages/default.aspx>, Accessed date: July 2016.
- Boxall, A. B. A., Johnson, P., Smith, E. J., Sinclair, C. J., Stutt, E., & Levy, L. S. (2006). Uptake of veterinary medicines from soils into plants. *Journal of Agricultural and Food Chemistry*, 54, 2288–2297.
- Carter, L. J., Harris, E., Williams, M., Ryan, J. J., Kookana, R. S., & Boxall, A. B. A. (2014). Fate and uptake of pharmaceuticals in soil–plant systems. *Journal of Agricultural and Food Chemistry*, 62, 816–825.
- Cha, J., & Cupples, A. M. (2010). Triclocarban and triclosan biodegradation at field concentrations and the resulting leaching potentials in three agricultural soils. *Chemosphere*, 81, 494–499.
- Chalew, T. E., & Halden, R. U. (2009). Environmental exposure of aquatic and terrestrial biota to triclosan and triclocarban. *JAWRA Journal of the American Water Resources Association*, 45, 4–13.
- CHEMSPIDER. (Royal Society of Chemistry) (2015). Chemistry database. <http://www.chemspider.com/Chemical-Structure.5363.html>, Accessed date: February 2016.
- Chen, J., Ahn, K. C., Gee, N. A., Ahmed, M. I., Duleba, A. J., Zhao, L., ... Lasley, B. L. (2008). Triclocarban enhances testosterone action: A new type of endocrine disruptor? *Endocrinology*, 149, 1173–1179.
- Chen, X., Nielsen, J. L., Furgal, K., Liu, Y., Lolas, I. B., & Bester, K. (2011). Biodegradation of triclosan and formation of methyl-triclosan in activated sludge under aerobic conditions. *Chemosphere*, 84, 452–456.
- Chiou, C. T., Sheng, G., & Manes, M. (2001). A partition-limited model for the plant uptake of organic contaminants from soil and water. *Environmental Science & Technology*, 35, 1437–1444.
- Chitescu, C. L., Nicolau, A. I., Romkens, P., & Van der Fels-Klerx, H. J. (2014). Quantitative modelling to estimate the transfer of pharmaceuticals through the food production system. *Journal of Environmental Science and Health, Part B*, 49, 457–467.
- Chu, S., & Metcalfe, C. D. (2007). Simultaneous determination of triclocarban and triclosan in municipal biosolids by liquid chromatography tandem mass spectrometry. *Journal of Chromatography A*, 1164, 212–218.
- Clarke, B. O., & Smith, S. R. (2011). Review of ‘emerging’ organic contaminants in biosolids and assessment of international research priorities for the agricultural use of biosolids. *Environment International*, 37, 226–247.
- Clarke, R., Healy, M. G., Fenton, O., & Cummins, E. (2016). A quantitative risk ranking model to evaluate emerging organic contaminants in biosolid amended land and potential transport to drinking water. *Human and Ecological Risk Assessment: An International Journal*, 22, 958–990.
- Clarke, R., Peyton, D., Healy, M. G., Fenton, O., & Cummins, E. (2016). A quantitative risk assessment for metals in surface water following the application of biosolids to grassland. *Science of the Total Environment*, 566, 102–112.
- Clarke, R. M., & Cummins, E. (2014). Evaluation of “classic” and emerging contaminants resulting from the application of biosolids to agricultural lands: A review. *Human and Ecological Risk Assessment: An International Journal*, 21, 492–513.
- Coogan, M. A. (2007). *Bioaccumulation of Triclocarban, Triclosan, and Methyl-Triclosan in a North Texas Wastewater treatment plant Receiving Stream and Effects of Triclosan on Algal Lipid Synthesis*. USA: University of North Texas.
- Coogan, M. A., & Point, T. W. L. (2008). Snail bioaccumulation of triclocarban, triclosan, and methyltriclosan in a north Texas, USA, stream affected by wastewater treatment plant runoff. *Environmental Toxicology and Chemistry*, 27, 1788–1793.
- DAFM (Department of Agriculture, Fisheries and Marine) (2016). Fact sheet on Irish agriculture. <http://www.agriculture.gov.ie/media/migration/publications/2016/February2016Factsheet170216.pdf>, Accessed date: July 2016.
