The aims of this study were to: (1) undertake a thorough literature review of the spreading of treated sewage sludge (biosolids) on land to include analysis of potential impacts on environmental and human health; (2) examine, under controlled conditions in the laboratory and field, the impact of the landspreading of biosolids (on grassland) on surface runoff/subsurface drainage/shallow groundwater of nutrients, solids, metals, pathogens and some specified emerging contaminants identified in the literature review, when spread based on N and P application rates; and (3) to model and conduct a risk assessment of potential hazards of human health concern.

Identifying Risks

Implementation of European Union Directives in recent decades concerning the collection, treatment and discharge of wastewater, as well as technological advances in the upgrading and development of wastewater treatment plants, has resulted in an increase in the number of households connected to sewers and an increase in the production of sewage sludge (the by-product of wastewater treatment plants). Recycling to land is currently considered the most economical and beneficial way for municipal sewage sludge management. However, despite the many potential benefits of recycling municipal sewage sludge to land, there are many risks, which include the presence of emerging contaminants in the sewage sludge that may enter the food chain, and the potential for surface runoff of contaminants into receiving waters. This project found that although the application of biosolids poses no greater threat to surface water quality than the land application of dairy cattle slurry, there is a possibility that many non-priority elements and emerging contaminants, for which no legislation currently exists, may be applied to land without regulation, and may accumulate in the soils and enter the food chain.

Informing Policy

Current legislation governing the land application of municipal sewage sludge to land considers certain priority pollutants and bio-essential elements. However, other emerging contaminants may be inadvertently applied to land. Regulations should be extended to cover non-priority elements, pharmaceuticals and personal care products (PPCPs). Non-priority elements are relatively inexpensive to measure, but PPCPs are prohibitively expensive as well as being continuously evolving. Wastewater treatment plants may be upgraded to include treatment of emerging contaminants, but the potential presence of known, as well as currently unknown parameters, raises concerns over the continued application of biosolids to land in Ireland.
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- Office of Communications and Corporate Services
The EPA is assisted by an Advisory Committee of twelve members who meet regularly to discuss issues of concern and provide advice to the Board.
Health and Water Quality Impacts Arising from Land Spreading of Biosolids

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EPA Research Report

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The EPA Research Programme addresses the need for research in Ireland to inform policymakers and other stakeholders on a range of questions in relation to environmental protection. These reports are intended as contributions to the necessary debate on the protection of the environment.
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Executive Summary

Treated sewage sludge, commonly referred to as "biosolids", is the organic by-product of urban wastewater treatment. If appropriate treatment is applied, it may be reused as an agricultural fertiliser. Despite this benefit, several issues are associated with the reuse of municipal sewage sludge in agriculture. Although many of these are issues of perception, there is considerable concern over the presence of metals, nutrients, pathogens, pharmaceutical and personal care products (PPCPs), and other endocrine-disrupting and synthetic compounds in biosolids, which may cause environmental and human health problems.

The main aims of this research were to (1) quantify the range of concentrations of metals and two of the most abundant PPCPs in the world, the antimicrobials triclosan (TCS) and triclocarban (TCC), in biosolids from a range of wastewater treatment plants (WWTPs) in Ireland; (2) undertake a field-scale experiment to assess losses of nutrients (nitrogen and phosphorus), metals, TCS, TCC and microbial matter (total and faecal coliforms) after successive rainfall events on grassland onto which biosolids had been applied, and to compare the results with another commonly spread organic fertiliser, dairy cattle slurry (DCS); (3) measure the uptake of metals by ryegrass for a period of time after the application of biosolids; and (4) conduct a risk assessment of potential hazards to human health based on the experimental data.

Three types of biosolids commonly used in Ireland were examined as part of this study: anaerobically digested (AD), lime stabilised (LS) and thermally dried (TD) biosolids. Biosolids and DCS were surface applied in accordance with the legislation in Ireland. A rainfall simulator was used to generate surface runoff over three successive events (24 hours, 48 hours and 360 hours) after a single application.

