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6 **QUANTITATIVE RISK ASSESSMENT OF ANTIMICROBIALS IN THE FEED TO**
7 **FOOD CHAIN**

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9
10 **Rachel Clarke¹, Mark G Healy², Owen Fenton³ and Enda Cummins¹**

11
12 [Email: rachel.clarke.1@ucdconnect.ie]

13 [enda.cummins@ucd.ie]

14 ph. +353 7167476]

15 ¹School of Biosystems and Food Engineering, Agriculture and Food Science centre,
16 University College Dublin, Belfield, Dublin 4, Ireland

17 ²Civil Engineering, National University of Ireland, Galway, Co. Galway, Rep. of Ireland

18 ³Teagasc, Environment Research Centre, Johnstown Castle, Co. Wexford, Rep. of
19 Ireland

29 **Abstract**

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

55 **Keywords:** biosolids, contaminants, human exposure, risk

56

57 1. Introduction

58 The two most important farming sectors in Ireland are the milk and meat sectors,
59 accounting for approximately 69% of agricultural output (DAFM 2016). The value of
60 overall beef exports from Ireland was 2.27 billion in 2014 (EC 2016), while dairy exports
61 have grown to 3.1 billion in 2014 (IDIA 2016). There are currently 6.96 million cattle in
62 Ireland according to the June 2015 livestock survey (Bord bia 2016). The total land area
63 of Ireland is 6.9 million hectares of which 4.5 million hectares is used for agriculture
64 (DAFM 2016). Eighty one percent of the agricultural area is devoted to pasture, hay and
65 grass silage (3.6 million hectares) and 11% to rough grazing (Bord bia 2016). Under the
66 'Code of Good Practice for the use of biosolids in agriculture' (Fehily Timoney &
67 Company 1999) it states that there are constraints on grazing following application of
68 biosolids to agricultural land. '*Cattle should not be turned out onto pasture that has been*
69 *fertilised with biosolids until 3-6 weeks after the date of application*'. The interval
70 between application and commencement of grazing will depend on the level of
71 incorporation of biosolids into the soil (Fehily Timoney & Company 1999). In Ireland,
72 53,543 tonnes dry solids (tds) of biosolids are generated each year, of which 98% is
73 disposed to agricultural land. It has been predicted that this figure will grow to 96,442
74 tds/annum by 2040 (Irish Water 2016).

75 One of the main concerns for human health regarding the spreading of biosolids on
76 agricultural land is the potential uptake of contaminants into plants which may bio-
77 transfer into grazing animals that are subsequently consumed by humans. Studies have
78 shown that conventional wastewater treatment does not fully eliminate contaminants
79 such as pharmaceuticals (i.e. beta-blockers, carbamazepine, paracetamol and
80 diclofenac) (Jelić et al., 2012; Igos et al., 2012; Harris et al., 2012; Clarke and Cummins,
81 2014); therefore contaminants may still be present in the treated sludge. Biosolids are
82 rich in organic matter and may contain up to 38% organic carbon on a dry mass basis.
83 Therefore, repeated application of biosolids may greatly increase a soil's organic carbon
84 content (OC), leading to enhanced sorption or reduced chemical bioavailability (Fu et
85 al., 2016). On one hand, biosolid application is a direct point source for contaminants
86 into the environment (Clarke et al., 2016), on the other hand, increased organic matter

87 may inhibit plant uptake due to reduced bioavailability. A number of studies have
88 demonstrated the uptake of contaminants into plants (Boxall et al., 2006, Sabourin et
89 al., 2012, Prosser and Sibley, 2015). For example, Holling et al. (2012) showed how 3
90 of the 5 human pharmaceuticals (carbamazepine, sulfamethoxazole, salbutamol,
91 triclosan and trimethoprim) analysed were detected in the aerial parts and root tissue of
92 the cabbage plant with median measured concentrations of 317.6 ng g⁻¹ and 416.2 ng g⁻¹
93 for carbamazepine, 21.2 ng g⁻¹ and 187.6 ng g⁻¹ for salbutamol and 22.9 ng g⁻¹ and
94 1220.1 ng g⁻¹ for triclosan. Sabourin et al. (2012) evaluated the uptake of 118
95 pharmaceuticals, 17 hormones and 6 parabens in tomatoes, carrots, potatoes and
96 sweetcorn from soil fertilised with municipal biosolids at a regulated application rate.
97 Eight of the 141 analytes were detected in one or two crop replicates at concentrations
98 ranging from 0.33 to 6.25 ng g⁻¹ dry weight (dw), however the study showed that no
99 analytes were consistently detected above the detection limit in all treated plots. Carter
100 et al. (2014) also demonstrated how 5 of the 6 pharmaceuticals (carbamazepine,
101 diclofenac, fluoxetine, propranolol) and personal care product (triclosan) tested were
102 detected in the root tissue of the plant tissue over a 40 day growing period.

103 The main route of human exposure to many highly lipophilic contaminants is through
104 ingestion of contaminated agricultural products such as beef and milk (USEPA 2005).
105 Livestock can ingest contaminants from soil by grazing and/or feeding on harvested
106 forage. In countries where animals can graze all year round, average soil ingestion has
107 been estimated as 4.5% of the dry matter intake for sheep and 6% for cattle when
108 pasture was the only feed source (Duarte-Davidson and Jones, 1996). While there have
109 been many models developed to predict animal uptake, including relating bio-transfer
110 concentrations (BCF) in livestock to physio-chemical properties (Travis and Arms, 1988,
111 Rodrigues et al., 2012), there are significant knowledge gaps with regards to actual
112 contaminant concentrations in livestock following direct ingestion of grass from biosolid
113 amended agricultural land. Lupton et al. (2015) conducted a study to determine plasma
114 and tissue depletion kinetics in cattle. The cattle (2 steers and 4 heifers) were dosed
115 with perfluorooctane sulfate (PFOS) at 0.098 mg kg⁻¹ weight and 9.1 mg kg⁻¹,
116 respectively. Plasma depletion half-lives for steers and heifers were 120 ± 4.1 and 106
117 ± 23.1 days, respectively. Specific tissue depletion half-lives ranged from 36 to 385

118 days for intraperitoneal fat, back fat, muscle, liver, bone, and kidney. The results of the
119 experiment showed that PFOS in beef cattle had a sufficiently long depletion half-life to
120 permit accumulation in edible tissues.

121 The proportion of an organic or inorganic contaminant taken up by plant roots and its
122 translocation route within the plant depends on its physio-chemical properties (Goldstein
123 et al., 2014), the plant's physical characteristics and soil properties (Taylor-Smith,
124 2015). The log K_{ow} or log of the octanol water partitioning coefficient represents a
125 compound's propensity to partition into either polar or non-polar mediums (Fent et al.,
126 2006). Highly lipophilic contaminants characterised by high octanol water partitioning
127 coefficients ($K_{ow} > 3$ log unit) or low water solubilities, have a high tendency to be
128 absorbed by plant roots from water (Li et al., 2005). For example, Carter et al. (2014)
129 attributed the uptake of pharmaceuticals and a personal care product into radishes and
130 ryegrass to the physiochemical properties of the contaminants, including Henry's Law
131 constant, water solubility and octanol water partition coefficient. Wu et al. (2010) also
132 demonstrated how the K_{ow} predictions of contaminant behaviour in plants correlated
133 with the bioconcentration factor of the contaminants. Wild et al (1992) categorised non-
134 ionised organic contaminants with $\log K_{ow} > 4$ as having a high potential for retention in
135 plant roots. Thus, the octanol water partition coefficient (K_{ow}) has been suggested as a
136 reliable indicator of uptake behaviour (Goldstein et al., 2014). Various contaminants
137 bind strongly to sludge, soil and sediments, an attribute that relates to their persistence
138 in the environment. Lipophilic organic contaminants possess a greater tendency to
139 partition into plant root lipids than hydrophilic contaminants (Duarte-Davidson and
140 Jones, 1996). For instance, persistent organic pollutants (POPs) have a strong affinity
141 to absorb to the soil organic carbon (SOC) due to their hydrophobicity ($\log K_{ow} > 4$),
142 which makes them less mobile in soil (Sweetman et al., 2005), whereas, the natural
143 hormones 17β estradiol and estrone are highly soluble with a half-life estimated to be
144 1.5 - 29 days in soil (Carr et al., 2011). However, Mattina et al. (2003) reported that
145 zucchini (*Cucurbita pepo* L.) and spinach (*Spinacia oleracea*) can bioaccumulate soil
146 bound POPs. This was attributed to the physiological properties of the plant species.

147 Chemicals in soil enter plants primarily through the root system and the degree of
148 uptake from soil into root tissues appears to be proportional to the octanol-water
149 partition coefficient (K_{ow}).

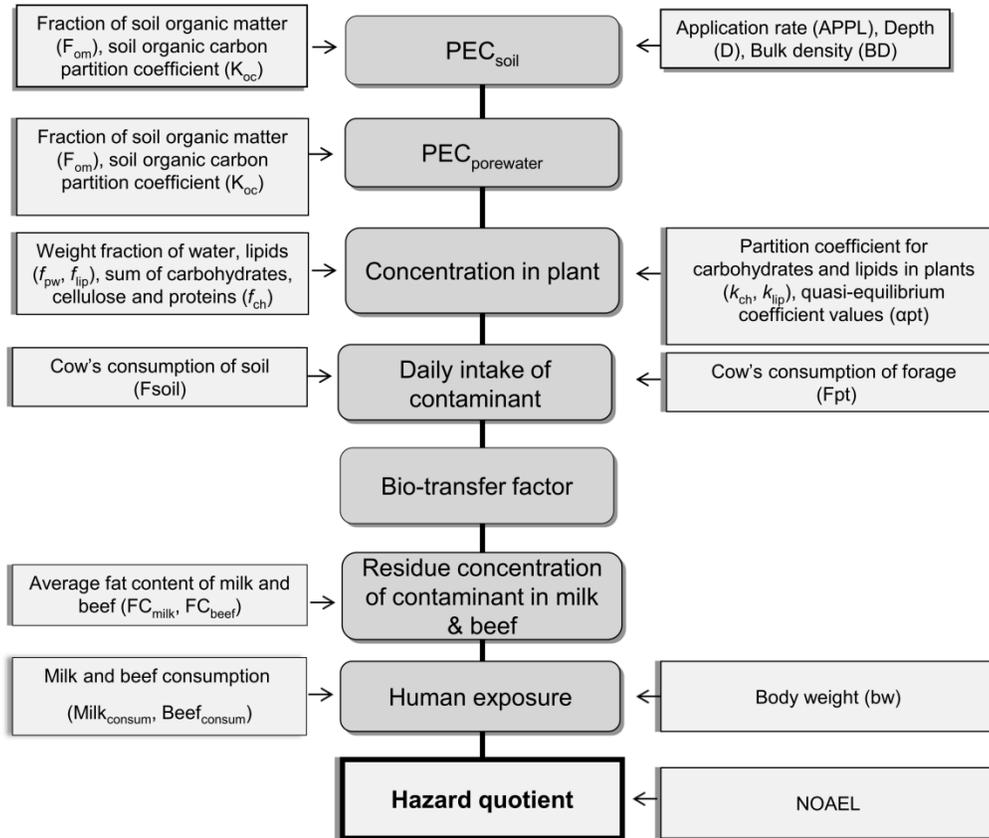
150 Since the 1960's antimicrobials triclosan (TCS) and triclocarban (TCC) have been in
151 use as antibacterial agents in many products such as toothpaste, soaps, creams, etc.
152 (Xia et al., 2010). Toxicological reports have shown that TCC has the potential to disrupt
153 excitation coupling in skeletal and cardiac muscles in humans (Clarke et al., 2016).
154 Studies suggest that TCS and TCC may persist in the sludge post wastewater
155 treatment. The US Environmental Protection Agency (2009) conducted a study on 84
156 WWTPs to analyse the sludge. TCC was detected in 100% of the samples at a
157 concentration range of 187 - 441,000 $\mu\text{g kg}^{-1}$, TCS was detected 94% of the time with a
158 concentration range of 430 - 133,000 $\mu\text{g kg}^{-1}$. Heidler et al. (2006) reported removal
159 efficiencies of TCS and TCC were 98 % and 97%, respectively. Ogunyoku and Young
160 (2014) studied removal efficiencies of conventional wastewater treatment on TCS and
161 TCC. Results show that TCS was more rapidly removed than TCC, indicating that TCS
162 was more readily bio-transformed than TCC.

