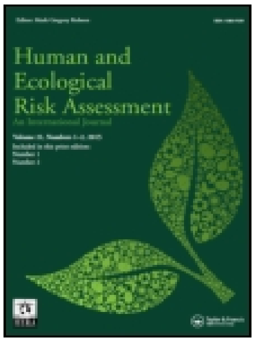

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A quantitative risk ranking model to evaluate emerging organic contaminants in biosolid amended land and potential transport to drinking water

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ABSTRACT

A quantitative risk ranking model was developed for human exposure to emerging contaminants (EC) following treated municipal sewage sludge (“biosolids”) application to Irish agricultural land. The model encompasses the predicted environmental concentration (PEC) in soil, surface runoff, groundwater, and subsequent drinking water ingestion by humans. Human exposure and subsequent risk was estimated for 16 organic contaminants using a Monte Carlo simulation approach. Nonylphenols ranked the highest across three environmental compartments: concentration in soil (PEC_{soil}), runoff (PEC_{runoff}), and groundwater ($PEC_{groundwater}$), which had mean values of 5.69 mg/kg, 1.15×10^{-2} μg/l, and 2.22×10^{-1} μg/l, respectively. Human health risk was estimated using the LC_{50} (chemical intake toxicity ratio, (RR)) as a toxicity endpoint combined with PEC_{runoff} and $PEC_{groundwater}$. NP ranked highest for LC_{50} combined with PEC_{runoff} and $PEC_{groundwater}$ (mean RR values 1.10×10^{-4} and 2.40×10^{-3}, respectively). The model highlighted triclocarban and triclosan as ECs requiring further investigation. A sensitivity analysis revealed that soil sorption coefficient and soil organic carbon were the most important parameters that affected model variance (correlation coefficient –0.89 and –0.30, respectively), highlighting the significance of contaminant and soil properties in influencing risk assessments. This model can help to prioritize emerging contaminants of concern requiring vigilance in environmental compartments.

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water; contaminants; biosolids; risk; soil; human consumption

Introduction

More than 10 million tons of sewage sludge was produced in the European Union (EU) in 2010 (Eurostat 2014). After appropriate treatment, the sludge (referred to as “biosolids”) may be applied, as an agricultural fertilizer, to land. There are considerable public acceptance issues surrounding the reuse of treated sludge as a fertilizer. The main fear is that the presence of organic contaminants in biosolids may accumulate in the food chain, or cause the

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contamination of soil and water (Cornu *et al.* 2001). Recent field biosolid studies have shown that organic (Topp *et al.* 2008) and inorganic contaminants (Gottschall *et al.* 2012) can be lost along surface runoff and leaching pathways, where land application is followed by episodic rainfall events. Emerging contaminants can be defined as any synthetic or naturally occurring chemical or microorganism that is not commonly monitored in the environment and cause known or suspected adverse ecological or human health effects (Careghini *et al.* 2014). The application of municipal biosolids on agricultural land could potentially be a widespread source of EC loss to surface and groundwater bodies (Smith 2009).

The quantity of biosolids applied to agricultural land within the European Union has increased in recent times. The impetus for this increase is as a result of four directives. The Sewage Sludge Directive 86/278/EEC (CEC 1986), The Urban Waste Water Treatment Directive (CEC 1991), which was amended by 98/15/EC in 2005, The Landfill Directive 1999/31/EC (CEC 1999), and The Waste Framework Directive 2008/98/EC (CEU 2012) encourage the reuse of biosolids whenever appropriate with an emphasis on protection of human health, water, soil, and air.

Many organic contaminants are resistant to removal during the sewage treatment process (Clarke and Cummins 2015), may persist as part of the effluent/sludge, or may be released directly or indirectly into the aquatic environment following land application of biosolids (Murray *et al.* 2010). Previous reports have indicated that land application of biosolids can be an important route through which contaminants enter the environment. McClellan and Halden (2010) reported the mean concentrations of 72 various pharmaceuticals and personal care products (PPCPs) detected in 110 archived biosolid samples that were collected by the USEPA in 2001. The report showed that triclosan and triclocarban were the most abundant contaminants with mean concentrations 12.6 ± 3.8 and 36 ± 8 mg/kg. Lapen *et al.* (2008) reported that following land application of biosolids (common rate of application 93,500 L ha^{-1}), PPCPs moved rapidly (within minutes) *via* soil macropores to tile drains (PPCPs concentration 10^1 to 10^3 ng/l). The authors noted that liquid municipal biosolids were spread and were applied in the wet season. Following from the Lapen study, Edwards *et al.* (2009) studied the fate of PPCPs following the application of dewatered biosolids on the same agricultural land one year after the Lapen study. The results show that concentrations of PPCPs were generally lower in the dewatered biosolids; this shows that the characteristics of the biosolids may influence the transport potential following land application. Similarly Xia *et al.* (2010) evaluated the levels of four organic contaminants in soils from field plots receiving annual applications of biosolids for 33 years. All four contaminants were detected in most of the biosolid concentrations ranging from hundreds of μg/kg to over 1000 mg/kg (dry weight basis). As the biosolid application rate increased, contaminant concentration increased. Three of the contaminants (TCC, TCS, and 4-NP) showed a rapid transformation and degraded in the soil under natural field conditions; however the fourth contaminant (polybrominated diphenyl ether [PBDE]) was slow to degrade and may result in accumulation.

Detection of contaminants in surface waters following land application of biosolids have been widely reported (Miao *et al.* 2005; Smith 2009; Gottschall *et al.* 2013). Sabourin *et al.* (2009) reported maximum concentrations of contaminants in runoff ranging from below detection limits to 109.7 ng/l following application of dewatered biosolids to agricultural land. Chari and Halden (2012) reported the detection of the hormones 17β estradiol and estrone in surface waters >30 days after land application of biosolids. Toxicity studies have shown adverse effects of steroidal hormones, even at low concentrations (~100 to >1000 ng^{-1}), on fish and aquatic invertebrates (Gottschall *et al.* 2013). Mompelat *et al.* (2009) also demonstrated that in Europe there are water quality and safety issues regarding the amount of pharmaceuticals detected in surface and drinking waters.

Due to their confirmed presence in biosolids-amended soils, the antimicrobials triclosan and triclocarban may have the potential to bioaccumulate in soil-dwelling organisms and thus exert toxic effects on higher organisms through trophic transfer (Higgins *et al.* 2011). Recent toxicological reports have shown that triclocarban has the potential to disrupt excitation-contraction coupling in skeletal and cardiac muscles in humans (Gautam *et al.* 2014). Triclocarban has been linked to endocrine disruption by amplifying androgen receptor-mediated activity in rats, and has also caused methemoglobinemia in humans exposed to boiled water containing 1.1% of triclocarban, which formed a primary aromatic amine (Palenske 2009). Recent reports have revealed that triclosan is capable of interfering with various hormones as a weak endocrine disruptor in multiple species, as well as impairing muscle contraction (Yueh *et al.* 2014). The results of these studies raise toxicological and public concerns at the lack of knowledge of the potential health effects associated with direct water ingestion. Hogenboom *et al.* (2009) reported concerns that such contamination can become an increasing problem for drinking water supplies, especially since the European Reach Legislation (Commission Regulation (EU) No. 1272/2013) may drive producers to develop newly designed, less lipophilic/bioaccumulative chemicals that may lead to higher mobility of the chemicals in aqueous media. Furthermore, the demands on drinking water treatment companies to remove hydrophilic compounds that are inherently more difficult to remove by traditional drinking water treatment techniques will be a challenge (Schriks *et al.* 2010). Most of the substances detected lack toxicity data to derive safe levels and have not yet been regulated (Mons *et al.* 2013). At present, the World Health Organisation (WHO; 2011) has derived approximately 125 statutory guideline values for contaminants in drinking water.

In addition to pharmaceuticals, emerging POPs such as PFOS and PFOA have frequently been detected in drinking water (Loos *et al.* 2007; Xiao *et al.* 2013). PFOS and PFOA are members of a large family of perfluorinated chemicals (PFCs), and their use has included performance chemicals such as surfactant in firefighting foam and emulsifiers in floor polish (Rumsby *et al.* 2009). Most of the environmental release is to water (98%) and the remainder is to air. They are immobile in soil and are non-biodegradable in sewage sludge (Rumsby *et al.* 2009). PFOS and PFOA detected in drinking water have become a significant concern to human health. Levels of PFOA and PFOS have been detected in drinking water at 2.4 and 8.1 ng/l, respectively (Loos *et al.* 2007). It has also been suggested that ongoing exposure to PFOA in drinking water at levels of 10 ng/l or 40 ng/l can increase serum PFOA levels by approximately 25% and 100%, respectively, from the general population background serum level of ~4 ng/ml (Olsen *et al.* 2003). The association of blood/serum PFOS and/or PFOA levels with hyperuricemia and with children attention deficit/hyperactivity disorder and lowered immune response to vaccinations have been reported (Olsen *et al.* 2003; Post *et al.* 2009; Steenland *et al.* 2010). As yet, there are no drinking water quality standards in the European Union; however, following incidents of pollution, some member states have issued guidance based on human health effects for PFOS and PFOA if found in drinking water. For example, the drinking water inspectorate in England and Wales has implemented a four tier system for drinking water companies to adopt for the monitoring and management of PFOS and PFOA in drinking water supplies. The inspectorate provides guidance on the levels of PFOS and PFOA that water companies should act upon to fulfil their statutory obligations to ensure the safety of drinking water. The drinking water commission in Germany also have issued guidance on the maximum values for PFOA and PFOS combined concentrations in drinking water with values ranging from 0.1 μg L^{-1}–5 μg L^{-1} (Rumsby *et al.* 2009).