The metals in the biosolids from the WWTPs examined were below the maximum allowable concentrations for use in agriculture in the European Union (EU). Some priority metals, such as antimony and tin, which are potentially harmful to human health, were identified in some of the samples analysed. As these parameters are not currently regulated, this means that a number of toxic metals, which are at concentrations of up to 40-times higher than their baseline concentrations in soils, are being applied to land without regulation. In the WWTPs examined, the concentrations of TCS and TCC were 0.61 and 0.08 µg g⁻¹, which are below the concentrations for these parameters measured in other countries. Similar to the findings for metals, the possibility exists that these potentially harmful, unregulated contaminants, for which no international standards currently exist for recycling in agriculture, may accumulate in the soil upon repeated application.

When losses of nutrients, metals and indicator microorganisms arising from biosolid-amended plots were compared with slurry treatments, biosolids did not pose a greater risk in terms of losses along the surface runoff pathway. The concentrations of TCS and TCC in surface runoff were also mainly below the limits of detection (90 ng L⁻¹ for TCS, 6 ng L⁻¹ for TCC). Furthermore, there was no significant difference in metal bioaccumulation for the ryegrass between plots that received biosolids and those that did not over the course of the study.

A literature review identified contaminants of concern based on relevant risk factors, persistence, bioaccumulation and toxicity (PBT). The contaminants identified were persistent organic pollutants (POPs), pharmaceuticals and PPCPs. A suite of 16 contaminants identified in the literature was further analysed in a risk-ranking model to include health-based risk endpoints. A probabilistic model was constructed in Excel 2010 (incorporating @Risk 6.0) to estimate human exposure to the organic contaminants that are contained within biosolids destined for land application. Nonylphenols ranked highest across all environmental compartments. The use of these contaminants is heavily restricted in the EU; however, because of their persistence, the bioaccumulation and toxicity of these compounds in the environment remains a concern. TCC and TCS also ranked highly, and may be considered potentially greater risks, as their use is not restricted and they are known to cause adverse health effects.

An exposure assessment model was further developed both for metals and for Escherichia coli (E. coli). The model considered exposure to metals and E. coli through surface water abstracted for drinking, taking account of surface runoff, dilution and water treatment.
effects. The likelihood of illness arising from exposure, and the severity of the resulting illness, were evaluated. Different dose–response relationships were characterised for the different pollutants with reference to the lifetime average daily dose (LADD) and hazard quotient (HQ) for metals, while a worst-case negative exponential dose–response model was used for *E. coli*. Of the three biosolids treatment scenarios considered, and with regard to LADD, the results showed that mean copper exposure concentrations for children were highest in all three rainfall events corresponding to the LS treatment (mean values of $2.07 \times 10^{-2}$, $2.07 \times 10^{-2}$ and $1.18 \times 10^{-2}$ µg kg$^{-1}$ bw d$^{-1}$). This was followed by adult copper exposure concentrations (mean values of $1.80 \times 10^{-2}$, $1.31 \times 10^{-3}$ and $9.21 \times 10^{-3}$ µg kg$^{-1}$ bw d$^{-1}$, for all three rainfall events). The results for the HQ showed that, of all the scenarios considered, the metal, copper, and biosolid treatment, LS, had the highest HQ for children for all three rainfall events, with mean child HQ values of $5.59 \times 10^{-3}$, $4.09 \times 10^{-3}$ and $3.18 \times 10^{-3}$ µg kg$^{-1}$ bw d$^{-1}$, respectively, followed by mean adult HQ values of $4.87 \times 10^{-3}$, $3.54 \times 10^{-3}$ and $2.49 \times 10^{-3}$ µg kg$^{-1}$ bw d$^{-1}$, respectively. However, these were still below the threshold value of risk (HQ < 0.01, no existing risk).