- Davis, E. F., Klosterhaus, S. L., & Stapleton, H. M. (2012). Measurement of flame retardants and triclosan in municipal sewage sludge and biosolids. *Environment International*, 40, 1–7.
- Dhillon, G. S., Kaur, S., Pulicharla, R., Brar, S. K., Cledon, M., Verma, M., & Surampalli, R. Y. (2015). Triclosan: current status, occurrence, environmental risks and bioaccumulation potential. *International Journal of Environmental Research and Public Health*, 12, 5657–5684.
- Duarte-Davidson, R., & Jones, K. C. (1996). Screening the environmental fate of organic contaminants in sewage sludge applied to agricultural soils: II. The potential for transfers to plants and grazing animals. *Science of the Total Environment*, 185, 59–70.
- EC (European Commission) (2003a). Technical guidance document on risk assessment in support of commission directive 93/67/EEC on risk assessment for new notified substances. *Commission regulation (EC) No 1488/94 on risk assessment for existing substances, Directive 98/8/EC of the European parliament and of the council concerning the placing of biocidal products on the market*[https://echa.europa.eu/documents/10162/16960216/tgdpart2\\_2ed\\_en.pdf](https://echa.europa.eu/documents/10162/16960216/tgdpart2_2ed_en.pdf), Accessed date: March 2016.
- EC (European Commission) (2003b). *Introduction to risk assessment*. [http://ec.europa.eu/health/ph\\_projects/2003/action3/docs/2003\\_3\\_09\\_a23\\_en.pdf](http://ec.europa.eu/health/ph_projects/2003/action3/docs/2003_3_09_a23_en.pdf), Accessed date: July 2016.
- EC (European Commission) (2005). Health and Consumer Protection Directorate-General, 2005. *Scientific Committee on Consumer Products - opinion on triclocarban*. Brussels, Belgium: European Commission Health and Consumer Protection Directorate-General.
- EC (European Commission) (2008). Scientific Committee on Consumer Products opinion on triclosan. [http://ec.europa.eu/health/ph\\_risk/committees/04\\_sccp/docs/sccp\\_o\\_166.pdf](http://ec.europa.eu/health/ph_risk/committees/04_sccp/docs/sccp_o_166.pdf), Accessed date: March 2015.
- EC (European Commission) (2016). Agriculture. [http://ec.europa.eu/ireland/news/key-eu-policy-areas/agriculture\\_en](http://ec.europa.eu/ireland/news/key-eu-policy-areas/agriculture_en), Accessed date: July 2016.
- Fehily Timoney and Company (1999). Codes of good practice for the use of biosolids in agriculture. <http://www.environ.ie/en/Publications/Environment/Water/FileDownload,17228,en.pdf>, Accessed date: June 2015.
- Fent, K., Weston, A. A., & Caminada, D. (2006). Ecotoxicology of human pharmaceuticals. *Aquatic Toxicology*, 76, 122–159.
- Fu, Q., Wu, X., Ye, Q., Ernst, F., & Gan, J. (2016). Biosolids inhibit bioavailability and plant uptake of triclosan and triclocarban. *Water Research*, 102, 117–124.
- Garcia-Santiago, X., Franco-Uria, A., Omil, F., & Lema, J. M. (2016). Risk assessment of persistent pharmaceuticals in biosolids: Dealing with uncertainty. *Journal of Hazardous Materials*, 302, 72–81.
- Gasperi, J., Geara, D., Lorgeoux, C., Bressy, A., Zedek, S., Rocher, V., ... Moilleron, R. (2014). First assessment of triclosan, triclocarban and paraben mass loads at a very large regional scale: Case of Paris conurbation (France). *Science of the Total Environment*, 493, 854–861.
- Goldstein, M., Shenker, M., & Chefetz, B. (2014). Insights into the uptake processes of wastewater-borne pharmaceuticals by vegetables. *Environmental Science & Technology*, 48, 5593–5600.
- Halden, R. U. (2014). On the need and speed of regulating triclosan and triclocarban in the United States. *Environmental Science & Technology*, 48, 3603–3611.