The widespread use of the surfactant NP and its short ethoxy chain precursors, NP1EO and NP2EO, has led to the detection of these contaminants in many environmental matrices such as water, sediment, air, and soil (Mao *et al.* 2012). Due to their physical–chemical characteristics such as high hydrophobicity and low solubility, NP, NP1EO, and NP2EO accumulates in the environmental compartments that are considered high in organic content, such as sewage sludge (Soares *et al.* 2008). Vikelsøe *et al.* (2002) reported significant concentrations of NP in soil samples (1450 μg/kg dw) exposed to high amounts of sludge applied (17 t dw/ha/yr) over a 25-year period. The soil was sampled twice with a 2-year interval and showed that concentrations of NP increased to 2430 μg/kg dw. This was compared to a low and normal sludge application rate showing NP concentrations of 0.04 and 0.01 μg/kg dw, respectively. The concentration of NP in surface runoff was 34 μg/kg dw following high sludge application.

NP, NP1EO, and NP2EO have been reported to exert a number of estrogenic responses on aquatic organisms, and have therefore been classified as endocrine disruptors by the WHO (WHO 2004; Gatidou *et al.* 2007). Exposure to the endocrine disruptor NP and its short ethoxy chain precursors, NP1EO and NP2EO, have been extensively studied in freshwater organisms, and can result in hermaphroditism, developmental abnormalities, reduction in larval survival and changes in the sex ratio in females which were found in the Pacific oyster, *Crassostrea gigas*, at concentrations as low as 1-100 μg/l (Vazquez-Duhalt *et al.* 2005). Low level exposure of NP to freshwater organisms can cause severe reproductive disorders ($<0.1\mu$g/l) and even death ($LC_{50} = 0.1$–5 mg/l; Roberts *et al.* 2006). However, NP and its ethoxylates were designated as priority hazardous substances (PHS) in the Waste Framework Directive 2000/60/EC (EC 2000) and most of their uses are currently regulated (Soares *et al.* 2008).

Although concentrations of NP have been detected at concentrations up to 55.3 μg/l in drinking water, adverse consequences from human exposure to NP, NP1EO, and NP2EO can take a variety of forms, including immuno-regulatory properties that can be crucial for normal foetal development (Vazquez-Duhalt *et al.* 2005). NP may be capable of inducing breast tumour cell proliferation and has the ability to mimic the natural hormone 17βestradiol by competing for the binding site for the receptor for the natural oestrogen (Soares *et al.* 2008). In 2008, the (WFD 2015) placed nonylphenols on a list of priority hazardous substances for which environmental quality standards were set. The threshold values of 0.3 μg/l in drinking water for annual average and the maximum allowable concentrations (2 μg/l) for surface waters were set (Water Framework Directive 2015).

Although certain POPs emissions are restricted under the Stockholm Convention (UN/EP 2001) because of persistent, bioaccumulative, and toxic properties in humans and wildlife, many researchers still report “legacy” POPs in environmental media around the world. Zennegg *et al.* (2013) recently reported temporal trends of POPs in biosolids. In their study, selected POPs were measured in biosolids between 1993 and 2012 from eight different wastewater treatment plants in Switzerland. There was a decreasing trend in biosolids of dioxin-like POPs (PCBs and PCDD/Fs) with a half-life of 9 to 12 years. However, there was no reduction in PBDEs, PFOAs and PFOSs. This study is indicative of the restrictions and exclusion of POPs (*e.g.*, PCBs, PCDD/Fs) in the last two decades and their reduction in the environment, however, recently restricted or excluded POPs (*e.g.*, PBDEs, PFOAs, and PFOSs) have yet to show a reducing trend.

The primary objective of this study is to develop a quantitative human health risk ranking model for "legacy" (*e.g.*, dioxin-like compounds) and EC in biosolids that are reused in Irish agriculture. All contaminants will be ranked to identify those that may pose the greatest risk to surface water, groundwater, drinking water and, ultimately, human health.

Methods and materials

Risk assessment

Risk assessment is a process of identifying potential adverse consequences along with their severity and likelihood (Jenkin *et al.* 2007). A probabilistic model adopted from Trevisan *et al.* (2009) was constructed to estimate human exposure to organic contaminants that are contained within biosolids destined for grassland application. The model includes four major compartments (Figure 1): concentration in top soil (PEC_{soil}), surface runoff (PEC_{runoff}), groundwater ($PEC_{groundwater}$), and the level of chemical intake or human exposure (HE). The chemical intake toxicity ratio (RR) (ratio of the measure of the effects (LC_{50}) to the estimated exposure) was used to model human risk. The probabilistic approach provides a complete scenario of potential organic contaminant exposures, compared to use of a single set of discrete inputs.

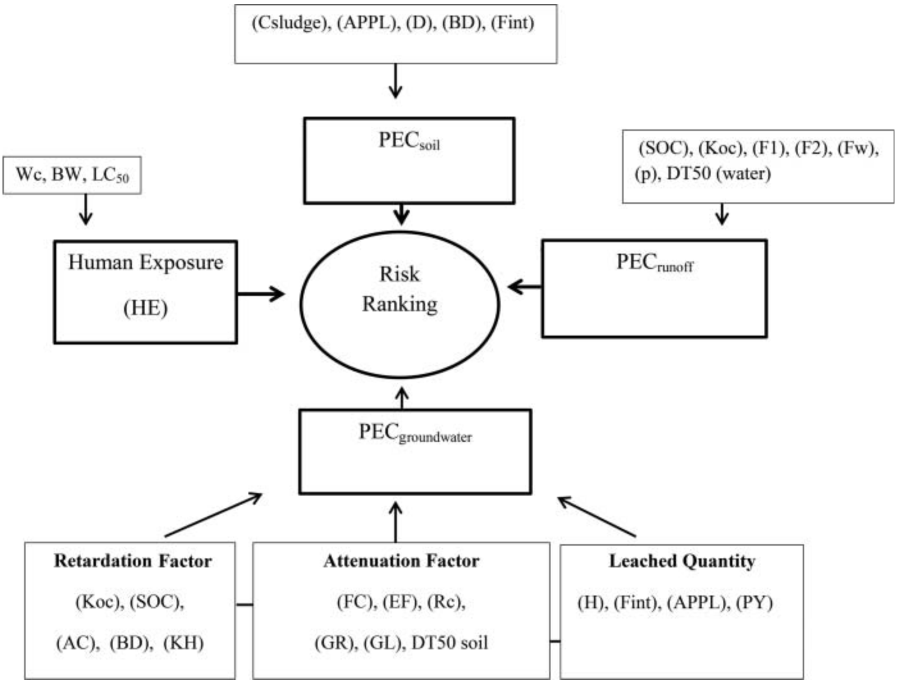

Figure 1. Flow diagram of inputs and outputs for the quantitative risk ranking process for organic contaminants. Note: Key for parameters can be found in Table 2.

Contaminants of concern

In order to identify hazardous contaminants it is important to know the fate and behavior of contaminants in the environment (*e.g.*, persistence, bioaccumulative, and toxicity). There are currently knowledge gaps regarding the toxicity and environmental fate for a considerable number of contaminants on the market. The contaminants analyzed in this study comprise a group of organic contaminants belonging to various categories and chosen based on persistence, bioaccumulation, and toxicity (PBT). The selected contaminants include dioxin-like compounds (polychlorinated biphenyls [PCBS], polychlorinated dibenzo-p-dioxin furrans [PCDD/Fs] and flame retardant [PBDEs], PPCPs [carbamazepine, triclosan, triclocarban, propranolol, and metoprolol], perfluorooctane sulfonate [PFOS] and perfluorooctanoate [PFOA] substances, natural hormones [estrone and estradiol], surfactants [nonylphenol (NP], its short ethoxy chain precursors nonylphenol mono-(NP1EO), and di-ethoxylate (NP2EO)), and the industrial chemical, bisphenol A (BPA). The chemical properties of all organic contaminants are presented in Table 1. Compounds that are expected to remain in the soil at the origin of application (Log Koc >3.5) are of exceptional concern, given the effectively longer organism exposure periods and the potential for increasing soil concentrations with repeated applications (Snyder *et al.* 2011).

Table 1. Chemical properties of organic contaminants.

				Half-life (DT50)	
Contaminant	Cas. no	Formula	Log Kow	Soil	Water
Dioxin-like compounds					
Polychlorinated biphenyl (PCBs)	1336-36-3	C12H5Cl5	4.53-8.35	360 d	180 d
Polychlorinated dibenzo dioxins/ furrans (PCDD/Fs)	Varied	C12H4Cl4O2 C12H4Cl4O2	5.6-8.1	360 d	180 d
Polybrominated diphenyl ethers (PBDEs)	Varied	C12H(10−x,y)Brx,yO	5.53-8.58	360 d	180 d
PPCPs					
(*Anti-epileptic*)					
Carbamazepine	298-46-4	C15H12N2O	2.25	75 d	38 d
(*Antimicrobial agents*)					
Triclosan	3380-34-5	C12H7Cl3O2	4.76	180 d	60 d
Triclocarban	101-20-2	C13H9Cl3N2O	4.2	120 d	60 d
(*Beta-blockers)*					
Propranolol	318-98-9	C16H21NO2 · HCl	3.48	30 d	15 d
Metoprolol	51384-51-1	C15H25NO3	1.88	75 d	15 d
Perfluorochemicals					
Perfluorooctane sulphonate (PFOS)	2795-39-3	C8F17SO3	4.88	360 d	180 d
Perfuorooctanoic acid (PFOA)	335-67-1	C8HF17O3S	6.30	360 d	180 d
Steroids					
Estrone E1	53-16-7	C18H22O2	3.13	75 d	38 d
Estradiol E2	50-28-2	C18H24O2	4.01	75 d	38 d
Alkyphenols					
NP	Varied	C15H24O	5.76	30 d	15 d
NP1EO	27986-36-3	C17H28O2	4.48	75 d	38 d
NP2EO	20427-84-3	C18H30O3	4.17	120 d	60 d
Industrial chemical					
Bisphenol A (BPA)	80-05-7	C15H16O2	4.20	75 d	38 d

All data was obtained from PBT profiler (USEPA 2012).