The results for viable *E. coli* consumed show that one of the sludges examined [an anaerobically digested sludge originating from a WWTP in the UK (ADUK)] was highest for the first and second rainfall events, with mean exposure values of $5.20 \times 10^{-1}$ MPN/100mL and $2.34 \times 10^{-1}$ MPN/100mL, respectively. The results for the probability of illness for healthy and immunocompromised populations showed that among immunocompromised populations the biosolids treatment ADUK (first and second rainfall events) had the greatest probability of illness/day, with mean probability values of $3.68 \times 10^{-3}$ and $2.1 \times 10^{-3}$ illnesses/day, respectively. The results indicate that the risk of illness was negligible for healthy individuals; however, care is required with immunocompromised individuals if the annual risk is greater than the threshold risk of illness ($10^{-4}$), as set by the US Environmental Protection Agency.

The overall conclusion from this study is that, although, in general, land applied biosolids pose no greater threat to water quality than DCS, cattle exclusion times from biosolid-amended fields may be overly strict (within the context of current exclusion criteria). A matter of concern is that unlegislated metals and PPCPs, which were found to be present in biosolids originating from a selection of the WWTPs examined in this study, may be inadvertently applied to land. With multiple applications over several years, these may build up in the soil and may enter the food chain; this gives rise to concerns over the continued application of biosolids to land in Ireland.
Figure ES1. Graphical abstract of project. ADIRE, anaerobically digested biosolids sourced in Ireland; ADVK, anaerobically digested biosolids sourced in the United Kingdom; Cu, copper; LS, lime stabilisation; Ni, nickel; TD, thermally dried; Zn, zinc.
1 Introduction

1.1 Overview

In the European Union (EU), implementation of directives and other legislative measures in recent decades concerning the collection, treatment and discharge of wastewater, as well as technological advances in the upgrading and development of wastewater treatment plants (WWTPs) (Robinson et al., 2012), has resulted in a rise in the number of households connected to sewers, which has increased the loadings on WWTPs. Production of untreated sewage sludge (the by-product of wastewater treatment plants) across the EU has increased from 5.5 million tonnes of dry matter (DM) in 1992 to an estimated 10 million tonnes in 2010 (Eurostat, 2014), with production expected to increase further to 13 million tonnes in all EU Member States by 2020 (EC, 2010).

Recycling to land is currently considered the most economical and beneficial method of municipal sewage sludge management (Haynes et al., 2009; Peters and Rowley, 2009; Healy et al., 2015). However, before this can occur, it must be treated to prevent harmful effects on soil, vegetation, animals and humans (EC, 2016). Chemical, thermal or biological treatments, which may include composting (USEPA, 2002), aerobic and anaerobic digestion (USEPA, 2006a), thermal drying (USEPA, 2006b), or lime stabilisation (USEPA, 2000), produce a stabilised organic material frequently referred to as “biosolids”.

Recycling biosolids provides many benefits to grassland: (1) their use completes the urban–rural cycle (Fehily Timoney and Company, 1999); (2) they may be used as a soil conditioner, improving its physical, chemical and biological properties, and reducing the possibility of soil erosion (Lucid et al., 2014); and, most importantly, (3) they are a cheap organic alternative to commercial fertiliser (Lu et al., 2012).

Many potential problems are associated with the land application of biosolids and these were reviewed by, among others, Lu et al. (2012) and Singh and Agrawa (2008). Among the main concerns are nutrient, metal, microbial and emerging contaminant losses in runoff (Dowdy et al., 1991; Eldridge et al., 2009; Wallace et al., 2014) and the accumulation of metals in both soil and crops after repeated applications of biosolids (McBride, 2003; Bai et al., 2010). For example, micro-plastics, which have been found in high concentrations in sewage sludge and have been detected on soils 15 years post-application (Zubris and Richards, 2005; Magnuson and Norén, 2014), may leach organic contaminants and/or endocrine disruptors and can be translocated to the human lymph and circulatory system upon ingestion (Hussain et al., 2001; Cole et al., 2011). Of particular concern is the content of the pharmaceutical and personal care products (PPCPs), in particular the antimicrobials triclosan (TCS) and triclocarban (TCC), in biosolids. Both compounds are commonly found to be the most abundant contaminants in biosolids (McClellan and Halden, 2010) and both are listed among the top contaminants of concern worldwide (Vericchi and Zambello, 2015). Even though their use has been restricted in the US (USFDA, 2015) and EU (EC, 2016), there may still be legacy issues in municipal sewage sludge and wastewater for some time to come.