163 Once introduced to the environment, TCS and TCC sorb to soils and sediment and are
164 not predicted to readily degrade (Aryal and Reinhold, 2011). Ying et al. (2007) reported
165 that TCS degraded faster than TCC by microbial processes in the soil under aerobic
166 conditions. However, the half-life in air was greater for TCS. Volatisation is not expected
167 to be a significant removal mechanism for TCC and TCS (Okunyoku and Young 2014).
168 Sorption (K_d) and persistence (measured as half-life, ($T_{1/2}$)) are considered the two
169 primary variables controlling the availability and hence offsite transport potential of
170 contaminants in soil (Fu et al., 2016). To describe the distribution of a chemical in soil,
171 the soil-water partition coefficient (K_d) is a suitable measure. The K_d is generally
172 proportional to the hydrophobicity of the compound and the amount of soil organic
173 matter. The experimental half-life of TCS and TCC in soil ranges from 87-231 and 18-58
174 days, respectively in aerobic soils with longer half-lives in anaerobic soils (Ying et al.,
175 2007). The half-life in air is estimated to be 1 d^{-1} for TCS and 0.75 d^{-1} for TCC (PBT
176 profiler 2012). Ying et al., (2007) reported half-lives in air of 0.66 d^{-1} for TCS and 0.75 d^{-1}

177 ¹ for TCC. TCS and TCC are both hydrophobic with log K_{ow} values of 4.76 and 4.90,
178 respectively (Dhillon et al., 2015). However, accumulation of these compounds has
179 been observed in plants (Aryal and Reinhold, 2011, Wu et al., 2012), animals (Coogan
180 and Point, 2008, Kinney et al., 2008, Higgins et al., 2011), humans (Allmyr et al., 2006)
181 and the potential of TCS and TCC as endocrine disruptors also shown (Chen et al.,
182 2008, Hinthner et al., 2011). Wu et al., (2012) demonstrated that after 60 days growth,
183 TCS and TCC had accumulated and translocated into above ground parts of the
184 soybean plant following application of biosolids and reclaimed waste water. Prosser et
185 al. (2015) reported the uptake of TCS and TCC in the edible portions of green pepper,
186 carrots, cucumber, tomato, radish and lettuce plants grown in biosolid amended land.
187 TCS was only detected in cucumber and radish up to 5.2 ng g⁻¹ dw, while TCC was
188 detected in carrot, green pepper, tomato and cucumber up to 5.7 ng g⁻¹ dw. It was
189 estimated that vegetable consumption represents less than 0.5 % of the acceptable
190 daily intake of TCS and TCC. Aryal and Reinhold (2011) measured concentrations of
191 TCS at approximately 20,000 and 40,000 ng g⁻¹ dw in the root and 8000 and 5000 ng g⁻¹
192 ¹ dw in the stem of pumpkin and zucchini plants, respectively despite a low
193 concentration of TCS (180 ng g⁻¹), however liquid biosolids were applied prior to
194 seeding and 8 weeks after seeding accounting for the high levels in the plants.

195 In this study, the aim was to develop a quantitative risk assessment model to estimate
196 the fate and translocation of antimicrobials (triclosan and triclocarban) into biosolid
197 receiving agricultural soils with transfer into animal forage and subsequent potential
198 transfer into the food production chain and potential human consumption/exposure
199 (Figure 1).

200



201

202 **Figure 1:** Flow diagram of inputs and outputs for the quantitative plant uptake and
 203 translocation into the food chain model.
 204

205 **2. Materials and methods**

206 **2.1 Predicted environmental concentration in soil**

207 The PEC_{soil} was estimated by developing a distribution of contaminant exposure based
 208 on the variability and uncertainty of the predicted environmental concentrations in
 209 biosolids. The concentration in the soil (C_{soil} ; $mg\ kg^{-1}$) immediately following a single
 210 biosolid application was calculated based on the concentration of the contaminant in the
 211 biosolids, application rate, crop intersection, mixing depth of soil and the soil bulk
 212 density following biosolids application according to the guidelines of the European
 213 Chemicals Bureau's Technical Guidance Document on Risk Assessment Part II
 214 (2003a).

215
$$C_{\text{soil}} = (C_{\text{sludge}} \times \text{APPL} \times (1 - \text{fint}/100)) / (D \times \text{BD}) \quad (1)$$

216 Where:

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

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

219 fint is the fraction intercepted by the crop (-)

220 D is the depth (m)

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

222 The peer reviewed literature was searched for Irish and European organic contaminant
223 concentrations in biosolids (Table 1) and probabilistic distributions were fitted to
224 characterise uncertainty/variability in the level of TCS and TCC in biosolids (Table 2).
225 Uncertainty regarding the application rate was represented using a triangular
226 distribution (minimum 300; mean 330; and maximum 520 g m^{-2}) (Table 3). The
227 application rate of biosolids was retrieved from Lucid et al. (2013). It was assumed that
228 the biosolids were spread on grassland.

229 The mixing depth (0.1 m) was obtained from the EU Technical Guidance Document on
230 Risk Assessment part II (2003a) and is representative of grassland as grassland is not
231 traditionally ploughed. The BD of soil (800 - 1000 kg/m^3) (uniform distribution) were
232 obtained from Vero et al. (2014), which is a typical range for the upper 100 mm of the
233 soil profile of Irish grasslands. It was assumed that the soil was poorly drained with OM
234 and BD ranges as stated. The fraction intercepted by the crop was based on tabular
235 interception fractions values as proposed by Linders et al., (2000) which were based on
236 field experiments found in the literature. The authors adopted the approach that
237 interception fraction plus the soil deposition fraction is unity ($\text{fint} + \text{Fsoil} = 1$). The study
238 focuses on interception rather than retention. Conceptually, it is assumed that both
239 interception and deposition on soil are instantaneous processes. Crop interception (fint)
240 was estimated to be a triangular distribution (minimum 0, most likely 10 and maximum
241 20%) assuming a worst-case scenario.

242

243 The degradation kinetics in soil was described using a first order reaction model. The
244 half-life (DT_{50}) of each contaminant was obtained from the peer reviewed literature and
245 shown in Table 3. To account for uncertainty and variability in the data, TCS and TCC
246 were assigned a uniform distribution. The dissipation rate constant 'k' is given by:

$$247 \quad k = \text{Ln}(2)/DT_{50\text{soil}} \quad (2)$$

248 Where DT_{50} is the half-life of the contaminant in soil.

249 A first order exponential decay model was used to estimate the degradation rate
250 constant 'k' (in day), DT_{50} and time 't' of TCS and TCC in soil.

$$251 \quad PEC_{\text{soil}} = C_{\text{soil}} \times e^{-kt} \quad (3)$$

252 Where PEC_{soil} is the concentration remaining in soil following dissipation. "t" is the time
253 the contaminant has in the soil prior to grazing. The Code of Good Practice for
254 Application of Biosolids on Agricultural Land states that cattle may not be turned out
255 onto grassland until at least 3-6 weeks following biosolid application. Therefore the
256 time 't' (in days) was assigned a uniform distribution (min 21, max 42) to allow for
257 constraints in allowing cattle to graze.

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

$$264 \quad C_{\text{applied}} = C_{\text{sludge}} \times \text{APPL} \times \textit{fint} \quad (4)$$

265 Where C_{applied} (mg m^2) is the concentration of biosolid remaining on the grass following
266 biosolid application.

267 When biosolids are applied to agricultural land, the field dissipation of the contaminants
268 contained within the biosolids is likely to be influenced by environmental conditions.
269 Variations in temperature and available moisture are likely to play an important role in
270 the dissipation of contaminants (Langdon et al., 2012). The half-life in air for both TCS

271 and TCC was obtained from the PBT Profiler (USEPA 2013) and Ying et al., (2007)
272 (Table 3). To account for variability and uncertainty in the data, a uniform distribution
273 (min 0.66, max 1) for TCS and (min 0.5, max 0.75) for TCC was assigned. A first order
274 exponential decay model was used to calculate the dissipation of the contaminants on
275 the swards of grass.

276 The same time 't' was used as above. It was assumed that 3 weeks had passed since
277 the land spreading of biosolids on agricultural grasslands. The amount of fresh grass in
278 kg per m² (P_d) was obtained from Agrinet Farm Management Software (2015) and it
279 was estimated that there was 18,000 kg of fresh grass per Ha. The overall
280 concentration of contaminant on grass was calculated according to equation 5.

$$281 \quad C_{\text{plant}} = C_{\text{applied}} \times e^{-kt} / P_d \quad (5)$$

282 Where C_{plant} (mg kg⁻¹) is the concentration remaining on the plant following dissipation.
283 P_d is the plant density (kg m²).

284 In the present study the approach developed by Chițescu et al. (2014) and Chiou et al.
285 (2001) is modified for use in Irish conditions (e.g. application rates, bulk density, cow's
286 consumption of forage and human consumption rates). The effective concentration of
287 contaminants available for plant uptake is the concentration of the contaminant in soil
288 interstitial (pore) water. Soil composition influences the concentration of the contaminant
289 in pore water, by its fraction of organic matter (F_{oc}) (Chițescu et al., 2014). The model is
290 expressed as:

$$291 \quad \text{PEC}_{\text{porewater}} = \text{PEC}_{\text{soil}} / (F_{\text{oc}} \times K_{\text{oc}}) \quad (6)$$

292
293 Where PEC_{porewater} is the contaminant concentration in the pore water (mg kg⁻¹). F_{oc} is
294 the fraction of organic matter content (F_{oc}) in the soil; and K_{oc} is the soil organic carbon-
295 water partitioning coefficient of the contaminants. K_{oc} is the organic carbon-soil sorption
296 coefficient (L kg⁻¹) (contaminant specific). Triangular distributions were used to model
297 K_{oc} uncertainty (Table 3). The fraction of organic matter content in the soil was obtained
298 from peer reviewed literature for F_{oc} in soil (2%, 5% and 7%) (Chalew and Halden
299 2009). To account for variability and uncertainty in the data, a uniform distribution (min

300 2%, max 7%) was assigned (Table 4). To convert the units from mg L⁻¹ to mg kg⁻¹, the
301 density of water was assumed.