- Halden, R. U., & Paull, D. H. (2005). Co-occurrence of triclocarban and triclosan in US water resources. *Environmental Science & Technology*, 39, 1420–1426.
- Harris, S., Cormican, M., & Cummins, E. (2012). Antimicrobial residues and anti-microbial-resistant bacteria: Impact on the microbial environment and risk to human health. *Human and Ecological Risk Assessment*, 18(4), 767–809.
- Heidler, J., Sapkota, A., & Halden, R. U. (2006). Partitioning, persistence, and accumulation in digested sludge of the topical antiseptic triclocarban during wastewater treatment. *Environmental Science & Technology*, 40, 3634–3639.
- Hendriks, A. J., Smitkova, H., & Huijbregts, M. A. (2007). A new twist on an old regression: Transfer of chemicals to beef and milk in human and ecological risk assessment. *Chemosphere*, 70, 46–56.
- Higgins, C. P., Paesani, Z. J., Abbot Chalew, T. E., Halden, R. U., & Hundal, L. S. (2011). Persistence of triclocarban and triclosan in soils after land application of biosolids and bioaccumulation in *Eisenia foetida*. *Environmental Toxicology and Chemistry*, 30, 556–563.
- Hinther, A., Bromba, C. M., Wulff, J. E., & Helbing, C. C. (2011). Effects of triclocarban, triclosan, and methyl triclosan on thyroid hormone action and stress in frog and mammalian culture systems. *Environmental Science & Technology*, 45, 5395–5402.
- Holling, C. S., Bailey, J. L., Heuvel, B. V., & Kinney, C. A. (2012). Uptake of human pharmaceuticals and personal care products by cabbage (*Brassica campestris*) from fortified and biosolids-amended soils. *Journal of Environmental Monitoring*, 14, 3029–3036.
- Hung, H.-W., Lin, T.-F., & Chiou, C. T. (2010). Partition coefficients of organic contaminants with carbohydrates. *Environmental Science & Technology*, 44, 5430–5436.
- ICOS (Irish Cooperative Organisation Society) (2009). International milk production comparisons. <http://www.icos.ie/supply-chain/transport-quality/>, Accessed date: July 2016.
- IDIA (Irish Dairy Industries Association) (2016). Industry overview. <http://www.fdi.ie/idia>, Accessed date: July 2016.
- Igos, E., Benetto, E., Venditti, S., Kohler, C., Cornelissen, A., Moeller, R., & Biwer, A. (2012). Is it better to remove pharmaceuticals in decentralized or conventional wastewater treatment plants? A life cycle assessment comparison. *Science of the Total Environment*, 438, 533–540.
- Irish Water (2015). National wastewater sludge management plan. Asset strategy. <http://www.water.ie/about-us/project-and-plans/wastewater-sludge-management/NWSMP-report.pdf>, Accessed date: July 2016.
- IUNA (Irish Universities Nutrition Alliance) (2005–2006). IUNA.net, Accessed date: March 2016.
- IUNA (Irish Universities Nutrition Alliance) (2011). National Adult Nutrition Survey. IUNA.net, Accessed date: March 2016.
- James, M. O., Li, W., Summerlot, D. P., Rowland-Faux, L., & Wood, C. E. (2010). Triclosan is a potent inhibitor of estradiol and estrone sulfonation in sheep placenta. *Environment International*, 36, 942–949.
- Jelic, A., Gros, M., Petrovic, M., Ginebreda, A., & Barcelo, D. (2012). Occurrence and elimination of pharmaceuticals during conventional wastewater treatment. *Emerging and Priority Pollutants in Rivers*. Springer.
- King, M. K. (2010). *Evaluation of the developmental effects and bioaccumulation potential of triclosan and triclocarban using the South African clawed frog, Xenopus laevis*.
- Kinney, C. A., Furlong, E. T., Kolpin, D. W., Burkhardt, M. R., Zaugg, S. D., Werner, S. L., ... Benotti, M. J. (2008). Bioaccumulation of pharmaceuticals and other anthropogenic waste indicators in earthworms from agricultural soil amended with biosolid or swine manure. *Environmental Science & Technology*, 42, 1863–1870.