Biosolids may provide an excellent opportunity to improve crop profit margins and reduce potential environmental impact by means of reducing the input costs of production, use and distribution of chemical fertilisers. However, there is a need for continued research into land spreading practices to ensure that environmental losses and associated concerns are minimised.

1.2 Objectives

The following were the specific objectives of this project:

- To characterise the metal, TCS and TCC content of biosolids originating from a number of WWTPs in Ireland.
- To quantify losses of nutrients [nitrogen (N), phosphorus (P)], metals [copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), cadmium (Cd), chromium (Cr)], TCS and TCC, and microbes [total coliforms (TC) and faecal coliforms (FC)], in surface runoff from experimental micro-plots at time intervals of 24, 48 and 360 hours, following application of three types of biosolids at the legal application rate based on current EU legislation. The losses arising from no application (the study control) and the application...
of the biosolids to grassland were compared to losses on similar micro-plots following application of another organic fertiliser that is commonly spread in Ireland, dairy cattle slurry (DCS).

- To measure the bioaccumulation of metals in rye-grass for a period of up to 130 days after the land application of biosolids.

- To develop a risk assessment model of human exposure to pathogens and emerging pollutants resulting from the application of urban wastewater biosolids to agricultural land.
2  Literature Review on Risks Arising from Application of Biosolids to Land

2.1  Overview

The aim of this chapter is to examine the recovery of nutrients and other compounds, such as volatile fatty acids, polymers and proteins, from municipal sewage sludge. Due to the increasing awareness regarding risks to the environment and human health, the application of sewage sludge to land, following treatment, as a fertiliser in agricultural systems has come under increased scrutiny. Therefore, any potential benefits accruing from the reuse of sewage sludge are considered after having been compared with possible adverse impacts associated with its use. This chapter is an abridged version of Healy et al. (2015).

2.2  Introduction

“Sludge” is residual-treated or untreated sludge from urban WWTPs (S.I. 254 of 2001). Due to the treatment processes employed in WWTPs, metals, trace organic compounds and pathogenic organisms are concentrated in sludge. However, it is also rich in nutrients and contains valuable organic matter (OM). European policy promotes a “circular economy” (EC, 2014) and emphasises a hierarchy of waste management, including prevention, preparing for reuse, recycling, other recovery and disposal (EU, 2008). This has prompted efforts within municipal sewage sludge management to utilise sewage sludge as a commodity. The terminology “biosolids” reflects the effort to consider these materials as potential resources (Isaac and Boothroyd, 1996).

The use of biosolids in agriculture provides the necessary nutrients and micronutrients necessary for plant and crop growth. They may be used as a soil conditioner, improving its physical and chemical properties (Hargreaves et al., 2008) and reducing the possibility of soil erosion (Lucid et al., 2014). Their use also addresses EU policy on sustainability and reuse of resources (EC, 2014). Numerous studies have documented their efficacy in increasing crop yields and their use in biofuel cropping systems, and in general, biosolids applications to land have been found to have a statistically significant impact on crop yields (e.g. Latare et al., 2014; Liu et al., 2015) and soil phosphorus (Shu et al., 2016), while having negligible adverse ecological impacts (Xue et al., 2015). A selection of recent studies that report impacts of biosolids application on crop growth, soil fertility and water holding capacity, is shown in Table 2.1.