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

$$305 \quad C_{pt} = \alpha_{pt} \times PEC_{porewater} \times [f_{pw} + f_{ch} \times K_{ch} + f_{lip} \times K_{lip}] \quad (7)$$

306 Where C_{pt} is the concentration of the contaminant in the plant on a fresh weight base
307 (mg kg⁻¹); f_{pw} , f_{lip} and f_{ch} are the weight fraction of, respectively, water, lipids and the sum
308 of carbohydrates, cellulose, and proteins in the plant; K_{lip} is the partition coefficient for
309 the lipids fraction of the plant assumed to be equal to the log K_{ow} ; K_{ch} is the partition
310 coefficient for the carbohydrate fraction of the plant, available according to K_{ow} . The
311 symbol α_{pt} is the quasi-equilibrium factor, defined as the ratio of the respective
312 concentration of the contaminant in plant water and external water. Thus, $\alpha_{pt} = 1$
313 denotes the state of equilibrium. $\alpha_{pt} < 1$ is a measure of the extent of approach to
314 equilibrium (Chiou et al., 2001). The quasi-equilibrium coefficient values are based on
315 the overall hydrophilic to lipophilic trend of the solutes in that more water soluble
316 compounds have α_{pt} values close to 1 and that the α_{pt} values for lipophilic contaminants
317 (high K_{ow} values) are less than 1 (Chiou et al., 2001). Therefore a value of 0.1 was
318 assigned for both contaminants.

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

$$327 \quad \text{Log } K_{ch} = 1.23 \text{ log } K_{ow} - 2.42 \quad (8)$$

328 2.2 Plant to animal transfer model

329 The daily intake of TCS and TCC in cow's was calculated as the sum of the intake of
330 contaminant in soil (PEC_{soil} mg kg⁻¹), the cow's consumption of soil (F_{soil} , kg⁻¹ d⁻¹), the
331 concentration of the contaminant in the plant (C_{pt}) on a fresh weight basis (mg kg⁻¹) and
332 the cow's consumption of the forage (F_{pt} , kg⁻¹ d⁻¹). Daily intake of a contaminant by a
333 cow is proportional to the amount of forage ingested and the degree of contamination of
334 the particular forage (Chițescu et al., 2014).

335
$$DI = PEC_{soil} \times F_{soil} + C_{pt} \times F_{pt} + C_{plant} \times F_{pt} \quad (9)$$

336

337 Where DI is the daily intake of the contaminant (mg d⁻¹); F_{soil} is the cows consumption of
338 soil. Chițescu et al. (2014) proposed a value of 0.1 kg d⁻¹, whilst Duarte Davidson and
339 Jones (1996) proposed that a cow consumes 0.9 kg d⁻¹ of soil. To account for the
340 uncertainty, a uniform distribution (min 0.1, max 0.9) was assigned (Table 5). The cow's
341 consumption of forage is between 12 and 18 kg d⁻¹ dry matter (Mc Gilloway and Mayne
342 1996), and it was assumed that dairy and beef cows consumed the same amount.
343 Therefore a uniform distribution was also assigned to account for variability and
344 uncertainty. This model also takes into account the consumption of the contaminant that
345 remained on the swards of grass following biosolid application and dissipation rates.

346 2.3 The bio-transfer factor

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

358
$$\text{Log BTF}_b [\text{mg kg}^{-1}/\text{mg d}^{-1}] = - 7.735 + 1.033 \times \log k_{ow} \quad (10)$$

359
$$\text{Log BTF}_m [\text{mg kg}^{-1}/ \text{mg d}^{-1}] = - 8.056 + 0.992 \times \log K_{ow} \quad (11)$$

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

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

363
$$C_{m/b} = \text{BTF} (b, m) \times \text{DI} \times \text{FC}_{(\text{milk}, \text{beef})} \quad (12)$$

364 Where C_m and C_b is the TCS and TCC residue concentrations in beef and milk (mg d^{-1});
365 FC_{milk} and FC_{beef} is the average fat content of milk and beef. The average fat content of
366 milk (FC_{milk}) as reported by the Irish Cooperative Organisation Society (ICOS) (2009) is
367 3.7%. Chițescu et al. (2014) used a value of 4%. To account for uncertainty in the data,
368 a uniform distribution was assigned (Table 6). The average fat content in beef (FC_{beef})
369 tissue can range widely from 7.5% to over 27% (Hendriks et al., 2007). A uniform
370 distribution was used to account for uncertainty.

371

372 2.4 Human exposure

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

375
$$\text{HE} = C_{\text{milk/beef}} \times M_c / \text{bw} \quad (13)$$

376 Where HE is human exposure ($\text{mg kg}^{-1} \text{bw d}^{-1}$); M_c is the consumption of milk or beef a
377 day, and bw is the body weight of the individual. The consumption of milk and beef was
378 based on several studies conducted by The Irish Universities Nutrition Alliance (IUNA).
379 The National pre-school Nutrition Survey investigated the habitual food and drink
380 consumption, health and lifestyle characteristics and assessed the body weight status in
381 500 pre-school children aged 1-4 years and living in the Republic of Ireland between
382 2010 and 2011. The National Children's Food Survey (2003-2004) assessed the
383 consumption and body weights of 594 children (IUNA 2005). The National Teens' Food
384 Survey (2005-2006) investigated habitual food and drink consumption and health and
385 lifestyle characteristics in 441 teenagers aged 13-17 years from the Republic of Ireland.

386 The National Adult Nutrition Survey (2011) assessed the consumption and body weights
387 of 1500 Irish consumers (IUNA 2011). A log normal distribution was used to model the
388 uncertainty regarding the intake of milk and beef. A summary of all human exposure
389 model inputs are provided in Tables 7 and 8.

390

391 2.5 Acceptable daily intake

392 The acceptable daily intake (ADI) procedure has been used to calculate permissible
393 chronic exposure levels for humans based on non-carcinogenic effects. The ADI is the
394 amount of contaminant a human can be exposed to each day over a long time (usually
395 lifetime) without suffering harmful effects. It is determined by applying safety factors (to
396 account for uncertainty in the data) to the highest dose in human or animal studies
397 which has been demonstrated not to cause adverse effects (NOAEL) (EC 2003b). In
398 determining the ADI the no observed adverse effects level (NOAEL) is divided by a
399 safety factor in order to provide a margin of safety for allowable human exposure. A
400 safety factor of 300 was applied in accordance with the European Commission Health
401 and Consumer Protection Directorate-General (2005) and is composed of three factors;
402 10 is for each intra-species, 10 is for each inter-species and 3 is for a limited database
403 of studies (Prosser et al., 2015).

$$404 \quad \text{ADI} = \text{NOAEL} / 10 \times 10 \times 3 \quad (14)$$

405 A NOAEL value of 25 mg kg⁻¹ bw d⁻¹ for TCS and TCC were obtained from Prosser et al.
406 (2015) and was based on a 90 day oral toxicity study with mice and a 2 year oral toxicity
407 study with rats, respectively. A NOAEL of 50 mg kg⁻¹ bw d⁻¹ for TCC was obtained from
408 the USEPA (2008) and was based on the reproductive toxicity of Sprague-Dawley rats
409 over an 80 day period. The Scientific Committee on Consumer Products (SCCP) (2008)
410 proposes a NOAEL of 12 mg kg⁻¹ bw d⁻¹ for TCS based on rat haemotoxicity studies as
411 the critical effect level against which human exposure to TCS is compared. Rodricks et
412 al. (2010) considered over 50 health endpoints and has developed a lower bound
413 benchmark dose level of 47 mg kg⁻¹ bw d⁻¹. To account for variability and uncertainty in
414 the data, a uniform distribution was assigned for both TCS (min 12, max 47 mg kg⁻¹ bw
415 d⁻¹) and TCC (min 25, max 50 mg kg⁻¹ bw d⁻¹) (Table 8).

416 Risk characterisation was quantified by potential non-carcinogenic risks, reflected by the
417 hazard quotient (HQ) – the ratio of the potential exposure to a substance and the level
418 at which no adverse effects are expected (the threshold toxicity reference value (RfD)).
419 If the HQ exceeds 1, there may be concern for non-carcinogenic risks (Lemly, 1996)
420 and range from <0.01 (no existing risk) to > 10 (risk is high). The reference dose value
421 (RfD) (mg kg⁻¹ bw d⁻¹) was calculated according to;

$$422 \quad \quad \quad RfD = NOAEL / UF \times MF \quad \quad \quad (15)$$

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

$$428 \quad \quad \quad HQ = HE / RfD \quad \quad \quad (16)$$

429

430 2.6 Sensitivity analysis

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

436

437 3. Results and discussion

438 The environmental fate of the antimicrobials triclosan and triclocarban were modelled
439 from biosolid application to plant uptake and bio-transfer to animal tissue with
440 subsequent human consumption of milk and beef. The model resulted in several output
441 distributions which include the PEC_{soil}, Concentration on grass (C_{plant}), Concentration in
442 plant tissue (C_{pt}), daily intake of contaminant (DI), and subsequent human exposure

443 (HE) through consumption of beef and milk and the acceptable daily dose based on
444 NOAEL values.

445 The results for the PEC_{soil} show that TCC had a greater concentration in biosolids
446 compared to TCS (mean values $3.90 \times 10^{-2} \text{ mg kg}^{-1}$, 5th and 95th percentile values 8.63
447 $\times 10^{-3}$ and 8.86×10^{-2} for TCC and $2.43 \times 10^{-2} \text{ mg kg}^{-1}$, 5th and 95th percentile values
448 5.19×10^{-3} and 5.37×10^{-2} for TCS) (Figure 2). TCS and TCC have similar chemical
449 properties. Both compounds are polychlorinated aromatic compounds which suggest
450 significant resistance to biodegradation and bio-transformation (Halden and Paull 2005);
451 however, concentrations detected in biosolids may differ.

452 The results for concentration of contaminant remaining on the plant (C_{plant}) show that
453 TCS had the greater concentration remaining (mean value $9.47 \times 10^{-10} \text{ mg kg}^{-1}$, 5th and
454 95th percentile values 5.54×10^{-18} and 3.13×10^{-9} , respectively), compared to TCC
455 (mean value $7.14 \times 10^{-12} \text{ mg kg}^{-1}$, 5th and 95th percentile values 8.28×10^{-28} and $1.69 \times$
456 10^{-11} , respectively). This is in agreement with previous studies investigating the
457 degradation potential of TCS and TCC which indicate that TCC is more persistent in the
458 environment. Cha and Cupples (2010) reported that TCC was more persistent than TCS
459 based on concentrations measured in the soil and the greater half-life values for TCC in
460 aerobic and anaerobic conditions.