- Langdon, K. A., Warne, M. S. J., Smernik, R. J., Shareef, A., & Kookana, R. S. (2012). Field dissipation of 4-nonylphenol, 4-t-octylphenol, triclosan and bisphenol A following

- land application of biosolids. *Chemosphere*, 86, 1050–1058.
- Lemly, A. D. (1996). Evaluation of the hazard quotient method for risk assessment of selenium. *Ecotoxicology and Environmental Safety*, 35, 156–162.
- Li, H., Sheng, G., Chiou, C. T., & Xu, O. (2005). Relation of organic contaminant equilibrium sorption and kinetic uptake in plants. *Environmental Science & Technology*, 39, 4864–4870.
- Linders, J., Mensink, H., Stephenson, G., Wauchope, D., & Racke, K. (2000). Foliar interception and retention values after pesticide application. A proposal for standardized values for environmental risk assessment (technical report). *Pure and Applied Chemistry*, 72, 2199–2218.
- Lucid, J. D., Fenton, O., & Healy, M. G. (2013). Estimation of maximum biosolids and meat and bone meal application to a low P index soil and a method to test for nutrient and metal losses. *Water, Air, & Soil Pollution*, 224, 1464–1475.
- Lupton, S. J., Dearfield, K. L., Johnson, J. J., Wagner, S., & Huwe, J. K. (2015). Perfluorooctane sulfonate plasma half-life determination and long-term tissue distribution in beef cattle (*Bos taurus*). *Journal of Agricultural and Food Chemistry*, 63, 10988–10994.
- Mc Gilloway, D. A., & Mayne, C. S. (1996). The importance of grass availability for the high genetic merit dairy cow. In P. C. Garnsworthy, J. Wiseman, & W. Haresign (Eds.). *Recent Advances in Animal Nutrition* 1996 (pp. 135–169). Chp. 8.
- McClellan, K., & Halden, R. U. (2010). Pharmaceuticals and personal care products in archived U.S. biosolids from the 2001 EPA national sewage sludge survey. *Water Research*, 44, 658–668.
- Ogunyoku, T. A., & Young, T. M. (2014). Removal of triclocarban and triclosan during municipal biosolid production. *Water environment research: A research publication of the Water Environment Federation*, 86, 197–203.
- Prosser, R., & Sibley, P. (2015). Human health risk assessment of pharmaceuticals and personal care products in plant tissue due to biosolids and manure amendments, and wastewater irrigation. *Environment International*, 75, 223–233.
- Rodricks, J. V., Swenberg, J. A., Borzelleca, J. F., Marronpot, R. R., & Shipp, A. M. (2010). Triclosan: A critical review of the experimental data and development of margins of safety for consumer products. *Critical Reviews in Toxicology*, 40, 422–484.
- Rodrigues, S., Pereira, M., Duarte, A., & Romkens, P. (2012). Soil–plant–animal transfer models to improve soil protection guidelines: A case study from Portugal. *Environment International*, 39, 27–37.
- Rudel, H., Bohmer, W., Muller, M., Fliedner, A., Ricking, M., Teubner, D., & Schroter-Kermani, C. (2013). Retrospective study of triclosan and methyl-triclosan residues in fish and suspended particulate matter: Results from the German Environmental Specimen Bank. *Chemosphere*, 91, 1517–1524.
- Sabourin, L., Duenk, P., Bonte-Gelok, S., Payne, M., Lapen, D. R., & Topp, E. (2012). Uptake of pharmaceuticals, hormones and parabens into vegetables grown in soil fertilized with municipal biosolids. *Science of the Total Environment*, 431, 233–236.
- Snyder, E. H., & O'Connor, G. A. (2013). Risk assessment of land-applied biosolids-borne triclocarban (TCC). *Science of the Total Environment*, 442, 437–444.
- Snyder, E. H., O'Connor, G. A., & McAvoy, D. C. (2010). Fate of 14 C–triclocarban in biosolids-amended soils. *Science of the Total Environment*, 408, 2726–2732.
- Taylor-Smith, A. (2015). *Pharmaceutical compounds in land-applied sludge and plant uptake: A review*. North Carolina State University.