Despite these benefits, however, there are several issues associated with the reuse of municipal sewage sludge in agriculture, which deserve particular attention. While many of these are issues of perception (Robinson et al., 2012), there is considerable concern about the presence of persistent and emerging contaminants in biosolids, the risk of contamination of soil and water (Fu et al., 2016), and the presence of toxic metals and pharmaceuticals in the sludge. The latter may build up in the soil and enter the food chain following continuous applications of biosolids to land (Latare et al., 2014).

2.3  Trends in Municipal Sewage Sludge Treatment

In Ireland, more than 53,000 tonnes of sewage sludge was produced in 2014, of which 79% was used on agricultural land as a fertiliser or soil enhancer (EPA, 2015). The amount of sewage sludge produced in Europe has generally increased, which is mainly attributable to the implementation of the Urban Waste Water Treatment Directive 91/271/EC (EU, 1991) and other legislative measures. The treatment and disposal of municipal sewage sludge presents a major challenge in wastewater management. As seen over the last decade, the upgrading and development of effective treatment plants has facilitated efforts to improve the quality of the effluent (i.e. removal of microorganisms, viruses, pollutants). Subsequently, legislation regarding municipal sewage sludge in the EU (Sewage Sludge Directive 86/278/EEC; EEC, 1986) and the USA (40 CFR Part 503; USEPA, 1995) has focused on effluent quality and potential contamination. Within the EU, treated municipal sewage sludge is defined as having undergone biological, chemical or heat treatment, long-term storage, or any other appropriate process so as to significantly reduce fermentability and any health hazards.
Land Spreading of Biosolids: Health and Water Quality Impacts

resulting from its use (EC, 2016). Physical-chemical treatment of wastewater has been widely practised, introducing biodegradation and chemical advanced oxidation for biological treatment (Mouri et al., 2013). In the treatment of wastewater, biological treatments, such as aerobic and anaerobic digestion, appear to be the more favoured option. Aerobic treatment has a high degree of treatment efficiency, while anaerobic biotechnology has significantly progressed, offering resource recovery and utilisation while still achieving the objective of waste control (Chan et al., 2009). With regard to sludge stabilisation, aerobic and anaerobic treatments are the most widely used methods of sewage sludge treatment. Within the EU, anaerobic and aerobic wastewater treatments appear to be the most common methods, with 24 out of 27 countries applying this method (Kelessidis and Stasinakis, 2012).

2.4 Legislation Covering Disposal of Sewage Sludge on Land


In the EU, the application of treated municipal sewage sludge to agricultural land is governed by EU Directive 86/278/EEC (EU, 1986), which requires that sewage sludge undergoes biological, chemical or heat treatment, long-term storage, or any other process to reduce the potential for health hazards associated with its use. In the EU, land application of treated municipal sewage sludge is typically based on its nutrient and metal content, although individual Member States often have more stringent limits than the directive (EC, 2010; Milieu Ltd et al., 2013a,b,c). Generally, when applying treated sewage sludge based on these guidelines and depending on the nutrient and metal content of the treated sewage sludge, P becomes the limiting factor for application.

In Ireland, the use of sewage sludge in agriculture is regulated by the Waste Management (Use of Sewage Sludge in Agriculture) Regulations (S.I. 148 of 1998 and S.I. 267 of 2001), which prescribe standards and limits on sludge used in agriculture subject to the carrying out of nutrient management plans. In addition, the “Code of Good Practice for the Use of Biosolids in Agriculture” sets out mandatory guidelines for producers, end-users and local authority regulators of sewage sludge used

<table>
<thead>
<tr>
<th>Country</th>
<th>Research focus</th>
<th>Application rate</th>
<th>% increase of parameter measured vs no biosolids application</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>USA</td>
<td>Switchgrass growth</td>
<td>0 kg N ha⁻¹</td>
<td>0</td>
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<td></td>
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</tr>
<tr>
<td>Canada</td>
<td>Soil test phosphorus</td>
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<td>Shu et al. (2016)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>28 Mg ha⁻¹</td>
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<tr>
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<td>0</td>
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Table 2.1. Impacts of biosolids application on soil fertility and plant productivity

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in agriculture. The use of sewage sludge in agriculture must also comply with the provisions of the EU (Good Agricultural Practice for Protection of Waters) Regulations, 2014, as amended (S.I. 31 of 2014), which specify the periods when land application of fertilisers is prohibited and limits application of fertilisers on the land.