Environmental fate model

In Ireland, over 82% of the population (4.5 million) (CSO 2014) rely on the public water supply for drinking water (Ireland EPA 2014). This water is sourced from rivers, lakes, and reservoirs, while groundwater accounts for between 20–25% of the drinking water supply (GSI 2014). The risk of organic contaminant leaching into surface water, groundwater, or lakes from land spreading of biosolids may be estimated by the predicted environmental concentration (PEC) models (soil, surface water, and groundwater), adopted from Trevisan *et al.* (2009), which was originally designed for the PECs of pesticides in ground and surface water in Germany. In the present study the approach developed by Trevisan and Padovani (Padovani *et al.* 2004; Trevisan *et al.* 2009), is modified for use in Irish conditions (*e.g.*, application rates, rainfall rates, bulk density, slope, and soil organic carbon [SOC]).

With regard to organic contaminants, there is concern when elevated levels are released into the environment or cause harm to humans. Therefore, the environmental fate of each contaminant selected was assessed using parameters such as the initial concentrations in biosolids potentially spread on agricultural grassland, the concentration in soil post application, potential subsequent runoff from land-spreading area into surface water, leaching into groundwater, consumption of water, and overall toxicity. Each parameter can be used as a basis to rank the human health risk from the consumption of contaminated water. Risk ranking can be used to prioritise substances for focused risk management (Labite and Cummins 2012). A flow diagram of the model processes are given in Figure 1.

Predicted environmental concentrations

PEC_{soil}

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 predicted concentration in the soil (PEC_{soil}; mg/kg) was calculated based on the concentration of the contaminant in the biosolids, application rate, mixing depth of soil and the soil bulk density following biosolids application (Trevisan *et al.* 2009):

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

where C_{sludge} is the concentration of the contaminant of interest in biosolids (g/m^2), APPL is the application rate of biosolids on agricultural land for one application (g/m^2), f_{int} is the fraction intercepted by the crop (-), D is the depth (m), and BD is the soil bulk density (kg/m^3).

Irish and European organic contaminant concentrations in biosolids were sourced from peer reviewed journals and probabilistic distributions were fitted (Table 2) to characterize uncertainty/variability. 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 *et al.* (2013). It was assumed that the biosolids were spread on grassland. The fraction intercepted by the crop was based on tabular interception fractions values as proposed by Linders *et al.* (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 ($F_{int} + F_{soil} = 1$). The study focuses on interception rather than

Table 2. Model inputs, distributions, and outputs.

Stage	Symbols	Description	Model/distribution	Units
		PEC_{soil}		
	C_{sludge}	Concentration in biosolids	Uniform or triangular (contaminant specific) (Table 3)	$\mu g/kg$
	APPL	Application rate	Triangular (300,330,520)	g/m^2
	D	Depth	0.02	m
	BD	Bulk density	Uniform (800,1000)	kg/m^3
	f_{int}	Crop interception	Triangular (0,10,20)	%
Output	PEC_{soil}		$C_{sludge} \times$ APPL $\times$ (1- fint) / (100 $\times$ BD $\times$ D)	mg/kg
		PEC_{runoff}		
	Koc	Soil adsorption coefficient	Contaminant specific (Table 3) (Uniform, triangular, normal)	m^3/kg
	SOC	Soil organic carbon content	Cumulative (min 1.4, max 55.8), [2.86,3.56,4.92,7,14, 26,40.82,48] [0.02,0.03,0.04,0.07, 0.14,0.4,0.4,0.5]	%
	f_1	Correction factor	$0.02153 \times SL + 0.001423 \times SL^2$	
	SL	Slope	6.7	%
	z	Water body distance	0.02	m
	f_2	Effect of water body distance	$0.83^{0.02}$	
	DT50	Half-life in water	Uniform (contaminant specific)	d
	f_w	Fraction of contaminant available for transport *via* runoff	$100 \times e^{-\Delta t Ln2/DT50} / 1 + Koc \times SOC$	–
	P	Maximum daily precipitation	Fixed value	mm/d
Output		PEC_{runoff}	APPL $\times$ (1- fint) $\times f_1 \times f_2 \times f_w$ / P	$\mu g/l$
		$PEC_{groundwater}$		
	K_H	Henry's constant	Contaminant specific (Table 3)	–
	Sc	Sand content	47	%
	Cc	Clay content	19	%
	OM	Organic matter	(Conversion SOC $\times$ 1.724)	%
	FC	Soil field capacity	$= 0.3486 - 0.018 \times \%\, Sc + 0.0039 \times \%\, Cc + 0.228 \times OM - 0.0738 \times BD / 1000$	cm^3/cm^3
	PD	Particle density	Fixed value (2650)	kg/m^3
	P	Porosity	1- (BD $\times$1000 / PD)	–
	AC	Air content	FC – P	–
	RF	Retardation factor	$= 1 + (BD \times Soc \times Koc/1000 / FC) + (AC \times K_H / FC)$	
	P	Precipitation	Uniform (750,1500)	mm/yr
	E	Evapotranspiration	Fixed value	mm/yr
	ER	Effective rainfall	P - ER	–
	GL	Groundwater level	Uniform (0.5, 2.5)	m
	RC	Recharge coefficient	Triangular (0.4,0.7,1)	–
	Q	Net recharge of water table	ER $\times$ RC	m/yr
	TR	Average residence time	GL $\times$ RF $\times$ FC / Q	L/yr

(*Continued*)

Table 2. (*Continued*)

Stage	Symbols	Description	Model/distribution	Units
	DT50	Half-life in soil	Contaminant specific (uniform, triangular)	d
	AF	Attenuation factor	EXP (- 0.693) × TR / DT50	–
	H	Thickness of water table	Fixed value (2.2)	m
Output	LQ	Leached quantity	2.739 × AF × APPL × (1-fint) / P × H	μg/l
		Human exposure		
	Wc	Water consumption	Adult Log normal (0.564,0.617), Child Log normal (0.238,0.208)	L
	BW	Body weight	Adult normal (78.1,16.5), Child normal (33,11.3)	Kg
	HE	Human exposure	Wc × $\text{PEC}_{\text{runoff}}$ / BW Wc × $\text{PEC}_{\text{groundwater}}$ / BW	μg/kg bw/d
	LC_{50}	Median lethal concentration	Contaminant specific (Uniform, triangular)	Mg/l
Risk Ratio (LC_{50})	RR	Model Outputs	LC_{50} / $\text{PEC}_{\text{runoff/groundwater}}$	

retention. Conceptually, it is assumed that both interception and deposition on soil are instantaneous processes. The interception values are related to the growth phases as defined in the BBCH (Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie) scale. The reasoning behind the proposed standard growth phase specific interception fractions per crop is the rapid acquirement of intercepted fractions that would otherwise be tedious and costly experiments. The estimates reflect the phenological development of a crop in time by taking all major growth phases into account. Crop interception can vary depending on the phenological stages (*i.e.*, bare soil/emergence has a value of 0 cm and potatoes >50 cm have an interception fraction of 89% (Trevisan *et al.* 2009). Crop interception (f_{int}) was estimated to be a triangular distribution (minimum 0, most likely 10 and maximum 20%) assuming a worst-case scenario. The mixing depth (0.02 m) was obtained from Padovani *et al.* (2004) and the BD of soil (800– 1000 kg/m^3) (uniform distribution) were obtained from Vero *et al.* (2014), which is a typical range for the upper 100 mm of the soil profile of Irish grasslands. It was assumed that the application rates incorporated into this research were applied according to the legal maximum rate to be applied to a phosphorus index 1 soil (S.I.610 of 2010), based on dry solid content of amendment.

PEC_{runoff}

The predicted concentration of organic contaminants in surface water due to runoff at a delivery point to surface water ($\text{PEC}_{\text{runoff}}$) retrieved from Trevisan *et al.* (2009) and modified to suit Irish conditions in this study, was calculated by:

$$\text{PEC}_{\text{runoff}} = \text{APPL} \times (1 - f_{int}) \times f_1 \times f_2 \times f_w \,/\, \text{P} \tag{2}$$

where f_1 is a correction factor that considers the field slope (6.7% based on an Irish field

Table 3. Organic contaminant input data.