461

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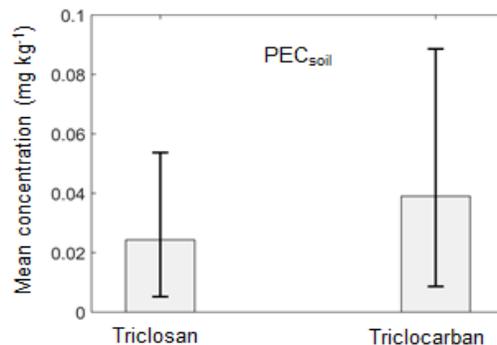


Figure 2: Mean concentrations of TCS and TCC in soil following a single biosolid application (PEC_{soil}) (mg kg^{-1}). Error bars denote 5th and 95th percentiles.

463

464

465 Concentrations in plant tissue were also greater for TCC with mean values 1.40×10^{-5}
466 mg kg^{-1} , 5th and 95th percentile values 1.22×10^{-6} and 4.46×10^{-5} , respectively,
467 compared to TCS $1.23 \times 10^{-5} \text{ mg kg}^{-1}$, 5th and 95th percentile values 6.45×10^{-7} and
468 5.03×10^{-5} , respectively. These results are in agreement with a study conducted by
469 García-Santiago et al., (2016) who demonstrated values of $8.66 \times 10^{-5} \text{ mg kg}^{-1}$ of TCS
470 in plant following a single application of biosolids. Plant uptake of TCS or TCC is a
471 function of many variables which include initial concentration in biosolids, behaviour of
472 contaminant in soil and plant type.

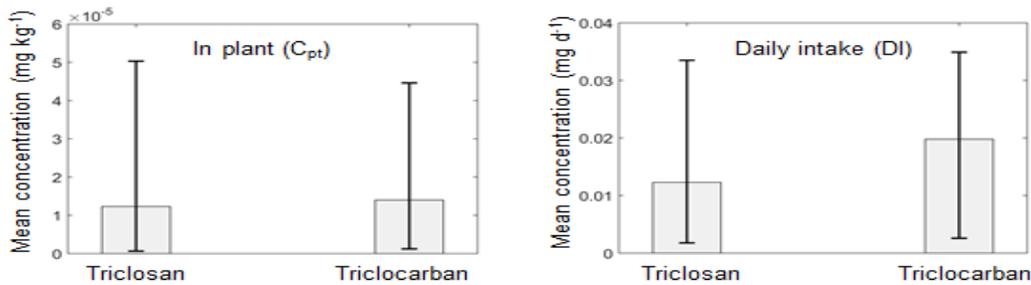


Figure 3: Mean concentrations of TCS and TCC in plant tissue (mg kg^{-1}) and daily cow intake (mg d^{-1}). Error bars denote 5th and 95th percentiles.

473

474

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

485 The daily intake results of TCS and TCC by cows show that TCC had a greater intake
486 rate, mean values $1.97 \times 10^{-2} \text{ mg d}^{-1}$, 5th and 95th percentile values 2.60×10^{-3} and 5.49
487 $\times 10^{-2}$, respectively, compared to TCS with $1.23 \times 10^{-2} \text{ mg d}^{-1}$, 5th and 95th percentile
488 values 1.78×10^{-3} and 3.35×10^{-2} , respectively (Figure 3). Variability in soil and feed
489 concentrations were included to account for uncertainty in the data. The concentration
490 of contaminant on the plant was also included. Depending on the grazing season,
491 concentrations of the contaminant in soil may vary. Concentrations of TCC were greater
492 than TCS in the soil, therefore it was expected that there would be greater
493 concentrations of TCC in the consumption of soil. The concentration in the feed (silage
494 or forage) is dominated by uptake factors previously mentioned (sorption and
495 persistence).

496

497 Predicted mean residue concentrations of TCS and TCC in beef show that
498 concentrations of TCC were greater than TCS in beef (mean value $1.47 \times 10^{-4} \text{ mg kg}^{-1}$,
499 5th and 95th percentile values 3.43×10^{-8} and 8.03×10^{-4} , respectively and mean value
500 $2.62 \times 10^{-6} \text{ mg kg}^{-1}$, 5th and 95th percentile values 2.97×10^{-7} and 7.91×10^{-6} ,
501 respectively) (Figure 4). Mean residue concentrations in milk show that TCC had a
502 higher concentration in milk than TCS (mean value $8.06 \times 10^{-6} \text{ mg kg}^{-1}$, 5th and 95th
503 percentile values 3.04×10^{-9} and 4.43×10^{-5} , respectively and mean value 1.81×10^{-7}
504 mg kg^{-1} , 5th and 95th percentile values 2.42×10^{-8} and 5.13×10^{-7} , respectively) (Figure
505 4). The hydrophilicities of TCC and TCS ($\log K_{ow}$ 4.9 and 4.6, respectively) indicate the
506 potential for bioaccumulation. It has been suggested that compounds with high $\log K_{ow}$
507 values and low water solubilities are the contaminants that have the greatest potential to
508 accumulate in animal tissues (Duarte-Davidson and Jones 1996). Studies have shown
509 that TCC and TCS can bioaccumulate in earthworms (Kinney et al., 2008), TCS in
510 sheep placenta (James et al., 2010) and humans (Adolfsson-Erici et al., 2002). TCS has
511 been found in human breast milk which indicates a route of exposure to the infant to
512 TCS *via* food (Allmyr et al., 2006). Bioaccumulation of TCS and TCC occurs in humans
513 but to a much lesser extent due to well-known detoxification reactions resulting in the
514 rapid elimination of parental TCS and TCC (Halden 2014). Contaminants with a higher

515 half-life $>36 \text{ d}^{-1}$ combined with a higher $\log K_{ow}$ value > 4.5 have been associated with
516 potential animal soil ingestion (Duarte-Davidson and Jones 1996).

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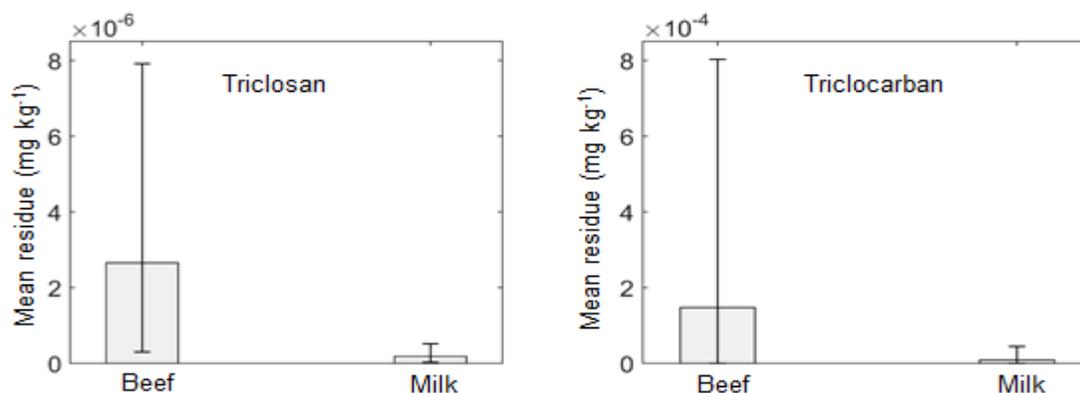


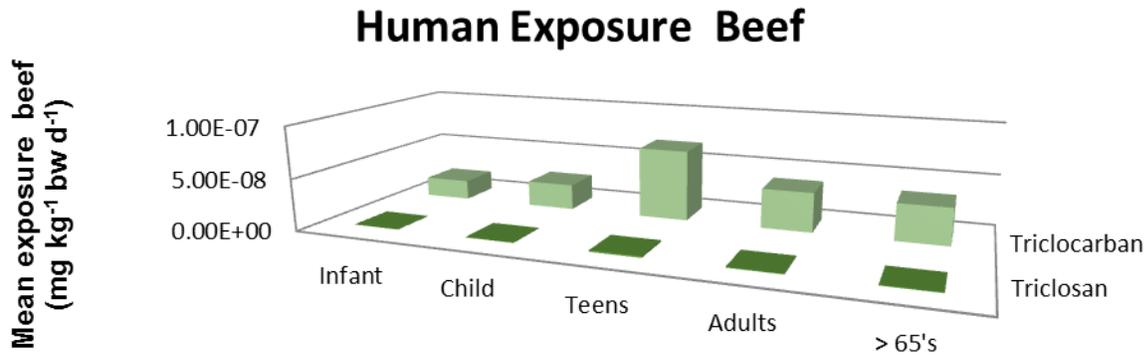
Figure 4: Mean residue concentrations of TCS and TCC in milk and beef (mg kg^{-1}).
Error bars denote 5th and 95th percentiles

519

520

521 The modelled results for human exposure to TCS and TCC in beef show that beef
522 consumption and contaminant concentration was greater for TCC and the teen group
523 (mean value $6.87 \times 10^{-8} \text{ mg kg}^{-1} \text{ bw d}^{-1}$) (Figure 5).

524



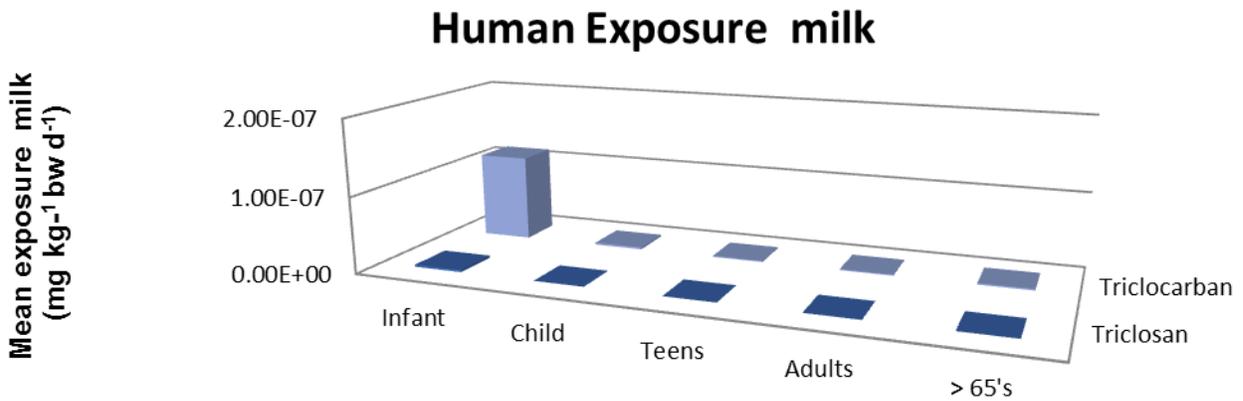
	Infant	Child	Teens	Adults	> 65's
■ Triclosan	3.59E-10	5.09E-10	1.39E-09	6.75E-10	5.74E-10
■ Triclocarban	1.93E-08	2.50E-08	6.87E-08	3.84E-08	3.57E-08

525

526 **Figure 5.** Mean exposure of TCS and TCC in beef ($\text{mg kg}^{-1} \text{bw d}^{-1}$)

527 Mean consumption of milk showed that TCC had the greatest concentration combined
 528 with the infant group (mean value $1.14 \times 10^{-7} \text{mg kg}^{-1} \text{bw d}^{-1}$) (Figure 6).