2.5 Existing and Emerging Issues Concerning the Reuse of Sludge on Land

2.5.1 Nutrient and metal losses

Phosphorus and reactive N losses to a surface water body originate from either the soil (chronic) or in runoff where episodic rainfall events follow land application of fertiliser (incidental sources) (Brennan et al., 2012). Such losses to a surface water body occur via primary drainage systems, runoff and/or groundwater discharges. Application of treated sewage sludge to soils may also contribute to soil test P build-up in soils, thereby contributing to chronic losses of P in runoff (Gerba and Smith, 2005). Dissolved reactive P losses may also be leached from an agricultural system to shallow groundwater (Galbally et al., 2013) and, where a connectivity exists, may affect surface water quality for long periods of time (Schulte et al., 2010; Fenton et al., 2011).

The metal content of treated sludge, and of the soil onto which it can be spread, is also regulated by legislation in Europe (86/278/EEC; EU, 1986). However, guidelines governing the application of treated sewage sludge to land (e.g. Fehily Timoney and Company, 1999) mean that it is frequently the case that application rates are determined by the nutrient content of the sludge and not its metal content (Lucid et al., 2013). Regardless, concerns have been raised about the potential for transfer of metals into water bodies, soil structures and, consequently, the food chain (Navas et al., 1999).

2.5.2 Pathogens

During wastewater treatment, the sludge component of the waste becomes separated from the water component. As the survival of many microorganisms and viruses in wastewater is linked to the solid fraction of the waste, the numbers of pathogens present in sludge may be much higher than the water component (Straub et al., 1992). Although treatment of municipal sewage sludge using lime, anaerobic digestion or temperature may substantially reduce pathogens, complete sterilisation is difficult to achieve (Sidhu and Toze, 2009) and some pathogens, particularly enteric viruses, may persist. Persistence may be related to factors such as temperature, pH, water content (of treated sludge) and sunlight exposure (Sidhu and Toze, 2009). Also, there is often resurgence in pathogen numbers post-treatment, known as the “regrowth” phenomenon. This may be linked to contamination within the centrifuge, reactivation of viable, but non-culturale, organisms (Higgins et al., 2007), storage conditions post-centrifugation (Zaleski et al., 2005) and proliferation of a resistant sub-population due to newly available niche space associated with reduction in biomass and activity (McKinley and Vestal, 1985).

The risk associated with sludge-derived pathogens is largely determined by their ability to survive and maintain viability in the soil environment after landspreading. Survival is determined by both soil and sludge characteristics. The major physico-chemical factors that influence the survival of microorganisms in soil are currently considered to be soil texture and structure, pH, moisture levels, temperature, UV radiation, nutrient and oxygen availability, and land management regimes [reviewed in van Elsas et al. (2011)]. In contrast, survival in sludge is primarily related to temperature, pH, water content (of treated sewage sludge), and sunlight exposure (Sidhu and Toze, 2009). Pertinent biotic interactions include antagonism from indigenous microorganisms and viruses, competition for resources, predation and occupation of niche space (van Elsas et al., 2002). Pathogen-specific biotic factors that influence survival include physiological status and initial inoculum concentration (van Veen et al., 1997).