Organic contaminant	Distribution	Min	Median	Max	References
Dioxin-like compounds					
PCBs					
Log Koc (L/kg)	Triangular[a]	0.01	4	6	[a] (Swackhamer and Armstrong 1987; Chin *et al.* 1988; Hansen *et al.* 1999; Jäntschi 2004; Rodenburg *et al.* 2010)
Csludge (mg/kg)	Uniform[b]	9.2		277	[b] (Stevens *et al.* 2003; Abad *et al.* 2005; Dąbrowska and Rosinska 2012; Jensen *et al.* 2012)
LC$_{50}$ (mg/l)	Uniform[c]	0.9		2.8	[c] (Nebeker and Puglisi 1974; Mayer *et al.* 1977; Veith *et al.* 1979; Foekema *et al.* 2008)
Henry's law constant (K_H)	Uniform[d]	3.40E-03		6.90E-01	[d,e] (Christensen and Li 2014; Chemspider 2015)
DT50 half-life (days)-water	Uniform[e]	37.5		180	[e,f] (USEPA PBT 2012)
-soil	Uniform[f]	360		940	
PCDD/F					
Log Koc (L/kg)	Triangular[a]	5.2	7.1	8.1	[a] (Fiedler 2003)
Csludge (mg/kg)	Triangular[b]	0.005	0.8	2.4	[b] (McLachlan *et al.* 1996; Oleszek-Kudlak *et al.* 2005; Smith 2009)
LC$_{50}$ (mg/l)	Triangular[c]	0	0.3	1.1	[c] (Basmaa 2004; National Dioxins Programme 2004; Chem-UNEP 1996)
Henry's law constant (K_H)	Uniform[d]	2.70E-02		1.20E-01	[d] (Cohen *et al.* 2002)
DT50 half-life (days)-water	Uniform[e]	5		90	[e,f] (USEPA PBT 2012; Chemspider 2015)
-soil	Uniform[f]	395		708	
Fire retardant					
PBDEs					
Log Koc (L/kg)	Normal[a]	5.1		0.9	[a] (Knoth *et al.* 2007; Eljarrat *et al.* 2008)
Csludge (mg/kg)	Triangular[b]	53.6	467	1,295	[b] (Hellström 2000; Key *et al.* 2009; Usenko *et al.* 2011)
LC$_{50}$ (mg/l)	Uniform[c]	0		6.8	[c] (NICNIS 2007; Bramwell *et al.* 2014)
Henry's law constant (K_H)	Uniform[d]	3.30E+03		1.70E+02	[d] (Sander 1999)
DT50 half-life (days)-water	Uniform[e]	90		150	[e,f] (USEPA PBT 2012; Chemspider 2015)
-soil	Uniform[f]	1440		3600	
Pharmaceuticals and personal care products (PPCPs)					
Carbamazepine					
Log Koc (L/kg)	Normal[a]	2.6		0.7	[a] (Scheytt *et al.* 2005, 2006)
Csludge (mg/kg)	Uniform[b]	0		1,300	[b] (Miao *et al.* 2005; Díaz-Cruz *et al.* 2009)
LC$_{50}$ (mg/l)	Triangular[c]	0	41	61	[c] (Kim *et al.* 2007, 2009)
Henry's law constant (K_H)	Uniform[d]	5.60E-02		3.90E-07	[d,e] (Chemsider 2015; Kummerer 2013)
DT50 half-life (days)-water	Uniform[e]	37.5		38	[f] (Walters *et al.* 2010; USEPA PBT profiler 2012)
-soil	Uniform[f]	75		495	

(Continued)

Table 3. (*Continued*)

Organic contaminant	Distribution	Min	Median	Max	References
Triclosan					
Log Koc (L/kg)	Normal[a]	4.1		0.3	[a](Barron *et al.* 2009; Agyin-Birikorang *et al.* 2010; Chen *et al.* 2011; WFD 2012; Gasperi *et al.* 2014)
Csludge (mg/kg)	Triangular[b]	7.5	4859	14,400	[b](Walters *et al.* 2010; Clarke and Smith 2011; Davis *et al.* 2012) [c](Orvos *et al.* 2002; Santa Cruz Biotechnology 2009; Wirud-Ingredients 2015)
LC$_{50}$ (mg/l)	Uniform[c]	0.005		0.9	[d](Thompson *et al.* 2005)
Henry's law constant (K_H)	Uniform[d]	5.20E-04		1.30E-03	[e](Chemsider 2015)
DT50 half-life (days)-water	Uniform[e]	0		60	[f](Wu *et al.* 2009)
-soil	Uniform[f]	87		231	
Triclocarban					
Log Koc (L/kg)	Triangular[a]	0	3.1	4.6	[a](Ying *et al.* 2007; Cha and Cupples 2010; King 2010)
Csludge (mg/kg)	Triangular[b]	0	14,113	39,628	[b](Chu and Metcalfe 2007; Snyder *et al.* 2010; McClellan and Halden 2010)
LC$_{50}$ (mg/l)	Triangular[c]	0.003	0.08	0.1	[c](Palenske 2009; Higgins *et al.* 2011; Maruya *et al.* 2014)
Henry's law constant (K_H)	Uniform[d]	3.60E-05		8.30E-06	[d,e](USEPA PBT profiler 2012; Chemspider 2015)
DT50 half-life (days)-water	Uniform[e]	0		60	[f](Ying *et al.* 2007; Walters *et al.* 2010)
-soil	Uniform[f]	18		120	
Propranolol					
Log Koc (L/kg)	Uniform[a]	2.3		3.9	[a](Maurer *et al.* 2007; Morais *et al.* 2013; Burke *et al.* 2013)
Csludge (mg/kg)	Triangular[b]	0.002	0.3	0.96	[b](Lee and Choi 2007; Radjenovic *et al.* 2009; Barron *et al.* 2010)
LC$_{50}$ (mg/l)	Triangular[c]	0	10	30	[c](Stanley *et al.* 2006; Verlicchi *et al.* 2012)
Henry's law constant (K_H)	Uniform[d]	2.90E-09		5.10E-07	[d](Toxnet 2014a)
DT50 half-life (days)-water	Uniform[e]	0		15	[e](Schnaak *et al.* 1997; Caminada *et al.* 2006)
-soil	Uniform[f]	0		30	[f](Chemspider 2015)
Metoprolol					
Log Koc (L/kg)	Uniform[a]	1.2		2.4	[a](Maurer *et al.* 2007; Morais *et al.* 2013; Burke *et al.* 2013)
Csludge (mg/kg)	Triangular[b]	0.03	0.3	0.8	[b](Ternes 1998; Barron *et al.* 2008)
LC$_{50}$ (mg/l)	Uniform[c]	50		170	[c](Sun *et al.* 2014; USP 2011; American Reagent 2010)
Henry's law constant (K_H)	Uniform[d]	5.00E-10		3.60E-08	[d](Vieno *et al.* 2007)
DT50 half-life (days)-water	Uniform[e]	0.3		7.5	[e,f](USEPA PBT profiler 2012; Chemspider 2015)
-soil	Uniform[f]	0		54	

(*Continued*)

Table 3. (*Continued*)

Organic contaminant	Distribution	Min	Median	Max	References
Industrial chemical					
BPA					
Log Koc (L/kg)	Triangular[a]	2.3	2.8	3.9	[a](Clara *et al.* 2004; Ivashechkin *et al.* 2004; Cunha *et al.* 2012; Yu *et al.* 2013)
Csludge (mg/kg)	Uniform[b]	79.1		1778.9	[b](Lee and Peart 2002)
LC$_{50}$ (mg/l)	Uniform[c]	0.002		12.2	[c](CEPA 2009; Faheem and Lone 2013; Hussain and Mirza 2013)
Henry's law constant (K_H)	Uniform[d]	3.60E-08		1.60E-07	[d](Toxnet 2014b; Guidechem 2015)
DT50 half-life (days)-water	Uniform[e]	0.5		3	[e](Klecka *et al.* 2001)
-soil	Uniform[f]	3		37.5	[f](Flint *et al.* 2012)
Perfluorochemicals					
PFOS					
Log Koc (L/kg)	Uniform	2.1		4.9	[a](Zareitalabad *et al.* 2013)
Csludge (mg/kg)	Lognormal	9.2		15.4	[b](Ye *et al.* 2009; Zheng *et al.* 2012)
LC$_{50}$ (mg/l)	Uniform	5.2		107	[c](Ji *et al.* 2008)
Henry's law constant (K_H)	Uniform	1.10E-05		4.90E-04	[d](Brooke *et al.* 2004; Benford *et al.* 2008)
DT50 half-life (days)-water	Uniform	180		285	[e](Environment Canada 2013)
-soil	Uniform	0		360	[f](USEPA PBT profiler 2012)
PFOA					
Log Koc (L/kg)	Triangular[a]	1.3	2.5	5	[a](Zareitalabad *et al.* 2013)
Csludge (mg/kg)	Triangular[b]	0	75	225	[b](Jensen *et al.* 2012)
LC$_{50}$ (mg/l)	Uniform[c]	300		1005	[c](Ji *et al.* 2008; Ye *et al.* 2009; Zheng *et al.* 2012)
Henry's law constant (K_H)	Uniform[d]	3.70E+00		3.30E+02	[d](Harrad 2009)
DT50 half-life (days)-water	Uniform[e]	0		180	[e,f](USEPA PBT profiler 2012; Zheng *et al.* 2012)
-soil	Uniform[f]	0		360	
Steroids					
17β-estradiol					
Log Koc (L/kg)	Triangular	2.9	3.8	5.6	[a](Zoetis 2014)
Csludge (mg/kg)	Triangular	6.9	21	36	[b](Combalbert and Hernandez-Raquet 2010)
LC$_{50}$ (mg/l)	Uniform	0.4		3.5	[c](Kang *et al.* 2002; EU Council 2011)
Henry's law constant (K_H)	Uniform	2.80E-08		3.10E-08	[d](NIEHS 2014; Chemspider 2015)
DT50 half-life (days)-water	Uniform	3		46	[e](Nagpal and Meays 2009)
-soil	Uniform	0		3	[f](USEPA PBT profiler 2012)
Estrone					
Log Koc (L/kg)	Uniform[a]	2.7		5.6	[a](Pfizer 2007; Snyder and Snyder 2009)
Csludge (mg/kg)	Triangular[b]	0	12	36	[b](Combalbert and Hernandez-Raquet 2010; Clarke and Smith 2011)
LC$_{50}$ (mg/l)	Uniform[c]	0		12	[c](Kang *et al.* 2002; Kashiwada *et al.* 2002; EU Council 2011)