529



	Infant	Child	Teens	Adults	> 65's
■ Triclosan	2.68E-09	6.44E-11	3.10E-11	3.11E-11	4.28E-11
■ Triclocarban	1.14E-07	2.49E-09	1.36E-09	1.39E-09	1.85E-09

530

531 **Figure 6.** Mean exposure of TCS and TCC in milk ($\text{mg kg}^{-1} \text{bw d}^{-1}$)

532

533 None of the human exposure values exceeded the ADI (mean ADI values of 0.058 mg
534 $\text{kg}^{-1} \text{bw d}^{-1}$ for TCS and $0.1 \text{ mg kg}^{-1} \text{bw d}^{-1}$ for TCC, respectively). Prosser et al. (2014)
535 estimated ADI values of $0.083 \text{ mg kg}^{-1} \text{bw d}^{-1}$ for TCS and TCC based on a NOAEL
536 value of $25 \text{ mg kg}^{-1} \text{bw d}^{-1}$ and an uncertainty factor of 300. Blanset et al. (2007)
537 estimated the ADI for TCS at $0.05 \text{ mg kg}^{-1} \text{bw d}^{-1}$ and concluded that, based on TCS
538 levels typically measured in drinking water, the risk to human health is minimal. The
539 European Union Health and Consumer Protection Directorate-General have set an ADI
540 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
541 ADI for TCS has been established yet. Concentrations of TCS and TCC have been
542 detected in humans following consumption of food products. For example, Chitescu, et
543 al. (2014) showed how 3 pharmaceuticals (sulfamethoxazole, ketoconazole and
544 oxytetracycline) were transferred from contaminated soil through plant uptake and into
545 the dairy food production chain. The results showed that the pharmaceuticals did
546 contaminate the dairy cow's milk and meat due to the ingestion of contaminated feed by
547 the cattle. However, human exposure results were below the ADI for all 3
548 pharmaceuticals and represented a minor risk. Aryal and Reinhold (2011) demonstrated
549 that exposure to TCS and TCC from consumption of pumpkin and zucchini plants
550 following biosolid application (3.25 dry tons per acre) was 2 orders less than exposure
551 from using products that contained TCS and TCC, 35 times greater than exposure to
552 drinking water and 250 times greater than exposure from eating soybeans grown in
553 fields amended with biosolids.

554 The results for the mean HQ based on the NOAEL and milk and beef exposure show
555 that for TCS and milk, potential toxicity risk was greater for the infant group (mean HQ
556 value 1.32×10^{-7} and 95th percentile value 4.5×10^{-7}) while for TCS and beef (mean
557 HQ value 1.02×10^{-5} and 95th percentile value 6.8×10^{-8}), the teen group had the
558 greatest level of toxicity risk (Table 9). The mean HQ results for milk toxicity risk
559 combined with TCC show that the infant group had the highest level of risk (mean HQ
560 value 3.9×10^{-6} and 95th percentile value 1.9×10^{-5}), whilst, the level of toxicity risk for
561 beef showed that the teen group had the highest level (mean HQ value 2.40×10^{-6} and
562 95th percentile value 1.1×10^{-5}) (Table 9). All HQ values were below the threshold value
563 of risk (HQ < 0.01). Therefore this study concludes that there is no appreciable risk to

564 human health. The HQ results show that of all the age categories investigated, the
565 groups more susceptible to toxicity risk are the infant group and the teen group. Prosser
566 and Sibley (2015) reported that the HQ for triclosan in the root of radish plants following
567 amendments with biosolids (single application) was 0.2 for toddlers. García-Santiago et
568 al. (2016) studied the environmental fate and the risks of persistent cosmetics and
569 pharmaceutical compounds following sludge application considering bio-transfer to
570 meat and milk, crops, dermal and inhalation with soil particles and human exposure.
571 The study revealed hazard quotient values of 0.28 for TCS with a 95th percentile of 0.95
572 for root plant ingestion which could pose a potential hazard to human health. Synder
573 and O Connor (2013) performed a two-tiered human health and ecological risk
574 assessment of land applied biosolids-borne TCC. Hazard quotients were calculated to
575 estimate risk for 14 exposure pathways identified in the USEPA Part 503 Biosolids Rule
576 Risk Assessment looking at 100 year application and 'worst case' scenarios. The
577 majority of biosolids-borne TCC exposure pathways resulted in HQ >1. Two pathways
578 exceeded the HQ, the biosolid to predator pathway and biosolid to aquatic organism
579 pathway. The study concluded that there was an unacceptable risk associated with TCC
580 in land applied biosolids.

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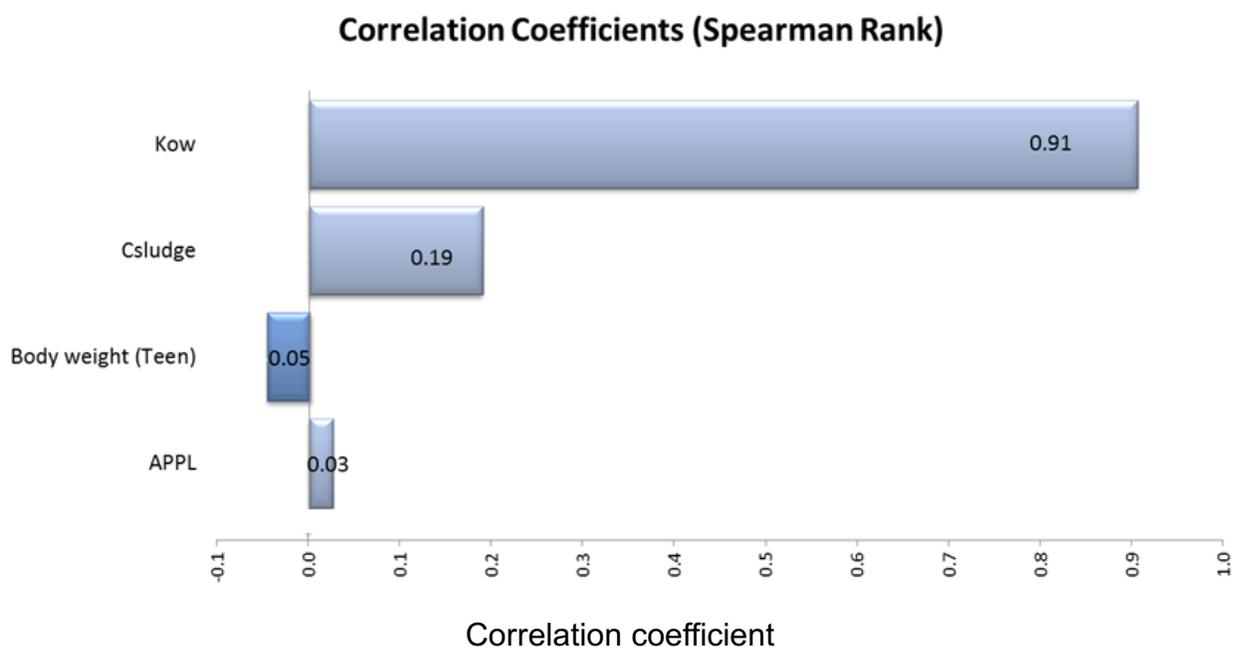
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591 A sensitivity analysis based on the rank order correlation coefficient was conducted for
592 TCC as this contaminant had the highest concentration in biosolids right through to
593 consumption. Sensitivity analysis assesses how the model predictions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

605



606

607

608 **Figure 7.** Sensitivity analysis of correlation coefficient (Spearman Rank) model input

609 4. Conclusion

610 In this study detectable concentrations of TCS and TCC in biosolids estimated from the
611 peer review literature were evaluated to assess their environmental fate in soil and
612 plants, transfer into animal tissues and translocation into the food chain through the
613 consumption of beef or milk. Introduction of these compounds to the environment is
614 mainly through biosolid spreading as most of the TCS and TCC mass entering the
615 WWTP is attached to the particles in the wastewater and most of the mass outgoing is
616 contained in the biosolids. The PEC_{soil} showed that concentrations for TCC were greater
617 than TCS; this was due to the overall concentration in the biosolids and greater half-life.
618 This trend continued throughout the model, however, it cannot be attributed to the initial
619 concentrations in the biosolids alone, rather factors such as sorption and persistence
620 dictates the behaviour of the contaminant in the soil. Both compounds are highly
621 lipophilic and rarely found in soil solution, are preferably found in roots due to the
622 contact with soil particles. This attribute also results in a higher bioaccumulation in beef
623 and milk. Predicted human exposure to TCS and TCC through beef and milk showed
624 that there was no appreciable risks as all values were well below the ADI. A hazard
625 quotient (HQ) was also calculated and the results showed that there was no appreciable
626 risk as all values were < 0.01 . The study showed that infants and teens had the highest
627 level of exposure through milk and beef, respectively, as the data obtained from
628 consumer consumption studies show that these age categories typically consume more
629 milk and beef. Sensitivity analysis showed that the K_{ow} and the initial concentration of
630 the contaminants in biosolids as being the parameters of importance. Once introduced
631 into the soil, concentrations of TCS and TCC may decrease over time as a result of a
632 variety of dissipation processes. The study does highlight a route into which TCS and
633 TCC may enter the food chain through the spreading of biosolids. The fact that they are
634 highly lipophilic may hinder their progress along the food chain; however, their
635 persistence in soil may introduce other consequences such as resistance to antibiotics.
636 While exposure would appear to be small for humans, more research needs to be
637 conducted to evaluate if the continued use of TCS and TCC may exacerbate the issue
638 of antibiotic resistance, which may be another inadvertent consequence of the use of
639 antimicrobials.

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643 5. Reference

- 644 Adolfsson-Erici, M., Petterson, M., Parkkonen, J. & Sturve, J. (2002). Triclosan, a
645 commonly used bactericide found in human milk and in the aquatic environment
646 in Sweden. *Chemosphere*, 46, 1485-1489.
- 647 Agrinet. (2015). Farm Management Software. Available at <http://www.agrinet.ie/>.
648 Accessed August 2016.
- 649 Agyin-Birikorang, S., Miller, M. & O'Connor, G. A. (2010). Retention-release
650 characteristics of triclocarban and triclosan in biosolids, soils, and biosolids-
651 amended soils. *Environmental Toxicology and Chemistry*, 29, 1925-1933.
- 652 Allmyr, M., Adolfsson-Erici, M., McLachlan, M. S. & Sandborgh-Englund, G. (2006).
653 Triclosan in plasma and milk from Swedish nursing mothers and their exposure
654 via personal care products. *Science of the Total Environment*, 372, 87-93.
- 655 Aryal, N. & Reinhold, D. M. (2011). Phytoaccumulation of antimicrobials from biosolids:
656 Impacts on environmental fate and relevance to human exposure. *Water*
657 *Research*, 45, 5545-5552.
- 658 Banihashemi, B. & Droste, R. L. (2014). Sorption–desorption and biosorption of
659 bisphenol A, triclosan, and 17 α -ethinylestradiol to sewage sludge. *Science of The*
660 *Total Environment*, 487, 813-821.
- 661 Barron, L., Havel, J., Purcell, M., Szpak, M., Kelleher, B. & Paull, B. (2009). Predicting
662 sorption of pharmaceuticals and personal care products onto soil and digested
663 sludge using artificial neural networks. *Analyst*, 134, 663-670.
- 664 Bianset, D.L., Zhang, J. and Robson, M.G., (2007). Probabilistic estimates of lifetime
665 daily doses from consumption of drinking water containing trace levels of N, N-
666 diethyl-meta-toluamide (DEET), triclosan, or acetaminophen and the associated
667 risk to human health. *Human and Ecological Risk Assessment*, 13(3), pp.615-
668 631.
- 669 BORD BIA (Irish Food Board). (2016). Factsheet on the Irish Agriculture and Food and
670 Drink Sector. Available at
671 <http://www.bordbia.ie/industry/buyers/industryinfo/agri/pages/default.aspx>.
672 Accessed July 2016.