Following landspreading, there are two main scenarios that can lead to human infection. First, pathogens may be transported via overland or sub-surface flow to surface and groundwater, and infection may arise via (accidental) ingestion of contaminated water (recreational) (Tyrrel and Quinton, 2003). Second, it is possible that viable pathogens could be present on the crop surface following biosolids application, or may become internalised within the crop tissue where they are protected from conventional sanitisation (Solomon et al., 2002). In this case, a person may become infected if they consume the contaminated produce.
2.5.3 Persistent organic pollutants, pharmaceutical and personal care products

The United Nations Environment Programme (UNEP) defines persistent organic pollutants (POPs) as organic compounds that, to a varying degree, resist photolytic, biological and chemical degradation (UNEP, 2013). They are characterised by low water solubility and high lipid solubility, which gives them high potential for bioaccumulation; they also have a long half-life in soil, sediment, air or biota. The lipophilic nature of POPs means that they appear at higher concentrations in fat-containing foods, including fish, meat, eggs and milk, and so traces of POPs are found in the human body (UNEP, 2013). Therefore, food consumption is considered the main pathway of human exposure to POPs for the majority of the population. High lipophilicity results in the substance bio-concentrating from the surrounding medium into the organism; combined with environmental persistence and a resistance to biological degradation, lipophilicity also results in biomagnification through the food chain (Smith and Riddell-Black, 2007). Some cancers, birth defects, dysfunctional immune and reproductive systems, and even some diminished intelligence, are suspected to be related to exposure to these chemicals (UNEP, 2013).

Pharmaceuticals and personal care products refer to any product used by individuals for personal health or cosmetic reasons, or those that are used in agriculture to enhance growth or health of livestock. PPCPs comprise a diverse collection of thousands of chemical substances, including prescription and over-the-counter therapeutic drugs, veterinary drugs, fragrances and cosmetics (Yang et al., 2011). Pharmaceuticals are specifically designed to alter both biochemical and physiological functions of biological systems in humans and animals. However, these features can unintentionally affect soil or aquatic organisms should their habitats become contaminated with these chemicals. Unlike pharmaceuticals, personal care products are directly washed into wastewater during showering and bathing, and thus enter WWTPs and, subsequently, the environment (Richardson et al., 2005). PPCPs are referred to as “pseudo persistent” contaminants (i.e. high transformation/removal rates are counteracted by their continuous introduction into the environment) (Cooper et al., 2008). PPCPs are likely to be found in any body of water influenced by raw or treated wastewater, including rivers, lakes, streams and groundwater, many of which are used as a source of drinking water (Yang et al., 2011), as well as in agricultural soils. In addition, PPCPs with antimicrobial activity may contribute to antimicrobial resistance formation and persistence of antimicrobial bacteria.

2.6 Conclusions

The application of biosolids to agricultural soils can be sustainable and economical on the basis of the recycling of nutrients and the waste disposal of sewage sludge. However, it also has potential risks with respect to the potential for the build-up of contaminants in the soil and the runoff of contaminants. There are also growing concerns that POPs and PPCPs may accumulate in the soil, which can then be taken up by plants and grazing animals and transferred to humans via the food chain. PPCPs are generally more water soluble in nature and tend to have shorter half-lives in soil compared to POPs. However, PPCPs are considered to be “pseudo persistent”, as their supply to the environment via biosolids is continually replenished. The risk of indirect exposure can occur through several pathways (consumption of food-crops, animal uptake of meat or milk, etc.). Risk assessment approaches have been adopted to assess the environmental fate of contaminants in biosolids, with quantitative structure–activity relationship (QSAR) model approaches dominating. Judicious selection of suitable modelling approaches is required to ensure accurate representation of human/environmental risks from emerging contaminants.

While the majority of sewage sludge is reused in agriculture, there are still public perception issues surrounding its use. For example, raw or treated sewage sludges are prohibited from being used on Bord Bia-certified farms (Bord Bia, 2010). In addition, before the publication of this report, no empirical information had been produced regarding the surface runoff of contaminants from land-applied treated sludge in Ireland. Moreover, fears regarding the uptake of metals by grass following land application, and potential incorporation into the food chain, still exist. These factors all have contributed to a general unease regarding the reuse of sewage sludge in agriculture.
3 Metal and PPCP Concentrations in Lime Stabilised, Thermally Dried and Anaerobically Digested Sewage Sludges

3.1 Overview

There is considerable debate and public sentiment about the land application of treated sewage sludges (biosolids). Concerns may be warranted, as many priority metal and PPCP pollutants may be present in biosolids. In this study, metal content and two of the most abundant PPCP components in sewage sludge, the anti-microbials TCS and TCC, were examined in biosolids from 16 WWTPs in Ireland.