(*Continued*)

Table 3. (*Continued*)

Organic contaminant	Distribution	Min	Median	Max	References
Henry's law constant (K_H)	Uniform[d]	4.80E-09		1.30E-06	[d](Chemspider 2015; National Center for Biotechnology Information nd)
DT50 half-life (days)-water	Uniform[e]	2		6	[e,f](Ying *et al.* 2002)
-soil	Uniform[f]	2		3	
Alkyphenols					
NP					
Log Koc (L/kg)	Triangular[a]	0.02	3.6	5.4	[a](Patrolecco *et al.* 2006; Shchegolikhina *et al.* 2014; Milinovic *et al.* 2015)
Csludge (mg/kg)	Triangular[b]	721,79	295,189	870,224	[b](La Guardia *et al.* 2009; Roig *et al.* 2012; Sánchez-Trujillo *et al.* 2014; Jones *et al.* 2014)
LC_{50} (mg/l)	Uniform[c]	0		1	[c](Hemmer *et al.* 2001; Kashiwada *et al.* 2002; WFD 2005)
Henry's law constant (K_H)	Uniform[d]	5.60E-02		1.40E-01	[d](Porter and Hayden 2002)
DT50 half-life (days)-water	Uniform[e]	8.2		16.5	[e](USEPA 2005)
-soil	Uniform[f]	4.5		37	[f](Marcomini *et al.* 1988)
NP1EO					
Log Koc (L/kg)	Uniform[a]	4		5.4	[a](Patrolecco *et al.* 2006)
Csludge (mg/kg)	Exponential[b]	14.2		802,042	[b](La Guardia *et al.* 2009)
LC_{50} (mg/l)	Uniform[c]	0.1		1	[c](Hemmer *et al.* 2001; Kashiwada *et al.* 2002; WFD 2005; Soares *et al.* 2008)
Henry's law constant (K_H)	Uniform[d]	5.80E-04		1.20E-03	[d,e](USEPA PBT profiler 2013; Chemspider 2015)
DT50 half-life (days)-water	Uniform[e]	0.3		7.5	[f](Marcomini *et al.* 1988)
-soil	Uniform[f]	7		360	
NP2EO					
Log Koc (L/kg)	Uniform[a]	0.001		6.5	[a](Aparicio *et al.* 2009)
Csludge (mg/kg)	Triangular[b]	19.3	148	928	[b](La Guardia *et al.* 2009; González *et al.* 2010)
LC_{50} (mg/l)	Triangular[c]	0.08	0.34	0.8	[c](TenEyck and Markee 2007)
Henry's law constant (K_H)	Uniform[d]	9.30E-06		3.80E-05	[d](Chemspider 2015)
DT50 half-life (days)-water	Uniform[e]	0		60	[e,f](ECHA 2013; Chemspider 2015)
-soil	Uniform[f]	8		360	[f](Marcomini *et al.* 1988)

study by Brennan *et al.* 2012) and estimated as:

$$f_1 = 0.02153 \times \text{Slope} + 0.001423 \times \text{Slope}^2 \quad \text{if slope} < 20\% \tag{3}$$

$f_1 = 1$ if slope $\geq 20\%$

f_2 is an empirical factor that takes into account the effect of the surface water body distance (z) and is calculated as:

$$f_2 = 0.83^z \tag{4}$$

f_w is the fraction of the contaminant available for transport *via* surface runoff (%), calculated as:

$$f_w = 100 \times e^{-\Delta \mathrm{t2/DT50}} / 1 + \mathrm{Koc} \times \mathrm{SOC} \tag{5}$$

where DT_{50} is the soil half-life of the selected contaminant in soil (days). Soil half-life is contaminant specific and based on mean DT_{50} values. A uniform distribution was used to model uncertainty in the data. Koc is the organic carbon-soil sorption coefficient (m^3/kg) (contaminant specific). The type of distributions applied to represent the uncertainty regarding Koc depended on the data collected for each individual contaminant, therefore uniform, triangular, and normal distributions were used to model Koc uncertainty (Table 3). SOC is the soil organic carbon (%). To capture the uncertainty in the data, a continuous empirical distribution in the form of a cumulative distribution ranging from 1.40 to 55.8% was used (Table 2) and were based on data from Fay *et al.* (2007).

Finally, P is the maximum daily precipitation (mm). The average maximum daily precipitation for the past 30 years in Ireland, obtained from meteorological data, was 84 mm/day (Met Eireann 2014).

Peer reviewed literature sources were examined for log Koc values for selected contaminants as each Koc is contaminant specific. Where Koc values were not available, regression equations from Lynam *et al.* (1990) were used to estimate log Koc based on *log* Kow which is defined as the ratio of a chemical's concentration in the octanol phase to its concentration in the aqueous phase of a two-phase octanol water system (USEPA 2009a). The Koc was calculated by:

$$\mathrm{Log}\,(\mathrm{Koc}) = 0.544 \times \log\,(\mathrm{Kow}) + 1.377 \tag{6}$$

To represent uncertainty in the data, a distribution was applied which was uniform, triangular or normal (contaminant specific) (Table 3).

Soil organic carbon content and Koc are two of the major factors influencing the mobility of organic contaminants (Labite and Cummins 2012). Increased SOC content influences the Koc, and will most probably reduce leaching and plant uptake, which will in turn increase the fraction of contaminant that accumulates in the soil (Sundstøl Eriksen *et al.* 2009). The database of SOC in Ireland was recently updated through the National Soil Database Project (Fay *et al.* 2007). A cumulative distribution was assigned with values ranging from 1.40% to 55.8% (Table 2). A correlation matrix was created so that the Koc and SOC inputs were not independent but correlated with each other. The dependence between the SOC and Koc was confined by implementing a uniform correlation coefficient with a minimum of 0.8 and a maximum of 0.95. These figures are based on studies by Barriuso *et al.* (1991) and Zhao *et al.* (2006). A previous study by Grathwohl (1990) reported similar results of the positive correlation between SOC and Koc, indicating that the SOC greatly influences the sorption of hydrophobic organic compounds in soil (Yang *et al.* 2013).

$PEC_{groundwater}$

The predicted environmental concentration of each contaminant in groundwater was calculated according to Trevisan *et al.* (2009) and Padovani *et al.* (2004), and was adapted for Irish conditions. The model incorporates the retardation factor (RF), average residence time (TR) in soil and subsoil, the attenuation factor (AF) and the leached quantity (LQ) (Figure 1). The RF represents the delay in the contaminant leaching with regard to the migration through the unsaturated zone (*i.e.*, soil). This leaching delay is due to contaminant sorption to soil and depends on the soil chemistry, contaminant gaseous and aqueous diffusion in soil (Paraiba and Spadotto 2002). The RF in this study was calculated according to Padovani *et al.* (2004) and is calculated by:

$$RF = 1 + (BD \times SOC \times Koc\ /\ FC) + (AC \times K_H\ /\ FC) \tag{7}$$

where FC is the soil field capacity (cm^3/cm^3), Koc is the soil sorption coefficient (cm^3/g), AC is the soil air content (%) and K_H is Henry's constant (-)

The soil field capacity is defined as the maximum amount of water that a soil can hold by capillary action before the water is drawn away by gravity, and was calculated according to:

$$FC = 0.3486 - 0.018 \times Sc(\%) + 0.0039 \times Cc(\%) + 0.0228 \times OM - 0.0738 \times BD \tag{8}$$

where the percentages of sand (Sc) (47%) and clay (Cc) (19%) were obtained from Brennan *et al.* (2012), OM is the organic matter (%) (equal to SOC × 1.724) (Labite and Cummins 2012), and BD is the bulk density (g/cm^3).

The soil air content was expressed as the difference in soil field capacity and porosity (P) (P – FC), and the latter was calculated according to:

$$P = 1 - BD \times 1000 / PD \tag{9}$$

where PD is the particle density (kg/m^3). A default value of 2650 kg/m^3 for particle density after Posudin (2014) was used, and is consistent with what is found in Irish soils (Labite and Cummins 2012).

Henry's constant (K_H) was retrieved from peer reviewed literature and was contaminant-specific (Table 3). Where there was a lack of dimensionless data, Eq. (10) was used to convert atm mol^{-1} to a dimensionless form. The dimensionless air partition coefficient K_H is given as:

$$K_H\left(atm\ m^3 mol^{-1}\right) = K_H' / RT \tag{10}$$

where R is the universal gas constant 0.08206 (L atm mol^{-1} K^{-1}) and T is temperature 298.15 (K).