673 Boxall, A. B. A., Johnson, P., Smith, E. J., Sinclair, C. J., Stutt, E. & Levy, L. S. (2006).
674 Uptake of Veterinary Medicines from Soils into Plants. *Journal of Agricultural and*
675 *Food Chemistry*, 54, 2288-2297.

676 Carr, D. L., Morse, A. N., Zak, J. C. & Anderson, T. A. (2011). Biological degradation of
677 common pharmaceuticals and personal care products in soils with high water
678 content. *Water, Air, & Soil Pollution*, 217, 127-134.

679 Carter, L. J., Harris, E., Williams, M., Ryan, J. J., Kookana, R. S. & Boxall, A. B. A.
680 (2014). Fate and Uptake of Pharmaceuticals in Soil–Plant Systems. *Journal of*
681 *Agricultural and Food Chemistry*, 62, 816-825.

682 Cha, J. & Cupples, A. M. (2010). Triclocarban and triclosan biodegradation at field
683 concentrations and the resulting leaching potentials in three agricultural soils.
684 *Chemosphere*, 81, 494-499.

685 Chalew, T. E. & Halden, R. U. (2009). Environmental exposure of aquatic and terrestrial
686 biota to triclosan and triclocarban1. *JAWRA Journal of the American Water*
687 *Resources Association*, 45, 4-13.

688 CHEMSPIDER. (Royal Society of Chemistry). (2015). Chemistry database. Available at
689 <http://www.chemspider.com/Chemical-Structure.5363.html>. Accessed February
690 2016.

691 Chen, J., Ahn, K. C., Gee, N. A., Ahmed, M. I., Duleba, A. J., Zhao, L., Gee, S. J.,
692 Hammock, B. D. & Lasley, B. L. (2008). Triclocarban enhances testosterone
693 action: a new type of endocrine disruptor? *Endocrinology*, 149, 1173-1179.

694 Chen, X., Nielsen, J. L., Furgal, K., Liu, Y., Lolas, I. B. & Bester, K. (2011).
695 Biodegradation of triclosan and formation of methyl-triclosan in activated sludge
696 under aerobic conditions. *Chemosphere*, 84, 452-456.

697 Chiou, C. T., Sheng, G. & Manes, M. (2001). A partition-limited model for the plant
698 uptake of organic contaminants from soil and water. *Environmental science &*
699 *technology*, 35, 1437-1444.

700 Chitescu, C. L., Nicolau, A. I., Romkens, P. & Van der Fels-Klerx, H. J. (2014).
701 Quantitative modelling to estimate the transfer of pharmaceuticals through the
702 food production system. *Journal of Environmental Science and Health, Part B*,
703 49, 457-467.

704 Chu, S. & Metcalfe, C. D. (2007). Simultaneous determination of triclocarban and
705 triclosan in municipal biosolids by liquid chromatography tandem mass
706 spectrometry. *Journal of Chromatography A*, 1164, 212-218.

707 Clarke, B. O. & Smith, S. R. (2011). Review of 'emerging' organic contaminants in
708 biosolids and assessment of international research priorities for the agricultural
709 use of biosolids. *Environment International*, 37, 226-247.

710 Clarke, R. M. & Cummins, E. (2014). Evaluation of "Classic" and Emerging
711 Contaminants Resulting from the Application of Biosolids to Agricultural Lands: A
712 Review. *Human and Ecological Risk Assessment: An International Journal*, 21,
713 492-513.

714 Clarke, R., Healy, M. G., Fenton, O. & Cummins, E. (2016). A quantitative risk ranking
715 model to evaluate emerging organic contaminants in biosolid amended land and
716 potential transport to drinking water. *Human and Ecological Risk Assessment: An
717 International Journal*, 22, 958-990.

718 Clarke, R., Peyton, D., Healy, M. G., Fenton, O. & Cummins, E. (2016). A quantitative
719 risk assessment for metals in surface water following the application of biosolids
720 to grassland. *Science of the Total Environment*, 566, 102-112.

721 Coogan, M. A. & Point, T. W. L. (2008). Snail bioaccumulation of triclocarban, triclosan,
722 and methyltriclosan in a north texas, usa, stream affected by wastewater
723 treatment plant runoff. *Environmental Toxicology and Chemistry*, 27, 1788-1793.

724 DAFM (Department of Agriculture, Fisheries and Marine). (2016). Fact sheet on Irish
725 agriculture. Available at
726 <http://www.agriculture.gov.ie/media/migration/publications/2016/February2016Factsheet170216.pdf> . Accessed July 2016.

727

728 Davis, E. F., Klosterhaus, S. L. & Stapleton, H. M. (2012). Measurement of flame
729 retardants and triclosan in municipal sewage sludge and biosolids. *Environment
730 International*, 40, 1-7.

731 Dettenmaier, E., CIVIL, U. S. U. & ENVIRONMENTAL (2008). *Measuring and Modeling
732 of Plant Root Uptake of Organic Chemicals*, Utah State University.

733 Dhillon, G. S., Kaur, S., Pulicharla, R., Brar, S. K., Cledon, M., Verma, M. & Surampalli,
734 R. Y. (2015). Triclosan: current status, occurrence, environmental risks and

735 bioaccumulation potential. *International journal of environmental research and*
736 *public health*, 12, 5657-5684.

737 Duarte-Davidson, R. & Jones, K. C. (1996). Screening the environmental fate of organic
738 contaminants in sewage sludge applied to agricultural soils: II. The potential for
739 transfers to plants and grazing animals. *Science of The Total Environment*, 185,
740 59-70.

741 EC (European Commission). (2003a). Technical Guidance Document on Risk
742 Assessment in support of Commission directive 93/67/EEC on Risk Assessment
743 for new notified substances. Commission regulation (EC) No1488/94 on Risk
744 Assessment for Existing substances, Directive 98/8/EC of the European
745 Parliament and of the Council concerning the placing of biocidal products on the
746 market. Available at
747 https://echa.europa.eu/documents/10162/16960216/tgdpart2_2ed_en.pdf.
748 Accessed March 2016

749 EC (European Commission). (2003b). Introduction to risk assessment. Available
750 at [http://ec.europa.eu/health/ph_projects/2003/action3/docs/2003_3_09_a23_en](http://ec.europa.eu/health/ph_projects/2003/action3/docs/2003_3_09_a23_en.pdf)
751 [.pdf](http://ec.europa.eu/health/ph_projects/2003/action3/docs/2003_3_09_a23_en.pdf). Accessed July 2016.

752 EC (European Commission). (2008). Scientific Committee on Consumer Products
753 Opinion on Triclosan. Available at
754 http://ec.europa.eu/health/ph_risk/committees/04_sccp/docs/sccp_o_166.pdf.
755 Accessed March 2015.

756 EC (European Commission). (2005). Health and Consumer Protection Directorate-
757 General, 2005. Scientific Committee on Consumer Products - Opinion on
758 Triclocarban. European Commission Health and Consumer Protection
759 Directorate-General, Brussels, Belgium.

760 EC (European Commission). (2016). Agriculture. Available at
761 http://ec.europa.eu/ireland/news/key-eu-policy-areas/agriculture_en. Accessed
762 July 2016.

763

764 Fehily Timoney and Company. (1999). Codes of Good Practice for the Use of Biosolids
765 in Agriculture. Available at

766 [http://www.environ.ie/en/Publications/Environment/Water/FileDownload,17228,e](http://www.environ.ie/en/Publications/Environment/Water/FileDownload,17228,en.pdf)
767 [n.pdf](http://www.environ.ie/en/Publications/Environment/Water/FileDownload,17228,en.pdf) . Accessed June 2015.

768 Fent, K., Weston, A. A. & Caminada, D. (2006). Ecotoxicology of human
769 pharmaceuticals. *Aquatic Toxicology*, 76, 122-159.

770 Fu, Q., Wu, X., Ye, Q., Ernst, F. & Gan, J. (2016). Biosolids inhibit bioavailability and
771 plant uptake of triclosan and triclocarban. *Water Research*, 102, 117-124.

772 Garcia-Santiago, X., Franco-Uria, A., Omil, F. and Lema, J.M., (2016). Risk assessment
773 of persistent pharmaceuticals in biosolids: Dealing with uncertainty. *Journal of*
774 *hazardous materials*, 302, pp.72-81.

775 Gasperi J., Geara, D., Lorgeoux, C., Bressy, A., Zedek, S., Rocher, V., EL Samrani, A.,
776 Chebbo, G. & Molilleron, R. (2014). First assessment of triclosan, triclocarban
777 and paraben mass loads at a very large regional scale: Case of Paris
778 conurbation (France). *Science of The Total Environment*, 493, 854-861.

779 Goldstein, M., Shenker, M. & Chefetz, B. (2014). Insights into the uptake processes of
780 wastewater-borne pharmaceuticals by vegetables. *Environmental science &*
781 *technology*, 48, 5593-5600.

782 Halden, R. U. (2014). On the Need and Speed of Regulating Triclosan and Triclocarban
783 in the United States. *Environmental Science & Technology*, 48, 3603-3611.

784 Halden, R. U. & Paull, D. H. (2005). Co-occurrence of triclocarban and triclosan in US
785 water resources. *Environmental Science & Technology*, 39, 1420-1426.

786 Harris S, Cormican M and Cummins E (2012). Antimicrobial residues and antimicrobial-
787 resistant bacteria: Impact on the microbial environment and risk to human health.
788 *Human and Ecological Risk Assessment*, 18 (4):767-809.

789 Heidler, J., Sapkota, A. & Halden, R. U. (2006). Partitioning, persistence, and
790 accumulation in digested sludge of the topical antiseptic triclocarban during
791 wastewater treatment. *Environmental science & technology*, 40, 3634-3639.

792 Hellstrom, T. (2000). Brominated flame retardants (PBDE and PBB) in sludge-a
793 problem. *The Swedish Water and Wastewater Association, Report No M*, 113,
794 31.

795 Hendriks, A. J., Smitkova, H. & Huijbregts, M. A. (2007). A new twist on an old
796 regression: transfer of chemicals to beef and milk in human and ecological risk
797 assessment. *Chemosphere*, 70, 46-56.

798 Higgins, C. P., Paesani, Z. J., Abbot Chalew, T. E., Halden, R. U. & Hundal, L. S.
799 (2011). Persistence of triclocarban and triclosan in soils after land application of
800 biosolids and bioaccumulation in *Eisenia foetida*. *Environmental Toxicology and*
801 *Chemistry*, 30, 556-563.

802 Hinthner, A., Bromba, C. M., Wulff, J. E. & Helbing, C. C. (2011). Effects of triclocarban,
803 triclosan, and methyl triclosan on thyroid hormone action and stress in frog and
804 mammalian culture systems. *Environmental science & technology*, 45, 5395-
805 5402.