- Thompson, A., Griffin, P., Stuetz, R., & Cartmell, E. (2005). The fate and removal of triclosan during wastewater treatment. *Water Environment Research*, 77, 63–67.
- Travis, C. C., & Arms, A. D. (1988). Bioconcentration of organics in beef, milk, and vegetation. *Environmental Science & Technology*, 22, 271–274.
- USEPA (U.S. Environmental Protection Agency) (2005). *Methodology for predicting cattle biotransfer factors, EPA contract number 68-W-03-042*. Research Triangle Institute <https://archive.epa.gov/epawaste/hazard/tsd/td/web/pdf/btfrreportfull05.pdf>, Accessed date: 20 February 2006.
- USEPA (US Environmental Protection Agency) (2008). Screening-level hazard characterisation of high production volume chemicals. [http://www.epa.gov/hpvis/hazchar/101202\\_Triclocarban\\_HC\\_INTERIM\\_March%202008.pdf](http://www.epa.gov/hpvis/hazchar/101202_Triclocarban_HC_INTERIM_March%202008.pdf), Accessed date: March 2015.
- USEPA (US Environmental Protection Agency) (2009). *Targeted national sewage sludge survey sampling and analysis technical report*. Washington, D.C.2009 (EPA-822-R-08-014).
- USEPA (US Environmental Protection Agency) (2013). PBT PROFILER. <http://www.pbtprofiler.net/>, Accessed date: May 2016.
- USEPA (US Environmental Protection Agency) (2015). Reference Dose (RfD): Description and use in health risk assessments. <https://www.epa.gov/iris/reference-dose-rfd-description-and-use-health-risk-assessments>, Accessed date: August 2016.
- Vero, S. E., Antille, D. L., Lalor, S. T. J., & Holden, N. M. (2014). Field evaluation of soil moisture deficit thresholds for limits to trafficability with slurry spreading equipment on grassland. *Soil Use and Management*, 30, 69–77.
- Walters, E., McClellan, K., & Halden, R. U. (2010). Occurrence and loss over three years of 72 pharmaceuticals and personal care products from biosolids–soil mixtures in outdoor mesocosms. *Water Research*, 44, 6011–6020.
- WFD (Water framework Directive) (2012). Proposed EQS for Water Framework Directive Annex VIII substances: triclosan (for consultation). <http://www.wfduk.org/sites/default/files/Media/Triclosan%20-%20UKTAG.pdf>, Accessed date: May 2012.
- Wild, S., & Jones, K. (1992). Organic chemicals entering agricultural soils in sewage sludges: Screening for their potential to transfer to crop plants and livestock. *Science of the Total Environment*, 119, 85–119.
- Wu, C., Spongberg, A. L., & Witter, J. D. (2009). Adsorption and degradation of triclosan and triclocarban in soils and biosolids-amended soils. *Journal of Agricultural and Food Chemistry*, 57, 4900–4905.
- Wu, C., Spongberg, A. L., Witter, J. D., Fang, M., & Czajkowski, K. P. (2010). Uptake of pharmaceutical and personal care products by soybean plants from soils applied with biosolids and irrigated with contaminated water. *Environmental Science & Technology*, 44, 6157–6161.
- Wu, C., Spongberg, A. L., Witter, J. D., & Sridhar, B. B. M. (2012). Transfer of wastewater associated pharmaceuticals and personal care products to crop plants from biosolids treated soil. *Ecotoxicology and Environmental Safety*, 85, 104–109.
- Xia, K., Hundal, L. S., Kumar, K., Armbrust, K., Cox, A. E., & Granato, T. C. (2010). Triclocarban, triclosan, polybrominated diphenyl ethers, and 4-nonylphenol in biosolids and in soil receiving 33-year biosolids application. *Environmental Toxicology and Chemistry*, 29, 597–605.
- Ying, G.-G., Yu, X.-Y., & Kookana, R. S. (2007). Biological degradation of triclocarban and triclosan in a soil under aerobic and anaerobic conditions and comparison with environmental fate modelling. *Environmental Pollution*, 150, 300–330.