The attenuation factor is a means of evaluating the amount of chemical transport through the unsaturated zone to the water table, and is defined as the fraction of the contaminant applied at soil surface that is likely to leach with values ranging from 0 to 1 (Labite and Cummins 2012). The attenuation factor was calculated according to Trevisan *et al.* (2009):

$$AF = \exp - TR \times \ln 2 / DT_{50} \tag{11}$$

where AF is the attenuation factor (-), DT_{50} is the half-life in soil (d) and is contaminant-specific and based on available data (Table 3). A uniform distribution was used to model uncertainty in soil half-life. Finally, TR is the average residence time (L/Yr) according to Trevisan *et al.* (2009) and is calculated by:

$$TR = GL \times RF \times FC / Q \tag{12}$$

where GL is the depth (groundwater level) of the water table layer (m), Q is the net recharge of the water table layer (m/yr) and can be estimated by rainfall and evaporation of the study area. The rainfall in Ireland is variable. Therefore, to capture the uncertainty a uniform distribution was assigned (minimum of 750, maximum of 1500 mm), while the evaporation rate was estimated to be 500 mm/yr (Met Eireann 2014).

The leached quantity was calculated after Trevisan *et al.* (2009):

$$LQ = 2.739 \times AF \times APPL \times (1 - fint) / P \times H \tag{13}$$

where LQ is the leached quantity (μg/l), AF is the attenuation factor (-) [from Eq. (11)], APPL is the application rate (g/cm^2), *fint* is the fraction intercepted from crop (%), and H is the thickness of the water table (m)

Human and environmental risk assessment

Human exposure. As a "worst case scenario" it was assumed that the water was abstracted for drinking pre-drinking water treatment. The amount of contaminant that may be ingested by humans through drinking water each day was estimated by:

$$HE = Wc \times PEC_{runoff} /_{groundwater} / BW \tag{14}$$

where HE is human exposure through chemical intake (μg/l), Wc is water consumption (L/day), and BW is body weight of an adult (kg). The amount of water consumed was based on two separate surveys conducted by The Irish Universities Nutrition Alliance (IUNA). The National Adult Nutrition Survey (2011) assessed the consumption and body weights of 1500 Irish consumers (IUNA 2011) and The National Children's Food Survey (2003–2004) assessed the consumption and body weights of 594 children (IUNA 2005). A log normal distribution was used to model the uncertainty regarding the intake of water and includes a mean consumption of 0.564 ± 0.617 L day^{-1} for adults and a mean consumption of 0.238 ± 0.208 L day^{-1} for children (IUNA 2005, 2011, respectively). A similar approach was taken by Cummins *et al.* (2010). Using the same surveys from IUNA, the average body weights of men and women and children were obtained. A normal distribution with a mean value of 78 ± 16.5 kg was used to model the variation in body weight for adults and a normal distribution with a mean value of 33 ± 11.3 kg was used to model variation in body weight for children (IUNA 2005, 2011, respectively). A summary of all model inputs and calculations is provided in Table 2.

Toxicity. Toxicity was calculated by using the lethal concentration (LC_{50}). There is a lack of mammalian toxicity data, particularly for emerging contaminates, hence the decision to use fish and fish embryos toxicity data. Fish have been used as sentinels for the quality of waters that serve as sources for human drinking water (Lammer *et al.* 2009). Using peer reviewed literature, the LC_{50} (fish and fish embryos) of each contaminant was obtained. The toxicity of organic contaminants on fish (acute toxicity) and fish embryos has been widely reported (Teneyck and Markee 2007; Ji *et al.* 2008; Kim *et al.* 2009: Ye *et al.* 2009), therefore, a considerable amount of experimental data could be compiled and reviewed. The application of the LC_{50} has gained acceptance among toxicologist and is generally the most highly rated test for assessing potential adverse effects of chemicals (Johnson and Finley 1980). Guilhermino *et al.* (2000) compared acute toxicity of 54 chemicals to *Daphnia magna*, expressed as 24- and 48-h LC_{50} values and the corresponding oral LD_{50} value for the rat. Results show a high correlation between acute toxicity of some chemicals to *D. magna* and the corresponding LD_{50} values for the rat. The study concluded that the *D. Magna* test was more specific than sensitive for an indication of toxicity to the rat and the use of invertebrates should be considered as a pre-screening method for assessment of the toxicity of new chemicals for classification and labeling purposes. Within the EU there are extensive regulatory requirements for fish acute toxicity data on individual chemicals both for environmental risk assessment and hazard classification (Braunbeck *et al.* 2005). A combination of uniform and triangular distributions was incorporated to capture variability. Full details of model data are available in Table 3. The risk ratio is the estimated and measured exposure to the toxicity. When the risk ratio is < 1, this would suggest that the level of risk is low. When the risk ratio is >1, the risk is high (Labite and Cummins 2012). To calculate the risk ratio (LC_{50}), the PEC_{runoff} and $PEC_{groundwater}$ was incorporated in:

$$RR(LC_{50}) = PEC_{runoff,\ groundwater} \,/\, LC_{50} \tag{15}$$

The entire model was constructed in Microsoft Excel 2010 (with the @Risk 6.0 add-on) (V4.5, Palisade Corporation, Newfield, NY) using Monte Carlo simulation techniques and run for 10,000 iterations.

Results and discussion

The environmental fate of selected organic contaminants was modeled from biosolid application to drinking water consumption. The risk ranking model resulted in several output distributions that can be used to compare the selected organic contaminants that are detected in biosolids and subsequently applied to agricultural land and their potential risk to human health. Outputs from the model include the PEC_{soil} of each contaminant, subsequent PEC_{runoff} to surface water and $PEC_{groundwater}$, the chemical intake risk ratio (RR) based on the LC_{50} combined with PEC_{runoff} and $PEC_{groundwater.}$ Table 4 shows the mean simulated ranking results according to PEC_{soil}, PEC_{runoff} and $PEC_{groundwater}$.

PEC_soil

The results of the PEC_{soil} indicate that from the contaminants analyzed, the contaminants NP and NP1EO and NP2EO ranked the highest, with mean PEC_{soil} values of 5.69 mg/kg, 1.72 mg/kg, and 1.44 mg/kg (95th percentiles 13.69 mg/kg, 5.14 mg/kg and 2.88 mg/kg, respectively) as shown in Table 4. This was attributed to the initial high concentrations of

Table 4. Risk ranking of $PEC_{soil,\ runoff,\ groundwater}$.

	Ranking according to $PEC_{soil,\ runoff,}$ and $_{groundwater}$ mean (5th, 95th percentiles)					
Contaminant	PEC_{soil} (mg/kg)	Rank	PEC_{runoff} (μg/l)	Rank	$PEC_{groundwater}$ (μg/l)	Rank
NP	5.69 (4.4e-01, 13.6)	1	1.1e-02 (1.8e-04, 4.8e-02)	1	2.22e-01 (0, 1.27)	1
NP1EO	1.72 (8.6e-02, 5.14)	2	4.1e-03 (1.7e-04, 1.3e-02)	2	1.84e-01 (9.1e-03, 5.5e-01)	2
NP2EO	1.44 (1.3e-01, 2.88)	3	3.3e-03 (1.3e-05, 9.9e-03)	3	9.11e-02 (0, 3.8e-01)	3
Triclocarban	2.7e-01 (4.3e-02, 6.1e-01)	4	8e-04 (6.2e-05, 2.1e-03)	4	1.4e-02 (1.9e-68, 6.4e-02)	4
Triclosan	3.1e-02 (2.3e-03,7.3e-02)	5	1.8e-04 (4.9e-06, 6.2e-04)	5	3.5e-04 (0, 1.4e-03)	6
BPA	1.7e-02 (3.1e-03, .3.4e 02)	6	6.6e-05 (1.0e-05,1.3e-04)	6	2.4e-0 (9.5e-77, 1.5e-03)	7
Carbamazepine	1.2e-02 (1.2e-03,2.5e-02)	7	4.7e-05 (4.2e-06, 1e-04)	7	1.1e-03 (3e-10,3.3e-03)	5
PBDEs	9e-03 (1.6e-03,2e-02)	8	6.2e-06 (1.9e-08, 2.9e-05)	8	2.1e-05 (0, .1.1e-04)	10
PCBs	1.7e-03 (1.3e-04, 4.2e-03)	9	3.4e-06 (2.3e-08, 1.3e-05)	10	9.5e-05 (1.5e-171, 4.6e-04)	9
PFOA	1.4e-03 (1.2e-04, 3.4e-03)	10	5.2e-06 (3.4e-07, 1.3e-05)	9	1e-04 (1.9e-40, 3.9e-04)	8
PFOS	5.6e-04 (2.1e-04,1.1e-03)	11	1.4e-06 (1.1e-07, 3.6e-06)	11	1.2e-05 (2.1e-224, 7e-05)	11
17βEstradiol	4.1e-04 (1.4e-04, 8e-03)	12	5.6e-07 (3.3e-08, 1.4e-06)	12	2.2e-07 (0, 2.4e-07)	14
Estrone	2.3e-04 (1.9e-05, 5.5e-04)	13	2.4e-07 (3.4e-09, 8.6e-06)	13	2.5e-06 (0, 1.6e-05)	12
PCDD/Fs	1.5e-05 (1.2e-06, 3.6e-05)	14	2.3e-10 (7.2e-13, 9.1e-10)	16	7.6e-16 (0, 3.5e-100)	16
Propranolol	6.2e-06 (4.7e-07,1.4e-05)	15	1.8e-08 (1.2e-09, 4.8e-08)	15	7.1e-08 (8.8e-84, 4.7e-07)	15
Metoprolol	5.8e-06 (1.0e-06,1.3e-05)	16	2.4e-08 (4.3e-09,5.2e-08)	14	6.6e-07 (4.8e-08, 1.7e-06)	13

NP, NP1EO, and NP2EO in the biosolids (mean values 103.8, 83.3, and 25.7 mg/kg, respectively), with NP and NP1EO exceeding the critical levels of 50 mg/kg dry mass (dm), as suggested by the European Union Working Document on Sludge 3rd draft (Ömeroğlu *et al.* 2015). Recently, Mao *et al.* (2012) reported that NP was the most abundant compound in raw wastewater as well as effluents from all stages of wastewater treatment. Furthermore, during wastewater treatment NP accumulated in sewage sludge at concentrations of several hundred mg/kg. This highlights the significance of effective wastewater treatment.