806 Holling, C. S., Bailey, J. L., Heuvel, B. V. & Kinney, C. A. (2012). Uptake of human
807 pharmaceuticals and personal care products by cabbage (*Brassica campestris*)
808 from fortified and biosolids-amended soils. *Journal of Environmental Monitoring*,
809 14, 3029-3036.

810 Hung, H.-W., Lin, T.-F. & Chiou, C. T. (2010). Partition coefficients of organic
811 contaminants with carbohydrates. *Environmental science & technology*, 44,
812 5430-5436.

813 ICOS (Irish Cooperative Organisation Society). (2009). International milk production
814 comparisons. Available at <http://www.icos.ie/supply-chain/transport-quality/>.
815 Accessed July 2016.

816 IDIA (Irish Dairy Industries Association). (2016). Industry overview. Available at
817 <http://www.fdi.ie/idia>. Accessed July 2016.

818 Igos, E., Benetto, E., Venditti, S., Kohler, C., Cornelissen, A., Moeller, R. & Biber, A.
819 (2012). Is it better to remove pharmaceuticals in decentralized or conventional
820 wastewater treatment plants? A life cycle assessment comparison. *Science of*
821 *The Total Environment*, 438, 533-540.

822 Irish Water. (2015). National Wastewater Sludge Management Plan. Asset
823 strategy. Available at [http://www.water.ie/about-us/project-and-plans/wastewater-](http://www.water.ie/about-us/project-and-plans/wastewater-sludge-management/NWSMP-report.pdf)
824 [sludge-management/NWSMP-report.pdf](http://www.water.ie/about-us/project-and-plans/wastewater-sludge-management/NWSMP-report.pdf). Accessed July 2016.

825 IUNA (Irish Universities Nutrition alliance). 2010-2011. The National pre-school
826 Nutrition Survey. Available at <http://www.iuna.net>. Accessed March 2016.

827 IUNA (Irish Universities Nutrition alliance). 2003-2004. The National Childrens Food
828 Survey. [Available at IUNA.net](#). Accessed March 2016.

829 IUNA (Irish Universities Nutrition alliance).2005-2006. Available at [IUNA.net](#). Accessed
830 March 2016.

831 IUNA (Irish Universities Nutrition alliance).2011. National Adult Nutrition Survey.
832 Available at [IUNA.net](#). Accessed March 2016.

833 James, M. O., LI, W., Summerlot, D. P., Rowland-Faux, L. & Wood, C. E. (2010).
834 Triclosan is a potent inhibitor of estradiol and estrone sulfonation in sheep
835 placenta. *Environment international*, 36, 942-949.

836 Jelic, A., Gros, M., Petrovic, M., Ginebreda, A. & Barcelo, D. (2012). Occurrence and
837 elimination of pharmaceuticals during conventional wastewater treatment.
838 *Emerging and Priority Pollutants in Rivers*. Springer.

839 King, M. K. (2010). Evaluation of the Developmental Effects and Bioaccumulation
840 Potential of Triclosan and Triclocarban Using the South African Clawed Frog,
841 *Xenopus Laevis*.

842 Kinney, C. A., Furlong, E. T., Kolpin, D. W., Burkhardt, M. R., Zaugg, S. D., Werner, S.
843 L., Bossio, J. P. & Benotti, M. J. (2008). Bioaccumulation of pharmaceuticals and
844 other anthropogenic waste indicators in earthworms from agricultural soil
845 amended with biosolid or swine manure. *Environmental Science & Technology*,
846 42, 1863-1870.

847 Langdon, K. A., Warne, M. S. J., Smernik, R. J., Shareef, A. & Kookana, R. S. (2012).
848 Field dissipation of 4-nonylphenol, 4-t-octylphenol, triclosan and bisphenol A
849 following land application of biosolids. *Chemosphere*, 86, 1050-1058.

850 Lemly, A. D. (1996). Evaluation of the hazard quotient method for risk assessment of
851 selenium. *Ecotoxicology and Environmental Safety*, 35, 156-162.

852 Li, H., Sheng, G., Chiou, C. T. & Xu, O. (2005). Relation of organic contaminant
853 equilibrium sorption and kinetic uptake in plants. *Environmental science &*
854 *technology*, 39, 4864-4870.

855 Linders, J., Mensink, H., Stephenson, G., Wauchope, D. & Racke, K. (2000). Foliar
856 interception and retention values after pesticide application. A proposal for
857 standardized values for environmental risk assessment (Technical report). *Pure*
858 *and applied chemistry*, 72, 2199-2218.

859 Lucid, J. D., Fenton, O., AND Healy, M.G. (2013). Estimation of maximum biosolids and
860 Meat and Bone Meal Application to a low P Index soil and a Method to test for
861 Nutrient and Metal Losses. *Water, Air, & Soil Pollution*, 224, 1464-1475.

862 Lupton, S. J., Dearfield, K. L., Johnson, J. J., Wagner, S. & Huwe, J. K. (2015).
863 Perfluorooctane Sulfonate Plasma Half-Life Determination and Long-Term
864 Tissue Distribution in Beef Cattle (*Bos taurus*). *Journal of agricultural and food*
865 *chemistry*, 63, 10988-10994.

866 Mattina, M. I., Lannucci-Berger, W., Musante, C. & White, J. C. (2003). Concurrent plant
867 uptake of heavy metals and persistent organic pollutants from soil. *Environmental*
868 *Pollution*, 124, 375-378.

869 McClellan, K. & Halden, R. U. (2010). Pharmaceuticals and personal care products in
870 archived U.S. biosolids from the 2001 EPA national sewage sludge survey.
871 *Water Research*, 44, 658-668.

872 Mc Gilloway D.A. and Mayne C.S. (1996). The importance of grass availability for the
873 high genetic merit dairy cow. In: 'Recent Advances in Animal Nutrition' 1996;
874 Eds. P.C. Garnsworthy, J. Wiseman and W. Haresign. Chp. 8, 135-169.

875 (OEHHA)(Office of Environmental Health Hazard Assessment). (2010). Triclocarban.
876 Potential designated chemical. May 24, 2010 meeting of Scientific Guidance
877 Panel (SGP). Biomonitoring California. Available at
878 <http://oehha.ca.gov/multimedia/biomon/pdf/052410Triclocarban.pdf>. Accessed
879 [May 2016](#)

880 Ogunyoku, T. A. & Young, T. M. (2014). Removal of Triclocarban and Triclosan during
881 Municipal Biosolid Production. *Water environment research : a research*
882 *publication of the Water Environment Federation*, 86, 197-203.

883 Prosser, R. & Sibley, P. (2015). Human health risk assessment of pharmaceuticals and
884 personal care products in plant tissue due to biosolids and manure amendments,
885 and wastewater irrigation. *Environment international*, 75, 223-233.

886 Rodricks JV, Swenberg JA, Borzelleca JF, Marronpot RR, Shipp AM. (2010). Triclosan: A
887 critical review of the experimental data and development of margins of safety for
888 consumer products. *Crit Rev Toxicol* 40:422–484.

889 Rodrigues, S., Pereira, M., Duarte, A. & Romkens, P. (2012). Soil–plant–animal transfer
890 models to improve soil protection guidelines: a case study from Portugal.
891 *Environment international*, 39, 27-37.

892 Rudel, H., Bohmer, W., Muller, M., Fliedner, A., Ricking, M., Teubner, D. & Schroter-
893 Kermani, C. (2013). Retrospective study of triclosan and methyl-triclosan
894 residues in fish and suspended particulate matter: Results from the German
895 Environmental Specimen Bank. *Chemosphere*, 91, 1517-1524.

896 Sabourin, L., Duenk, P., Bonte-Gelok, S., Payne, M., Lapen, D. R. & Topp, E. (2012).
897 Uptake of pharmaceuticals, hormones and parabens into vegetables grown in
898 soil fertilized with municipal biosolids. *Science of The Total Environment*, 431,
899 233-236.

900 Shone, M. & Wood, A. V. (1974). A Comparison of the Uptake and Translocation of
901 Some Organic Herbicides and a Systemic Fungicide by Barley I. ABSORPTION
902 IN EELATION TO PHYSICO-CHEMICAL PROPERTIES. *Journal of Experimental*
903 *Botany*, 25, 390-400.

904 Snyder, E. H., O'Connor, G. A. & McAvoy, D. C. (2010). Fate of 14 C–triclocarban in
905 biosolids-amended soils. *Science of the Total Environment*, 408, 2726-2732.

906 Snyder, E.H. and O'Connor, G.A., (2013). Risk assessment of land-applied biosolids-
907 borne triclocarban (TCC). *Science of the total environment*, 442, pp.437-444.

908 Statutory Instrument. (2010). S.I. No. 610 of 2010. European Union (Good Agricultural
909 Practice for Protection of Waters) Regulations 2010. The Statutory Office.
910 Available at <http://www.irishstatutebook.ie/eli/2010/si/610/made/en/pdf>
911 Accessed February 2016.

912 Sweetman, A. J., Valle, M. D., Prevedouros, K. & Jones, K. C. (2005). The role of soil
913 organic carbon in the global cycling of persistent organic pollutants (POPs):
914 interpreting and modelling field data. *Chemosphere*, 60, 959-972.

915 Taylor-Smith, A. (2015). *Pharmaceutical compounds in land-applied sludge and plant*
916 *uptake: A review*. North Carolina State University.

917 TCC, O. B.-B. T. (2009). *Fate, Transport, and Risk Assessment*. University of Florida.

918 Thompson, A., Griffin, P., Stuetz, R. & Cartmell, E. (2005). The fate and removal of
919 triclosan during wastewater treatment. *Water environment research*, 77, 63-67

920 Trapp, S. (2000). Modelling uptake into roots and subsequent translocation of neutral
921 and ionisable organic compounds. *Pest Management Science*, 56, 767-778.

922 Travis, C. C. & Arms, A. D. (1988). Bioconcentration of organics in beef, milk, and
923 vegetation. *Environmental science & technology*, 22, 271-274.

924 USEPA (US Environmental Protection Agency).(2008). Screening-level hazard
925 characterisation of high production volume chemicals. Available at
926 [http://www.epa.gov/hpvis/hazchar/101202 Triclocarban HC INTERIM March%](http://www.epa.gov/hpvis/hazchar/101202_Triclocarban_HC_INTERIM_March%202008.pdf)
927 [202008.pdf](http://www.epa.gov/hpvis/hazchar/101202_Triclocarban_HC_INTERIM_March%202008.pdf). Accessed March 2015.

928 USEPA (US Environmental Protection Agency).(2009). Targeted National Sewage
929 Sludge Survey Sampling and Analysis Technical Report. Washington, D.C: 2009.
930 EPA-822-R-08-014.

931 USEPA (US Environmental Protection Agency).(2013). PBT PROFILER. Available at
932 <http://www.pbtprofiler.net/>. Accessed May 2016.

933 USEPA (U.S. Environmental Protection Agency). (2005). Methodology for predicting
934 cattle biotransfer factors, EPA Contract Number 68-W-03-042, Research Triangle
935 Institute. Available at
936 <https://archive.epa.gov/epawaste/hazard/tsd/td/web/pdf/btfreportfull05.pdf>
937 [Accessed February 2016](https://archive.epa.gov/epawaste/hazard/tsd/td/web/pdf/btfreportfull05.pdf). Accessed June 2016.