PEC_{runoff} and $PEC_{groundwater}$

The results for PEC_{runoff} revealed that the highest values obtained were from NP, NP1EO and NP2EO, with mean values of 1.17×10^{-2} μg/l, 4.13×10^{-3} μg/l and 3.36×10^{-3} μg/l (95th percentiles 4.08×10^{-2} μg/l, 1.36×10^{-2} μg/l and 9.99×10^{-3} μg/l, respectively) (Table 4). Petrovic *et al.* (2002) reported surface water body concentrations of nonylphenols between 0.5 μg/l to 15 μg/l. This was attributed to the proximity of the wastewater treatment plant to the receiving waters and the initial concentrations.in the sludge (85 and 172 mg/kg). The fate of NP, NP1EO, and NP2EO in each environmental compartment (soil, surface runoff, and groundwater) is controlled primarily by its physical chemistry properties, which in turn influences its degradation. These compounds will partition favourably to organic matter due to being highly hydrophobic (log Koc $\approx$ 4.6) with low mobility and low water solubility (Soares *et al.* 2008). Therefore, comparisons of nonylphenol concentrations between soils and surface water will show higher concentrations in soils. NP, NP1EO, and NP2EO ranked highest for $PEC_{groundwater}$ with mean values of 2.2×10^{-1} μg/l, 1.8×10^{-2} μg/l and 9.1×10^{-2} μg/l (95th percentiles 1.2 μg/l, 5.5×10^{-1} μg/l and 3.8×10^{-1} μg/l, respectively) (Table 4). Following agricultural practices, Careghini *et al.* (2014) reported concentrations of nonylphenols in groundwater with values between below 0.01 (detection limit) and 0.90 μg/l. Engstrom *et al.* (2006) measured NP concentrations in biosolid amended soils over a 4- to 12-month period. Results show that leachate NP concentrations were never higher than 7.5 μg/L and decreased to 1.5 μg/L after a four month period. Nonylphenols were degraded in agricultural soil with a half-life in the order of months. The top ranking contaminants for PEC_{runoff} (NP, NP1EO, and NP2EO) are below the Water Framework Directive threshold value (0.3 μg/l) for annual average. Although the predicted concentrations in runoff and groundwater are below the threshold value, it is imperative to note that these contaminants are continuously released into the environment.

Human exposure/toxicity

Drinking water consumption was combined with body weight and PEC_{runoff} to yield a chemical exposure estimate. Results showed that the contaminants that ranked the highest for adult and child consumption were NP, NP1EO and NP2EO, with mean values of 8.6×10^{-5}, 3.1×10^{-5} and 2.5×10^{-5} μg/kg bw/d, respectively, for adult consumption and 9.27×10^{-5}, 3.4×10^{-5}, and 2.2×10^{-5} μg/kg bw/d, respectively, for child consumption (Table 5, Figure 2). HE and $PEC_{groundwater}$ showed that N,P NP1EO, and NP2EO ranked the highest for adult and child consumption, with mean values 1.6×10^{-3}, 1.4×10^{-3} and 7.0×10^{-4} μg/kg bw/d, respectively, for adult consumption and 2.0×10^{-3}, 1.6×10^{-3}, and 5.1×10^{-4} μg/kg bw/d, respectively, for child consumption (Table 5, Figure 3). There was a significant difference in human exposure between adult and child. Child consumption concentrations were higher in PEC_{runoff} and $PEC_{groundwater}$ for NP, NP1EO, and NP2EO (see below). Although all concentrations consumed are deemed low, children have different exposures to environmental contaminants from those of adults. Their physiological developments are often subjected to higher exposure of contaminants found in food, water and air. These exposures may be handled quite differently by an immature set of physiological systems to the way they are dealt with by adults. The WHO states that "the developmental component of a child's physiology is changing: maturing, differentiating and growing in phases known as 'developmental windows'. These 'critical windows of vulnerability' have no parallel in adult physiology and create unique risks for children exposed to contaminants that can

Table 5. Comparison of human exposure (consumption) between PEC_{runoff} and $PEC_{groundwater}$.

	Human exposure combined with PEC_{runoff} (μg/kg bw/d)			Human exposure combined with $PEC_{groundwater}$ (μg/kg bw/d)		
Contaminant	Adult	Child	Rank	Adult	Child	Rank
NP	8.5e-05	9.7e-05	1	1.5e-03	2.2e-03	1
NP1EO	3.0e-05	3.4e-05	2	1.4e-03	1.6e-03	2
NP2EO	2.8e-05	2.2e-05	3	7.2e-04	5.1e-04	3
Triclocarban	5.9e-06	6.1e-06	4	8.2e-05	9.0e-05	4
Triclosan	6e-06	1.3e-06	5	2.2e-06	2.6e-05	6
Carbamazepine	3.7e-07	4.0e-07	6	1.9e-06	2.5e-06	5
BPA	4.9e-07	1.3e-07	7	9e-06	4.5e-06	7
PBDES	4.5e-08	1.3e-08	8	1.6e-07	8.0e-07	8
PFOA	3.9e-08	1.2e-08	9	7.7e-07	3.2e-10	14
PCB	2.7e-08	5.9e-08	10	7.7e-07	2.4e-07	9
PFOS	1e-08	3.3e-08	11	8.9e-08	1.6e-07	10
17β Estradiol	4.2e-09	1.1e-09	12	1.5e-09	3.6e-12	15
Estrone	1.8e-09	4.8e-09	13	1.8e-08	2.2e-09	12
Metoprolol	1.8e-10	6.1e-10	14	5e-09	3.5e-09	11
Propranolol	1.3e-10	3.5e-10	15	5.4e-10	7.3e-10	13
PCDD/Fs	1.5e-12	7e-12	16	7.8e-18	1.3e-17	16

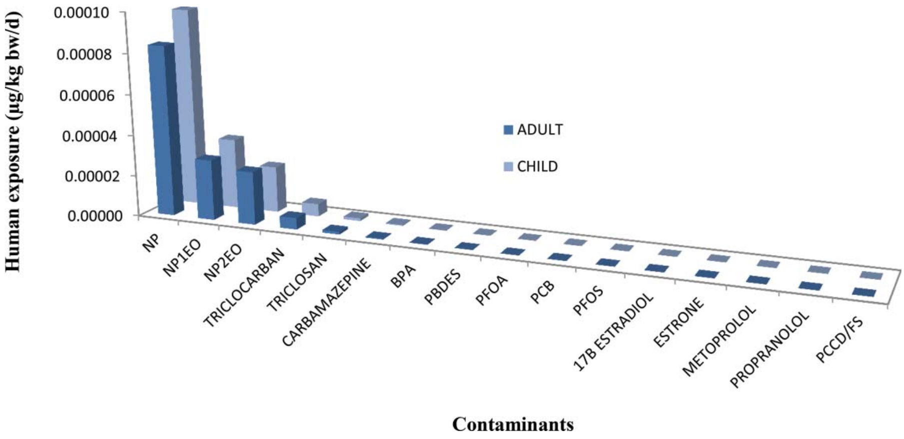

Figure 2. Human exposure based on PEC_{runoff} (adult and child).

alter normal function and structure" (WHO 2008, p 5). The daily intake values estimated in the present study for NP, NP1EO, and NP2EO are much lower than the 5 μg/kg bw/d tolerable daily intake (TDI) of NP proposed by the Danish Environmental Protection Agency (Ademollo *et al.* 2008; Li *et al.* 2010). Diet is the main route of exposure of nonylphenols in humans. Maggioni *et al.* (2013) evaluated concentrations of NP from 35 public drinking fountains and bottled mineral waters in Italian cities and found that NP was detected in ranges from 7.7 μg/l to 84 μg/l. Based on bottled water consumption, it was estimated that the average daily intake varies between 0.36 and 0.60 μg/d for adults (Careghini *et al.* 2014).

According to the LC_{50} risk ranking results for human exposure (LC_{50} RR PEC_{runoff}), the top ranked contaminants considered to be a risk to human health include NP, NP1EO and NP2EO with mean LC_{50} RR PEC_{runoff} values of 1.10×10^{-4}, 3.94×10^{-5}, and 1.35×10^{-5}, respectively (Table 6). Similarly, the results of the LC_{50} RR $PEC_{groundwater}$ also included NP, NP1EO, and NP2EO (Table 6). Mean values for LC_{50} RR $PEC_{groundwater}$ were 2.40×10^{-3}, 1.64×10^{-3}, and 3.66×10^{-4} for NP1EO, NP, and triclocarban, respectively. The

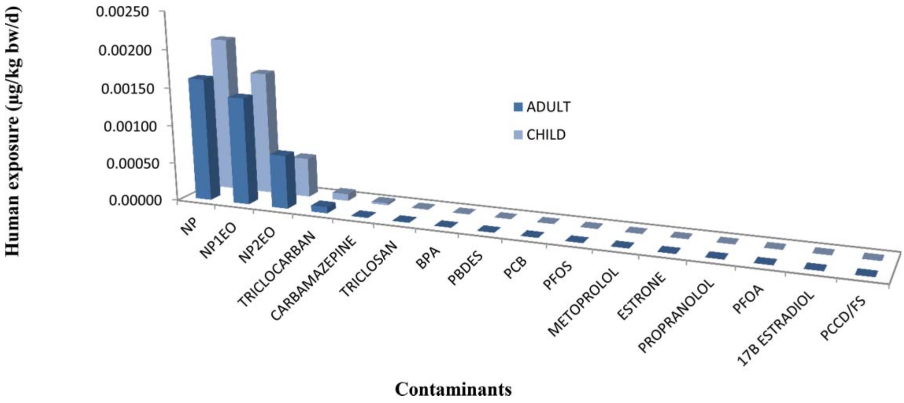

Figure 3. Human exposure based on $PEC_{groundwater}$ (adult and child).