938 USEPA (US Environmental Protection Agency). (2015). Reference Dose (RfD):
939 Description and Use in Health Risk Assessments. Available at
940 [https://www.epa.gov/iris/reference-dose-rfd-description-and-use-health-risk-](https://www.epa.gov/iris/reference-dose-rfd-description-and-use-health-risk-assessments)
941 [assessments](https://www.epa.gov/iris/reference-dose-rfd-description-and-use-health-risk-assessments) . Accessed August 2016.

942 Vero, S. E., Antille, D. L., Lalor, S. T. J. & Holden, N. M. (2014). Field evaluation of soil
943 moisture deficit thresholds for limits to trafficability with slurry spreading
944 equipment on grassland. *Soil Use and Management*, 30, 69-77.

945 Walters, E., McClellan, K. & Halden, R. U. (2010). Occurrence and loss over three years
946 of 72 pharmaceuticals and personal care products from biosolids–soil mixtures in
947 outdoor mesocosms. *Water Research*, 44, 6011-6020.

948 WFD (Water framework Directive). (2012). Proposed EQS for Water Framework
949 Directive Annex VIII substances: triclosan (For consultation). Available at
950 <http://www.wfduk.org/sites/default/files/Media/Triclosan%20-%20UKTAG.pdf>
951 Accessed May 2012.

952 Wild, S. & Jones, K. (1992). Organic chemicals entering agricultural soils in sewage
953 sludges: screening for their potential to transfer to crop plants and livestock.
954 *Science of the total environment*, 119, 85-119.

955 Wu, C., Spongberg, A. L. & Witter, J. D. (2009). Adsorption and degradation of triclosan
956 and triclocarban in soils and biosolids-amended soils. *Journal of agricultural and*
957 *food chemistry*, 57, 4900-4905.

958 Wu, C., Spongberg, A. L., Witter, J. D., Fang, M. & Czajkowski, K. P. (2010). Uptake of
959 Pharmaceutical and Personal Care Products by Soybean Plants from Soils
960 Applied with Biosolids and Irrigated with Contaminated Water. *Environmental*
961 *Science & Technology*, 44, 6157-6161.

962 Wu, C., Spongberg, A. L., Witter, J. D. & Sridhar, B. B. M. (2012). Transfer of
963 wastewater associated pharmaceuticals and personal care products to crop
964 plants from biosolids treated soil. *Ecotoxicology and Environmental Safety*, 85,
965 104-109.

966 Xia, K., Hundal, L. S., Kumar, K., Armbrust, K., Cox, A. E. & Granato, T. C. (2010).
967 Triclocarban, triclosan, polybrominated diphenyl ethers, and 4-nonylphenol in
968 biosolids and in soil receiving 33-year biosolids application. *Environmental*
969 *Toxicology and Chemistry*, 29, 597-605.

970 Ying, G.-G., Yu, X.-Y. & Kookana, R. S. (2007). Biological degradation of triclocarban
971 and triclosan in a soil under aerobic and anaerobic conditions and comparison
972 with environmental fate modelling. *Environmental Pollution*, 150, 300-30

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

	Concentration in biosolids ($\mu\text{g kg}^{-1}$)	
Triclosan	^a 1840	^a (Clarke and Smith 2011), ^b (Davis et al., 2012), ^c (Walters et al., 2010), ^d (Chu and Metcalf 2007)
	2830	
	3210	
	5993	
	^b 4370	
	1429	
	11843	
	12876	
	1265	
	^c 7860	
	^d 9080	
	11550	
	1490	
	1110	
	1510	
17950		
Triclocarban	^a 5970	^a (Chu and Metcalf 2007), ^b (Synder et al 2010), ^c (Mc Clellan and Halden 2010), ^d (Walters et al., 2010), ^e (Cha and Cupples 2010)
	3050	
	5490	
	4920	
	3300	
	3490	
	3700	
	4780	
	^b 19000	
	^c 36000	
	^d 2715	
	^e 4510	
	7085	

974 **Table 2:** Model inputs, distributions and outputs for PECsoil

Stage	Symbols	Description	Model / distribution	Units
PEC _{soil}				
	C_{sludge}	Concentration in biosolids	Uniform or triangular (contaminant specific, table 1)	mg kg ⁻¹
	APPL	Application rate	Triangular (300,330, 520)	kg m ⁻²
	f_{int}	Crop intersection	Triangular (0, 10, 20)	-
	D	Depth	0.1	m
	BD	Bulk density	Uniform (min 800, max 1000)	kg m ⁻³
Output	C_{soil}	$(C_{sludge} \times APPL \times (1 - f_{int} \times 100)) / (D \times BD)$		mg kg ⁻¹
	DT ₅₀	Half-life in soil	Uniform (contaminant specific, Table 3)	d ⁻¹
	k	Dissipation rate constant	$\ln(2)/DT_{50\ soil}$	d ⁻¹
	t	Time to graze	Uniform (min 21, max 42)	d ⁻¹
Output	PECsoil	Concentration of contaminant in soil following dissipation	$C_{soil} \times e^{-kt}$	mg kg ⁻¹
	$C_{applied}$	Concentration of contaminant applied on grass	$C_{sludge} \times APPL \times f_{int}$	mg m ⁻²
	DT ₅₀	Half-life in air	Uniform (contaminant specific, Table 3)	d ⁻¹
	K	Dissipation rate constant	$\ln(2)/DT_{50\ air}$	
	P_d	Plant density	1.8	kg m ⁻²
Output	C_{plant}	Concentration on plant following dissipation	$C_{applied} \times e^{-kt} / P_d$	mg kg ⁻¹

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977 Table 3: Properties of triclosan and triclocarban

Contaminant	Distribution	Min	Mean	Max	References
Triclosan					
Log K_{oc} (L kg ⁻¹)	Triangular ^a	2.7	4.0	4.7	^a Barron et al., 2009, Agyin-Birikorang et al 2010, Chen et al., 2011, Gasperi et al., 2011. WFD 2012.
C_{sludge} (mg kg ⁻¹)	Triangular ^b	1,110	7,298	19,676	^b Chu and Metcalfe 2007, Clarke & Smith 2011, Walters et al., 2010, Cha & Cupples. 2010, Davis et al., 2012.
K_{ow}	Triangular ^c	4.38	4.66	4.8	^c Coogan et al., 2007, Wu et al., 2009, Chen et al., 2011, Rudel et al., 2013, Banihashemi & Droste 2014, Chemspider
DT ₅₀ soil	Uniform ^d	87		231	^d Wu et al., 2009, Chemspider 2015.
DT ₅₀ air	Uniform ^e	0.66		1	^e USEPA 2013, Ying et al. 2007
Henry's Law Constant	Uniform ^f	1.3×10^{-3}		5.2×10^{-4}	^f Thompson et al 2005
Triclocarban					
Log K_{oc} (L kg ⁻¹)	Triangular ^f	3.59	4.06	4.9	^f Ying et al., 2007, King 2010, Cha & Cupples 2010, Chemspider 2015.
C_{sludge} (mg kg ⁻¹)	Triangular ^g	2,715	14,756	38,839	^g Chu & Metcalfe 2007, Snyder et al., 2010, Walters et al., 2010, Mc Clellan & Halden 2010, Cha & Cupples 2010.
K_{ow}	Uniform ^h	2.7		7.1	^h Wu et al., 2009, Agyin-Birikorang et al 2010, Oehha 2010,
DT ₅₀ soil	Uniform ⁱ	18		120	ⁱ Ying et al 2007, Walters et al 2010)
DT ₅₀ air	Uniform ^j	0.5		0.75	^j USEPA 2013, Ying et al 2007
Henry's Law Constant	Uniform ^k	3.6×10^{-5}		8.3×10^{-6}	^k chemspider 2015

979 Table 4. Model inputs, distributions and outputs for PEC_{porewater} and concentration of
 980 contaminant in external water

PEC _{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 ⁻¹
Concentration of contaminant in external water				
Output	PEC _{porewater}	Concentration of contaminant in external water	PEC _{soil} / (F _{oc} × K _{oc})	mg kg ⁻¹
	α _{pt}	Quasi-equilibrium factor	0.1	-
	f _{pw}	Weight fraction of water	89	%
	f _{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 × K _{ow} – 2.42	-
	f _{lip}	Weight fraction of lipids	97	%
	k _{lip}	Partition coefficient for lipid fraction of plant	K _{ow}	log
Output	C _{pt}	Concentration of contaminant in plant	C _{pt} = α _{pt} × PEC _{porewater} × [f _{pw} + f _{ch} × K _{ch} + f _{lip} × K _{lip}]	mg kg ⁻¹

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988 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	$\text{PEC}_{\text{soil}} \times F_{\text{soil}} + C_{\text{pt}} \times F_{\text{pt}} + C_{\text{plant}} \times F_{\text{pt}}$	mg d^{-1}

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1006 Table 6. Bio-transfer factor and residue for milk and beef

Bio-transfer factor				
	BTF _b	Bio-transfer factor beef	Log BTF = - 7,735 +1.033 log K _{ow}	[mg kg ⁻¹ /mg d ⁻¹]
	BTF _m	Bio-transfer factor milk	Log BTF = -8.056 +0.992 log K _{ow}	[mg kg ⁻¹ /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	BTF _m × DI × FC _{milk}	mg d ⁻¹
Output	C _{beef}	residue in beef	BTF _b × DI × FC _{beef}	mg d ⁻¹

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1026 **Table 7:** Mean consumption and standard deviation of milk and beef for individual age
 1027 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⁻¹)	220±193	9 ±13	10 ±14	13 ±19	17 ±19
Beef consumption (g d⁻¹)	2 ±5	5 ±11	30 ± 44	19 ±31	16 ±27

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1050 Table 8. Model inputs, distributions and outputs for human exposure and hazard
 1051 quotient.

Human exposure				
	bw	Body weight	Normal (Table 7)	kg
	Milk _{consum}	Milk consumption	Lognormal (Table 7)	kg d ⁻¹
Output	HE _{milk}	Human exposure milk	$C_{milk} \times \text{Milk}_{consum} / bw$	mg kg ⁻¹ bw d ⁻¹
	Beef _{consum}	Beef consumption	Lognormal (Table 7)	kg d ⁻¹
Output	HE _{beef}	Human exposure beef	$C_{beef} \times \text{beef}_{consum} / bw$	mg kg ⁻¹ bw d ⁻¹
	NOAEL	No observed adverse effects level	(TCS -min 12, max 47) (TCC- min 25, max 50)	mg kg ⁻¹ bw d ⁻¹
	SF	Safety factor	10 × 10 × 3	-
	ADI	Acceptable daily intake	NOAEL/safety factor	mg kg ⁻¹ bw d ⁻¹
	UF	Uncertainty factor	1000	-
	MF	Modifying factor	0.8	-
	R/D	Reference dose	NOAEL/ UF × MF	mg kg ⁻¹ bw d ⁻¹
Output	HQ	Hazard quotient	HE / R/D	-

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

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

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