Table 6. Comparison of human exposure (PEC_{runoff} and $PEC_{groundwater}$) combined with LC_{50}.

Ranking according to human health based risk Results based on mean PEC_{runoff} and $PEC_{groundwater}$ combined with LC_{50} (RR)				
Contaminant	(RR) PEC_{runoff}	Rank	(RR) $PEC_{groundwater}$	Rank
NP	1.10e-04	1	2.40e-03	1
NP1EO	3.94e-05	2	1.64e-03	2
NP2EO	1.35e-05	3	3.66e-04	3
Triclocarban	1.20e-05	4	1.80e-04	4
Triclosan	4.45e-06	5	7.40e-06	5
PCBs	4.45e-06	6	1.26e-06	6
BPA	2.92e-08	7	1.77e-07	7
PBDE	7.42e-09	8	9.68e-08	8
Carbamazepine	1.48e-09	9	3.79e-08	9
17β Estradiol	1.29E-09	10	2.40E-14	15
Estrone	1.61E-10	11	1.21E-10	11
PFOS	1.36E-10	12	1.76e-09	10
PFOA	1.23E-11	13	5.65E-14	14
PCDD/Fs	6.10E-12	14	1.56E-16	16
Propranolol	3.19E-12	15	6.95E-12	12
Metoprolol	2.11E-13	16	4.44E-12	13

contaminants that follow NP, NP1EO, and NP2EO in the rankings are the personal care products (PCPs), triclocarban, and triclosan. These PCPs are classed as fungicides and bacteriostats; therefore they have numerous applications in consumer markets. TCS and TCC can be found in soaps, detergents, and even in pacifiers (Halden 2014). On December 16, 2013, the US Food and Drug Administration proposed a rule that would require manufacturers to demonstrate the safety and effectiveness of antimicrobials (USFDA 2015). This proposal came 40 years after the first attempt to regulate TCC and TCS despite toxicological reports regarding its endocrine disrupting properties and protagonists of antibiotic drug resistance (Halden 2014). Recently, the European Chemicals Agency (ECHA) announced that TCS will be phased out for hygienic uses and replaced by more suitable alternative due to its toxic and bioaccumulative properties (ECHA 2015). As the nonylphenols are restricted in use in the EU under the 2003/53/EC Directive (EC 2003), which restricts the use and marketing of in Europe of products and product formulations that contain more than 0.1% NP or NPE (TFL 2007), it should see the decline of these products and its detection in the environment. However, detection of ECs such as triclocarban and triclosan are increasing in biosolids and surface waters. These chemicals are beginning to be classified as priority substances and substituted; however, their bioaccumulative properties may still persist in soil and water bodies for decades to come. With the anticipation of greener pharmaceuticals being developed, antimicrobials may be created that will be of low toxicity to terrestrial and aquatic organisms and rapidly degrade in the environment.

The risk ranking conducted in the present study can be used to highlight contaminants that may pose a risk to human health from potential transportation of contaminants in biosolids applied to agricultural land through to drinking water. A similar study has been conducted by Cooper *et al.* (2008) that ranked numerous pharmaceuticals that pose the greatest risk to the environment. Ranking was based on consumption rates, surface water concentrations, effluent concentrations, environmental and biological half-lives, mammal, fish and crustacean toxicity octanol-water partition coefficients (Kow) and solubility. The results show that the pharmaceuticals most commonly ranked were those that were classed as central nervous system drugs (*e.g.*, anti-inflammatory drugs and psychotherapeutic drugs) followed by cardiovascular drugs and anti-infective drugs. It was found that prescription usage greatly influenced what compounds ranked the highest. The study also highlighted the lack or abundance of data for some of the pharmaceuticals as some of the pharmaceuticals that ranked highest were driven by known toxicity. Coutu *et al.* (2012) also conducted a risk ranking of several pharmaceuticals in the environment. The study enables ranking of the hazard, not risk, to aquatic species and human health. Hazard assessment was based on the physio-chemical properties of each pharmaceutical. Results show that the hormones ethinylestradiol and testosterone ranked the highest along with the antibiotic erythromycin A. There are concerns that risk assessments focus on the contaminants that are commonly consumed, or on conventional contaminants such as legacy pollutants. Daughton (2014) describes a psychosocial phenomenon known as the "Matthew effect," omore commonly known as "the bandwagon effect." The authors describe the phenomenon as "the prominence of a few contaminants targeted for investigation is dictated largely by the attention devoted to them in the past." This can be seen by the repetitiveness of contaminants studies in peer-reviewed literature. The concern lies in the contaminants that escape scrutiny due to the lack of monitoring, available data, and regulation. This study highlights the need for more wide-ranging contaminant monitoring as many contaminants such as high production volume pharmaceuticals have not been detected in the environment but are estimated to be persistent and/or bioaccumulative.

Sensitivity analysis

A sensitivity analysis based on the rank order correlation coefficient was conducted for NP as this contaminant ranked the highest across all of the environmental compartments. Sensitivity analysis assesses how the model predictions are dependent on variability and uncertainty in the model's inputs. Results revealed that soil Koc and SOC were the most

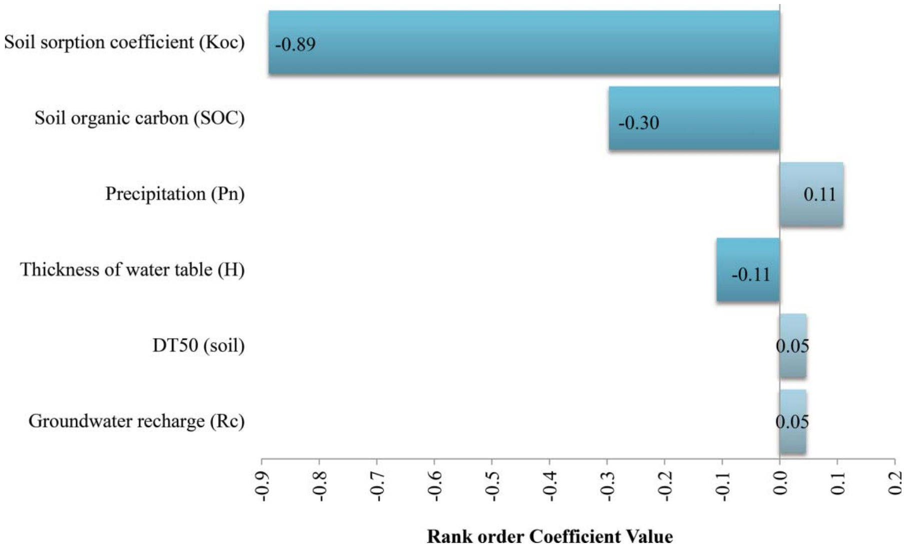

Figure 4. Sensitivity analysis of correlation coefficient (Spearman Rank) model input parameters for NP.

important parameters (correlation coefficient values –0.89 and –0.30, respectively) that affected the variance in model predictions (Figure 4). This highlights the importance of soil and site conditions (SOC, DT50-soil,) influencing the runoff and leaching of the contaminants to groundwater as well as contaminant properties (Koc) in influencing risk estimates. This directs future research in highlighting the need to reduce these uncertainty inputs to improve model estimates. Furthermore, the sensitivity analysis results also show water table depth, groundwater recharge and precipitation as other parameters of importance in the human health risk model.

Conclusions

A probalistic model was developed to rank "legacy" and EC according to PEC_{soil}, PEC_{runoff}, and $PEC_{groundwater}$ and resulting human health risk (RR). The highest rank obtained for PEC_{soil}, PEC_{runoff} and $PEC_{groundwater}$ was the surfactant NP and its ethoxylates NP1EO and NP2EO. The PEC_{runoff} and $PEC_{groundwater}$ were combined with drinking water consumption and body weights of adults and children to give the likely human exposure. The toxicity endpoint LC_{50} was combined with $PEC_{runoff\ and}$ $PEC_{groundwater}$ to give the chemical intake toxicity ratio (RR). The LC_{50} combined with PEC_{runoff} and $PEC_{groundwater}$ revealed that NP, NP1EO, and NP2EO ranked the highest. A sensitivity analysis revealed that Koc and SOC were the most important parameters that affected model variance. This indicates that the consistency of the unsaturated zone (soil/subsoil/bedrock) that biosolids are spread on and chemical properties are critical in controlling human health risk. Although the nonylphenols ranked highest in this study, it is important to note that these contaminants are either restricted or banned in Europe since 2005; therefore, these contaminants may be considered low risk. However, they still persist in the environment. The contaminants that ranked just below the nonylphenols, such as triclocarban and triclosan, can be considered more of an evolving risk as these contaminants are emerging and have only recently been restricted within the European Union. The model developed in this study is of importance for risk managers in providing a ranking of potential chemical hazards resulting from the spreading of biosolids on agricultural land, while highlighting some emerging contaminants requiring vigilance in the future.

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