For more technical detail on the metal analysis component of this study, the reader is referred to Healy et al. (2016a).

3.2 Introduction

In the EU, land application of biosolids is typically based on its nutrient and metal content, although individual Member States often have more stringent limits than governing directives (LeBlanc et al., 2008; EC, 2010; Milieu et al., 2013a,b,c). Guidelines govern the maximum allowable levels of nutrients and metals (e.g. Fehily Timoney and Company, 1999), although, as the metal content is normally low relative to the nutrient content of biosolids, application rates are frequently determined by the nutrient content of the biosolids and not by their metal content (Lucid et al., 2013). There is a potential risk of metal accumulation in the soil (Mamindy-Pajany et al., 2014), in plants (Latare et al., 2014), or in transport to groundwater, particularly if biosolids are applied in excess (McBride et al., 1999). Parameters that are also of concern are the PPCPs, which cannot be fully removed from WWTPs (Narumiya et al., 2013). Two PPCPs of particular interest are TCS, a broadspectrum bacteriostat and fungicide, and TCC, also a bacteriostat and fungicide, which are known toxins for humans and which are listed among the top contaminants of concern worldwide (Verlicchi and Zambello, 2015).

Due to the increasing awareness regarding potential risks to the environment and human health, the application of sewage sludge, following treatment, to land as a fertiliser in agricultural systems has come under increased scrutiny. As metals and PPCPs probably remain in the soil indefinitely, the characterisation of biosolids for these parameters prior to land application is important. The aim of this study was to: (1) examine if the metal content of biosolids from high population equivalent (PE) WWTPs in Ireland exceeded permitted limit values; (2) to establish a baseline for unregulated metals and potential pollutants of which little is known and from which other global studies may be compared; and (3) characterise the TCS and TCC content of the biosolids.

3.3 Materials and Methods

3.3.1 Sample collection and preparation

Biosolids were collected from 16 WWTPs or agglomerations, with PEs of up to approximately 2.3 million. Selection of the WWTPs was based on willingness to participate in this monitoring study, PE (large PEs were desirable) and geographical location (a good geographical spread was also desirable). Of the WWTPs examined, most received landfill leachate in low quantities [no greater than 2% of the total biological oxygen demand (BOD) loading on the WWTP], while others received industrial, commercial and domestic/septic tank sludge comprising up to 30% of the total influent BOD loading on the WWTP. Eight discrete samples of 100g were collected in clean low-density polyethylene containers (Fisher, UK) from each WWTP and stored at −20°C prior to analysis. The biosolid samples were freeze-dried at −50°C and pulvèrise in an agate ball mill with a rotational speed of 500 rpm for 5 minutes (repeated three times) using an 80 mL agate vial and balls (Ø 10 mm).

3.3.2 Elemental determination

A handheld X-ray fluorescence (XRF) analyser was employed to determine metal concentrations [Cd, Cr, Cu, iron (Fe), mercury (Hg), molybdenum (Mo), Ni, Pb, antimony (Sb), selenium (Se), tin (Sn), and Zn]. This
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A portable XRF system consists of a powerful X-ray tube [4W, gold anode] and a 30 cm² Silicon Drift Detector. An internal instrument standardisation was performed using an alloy chip, and sewage sludge certified reference materials (CRMs) were used for calibration/verification. Quality control included the use of instrumental blanks and analysis of duplicate samples. The performance of the method and stability of the instrument was evaluated by using CRMs for sewage sludge.

3.3.3 Triclosan and triclocarban determination

The method of analysis for TCS and TCC in the biosolids was in accordance with USEPA method 1694 (USEPA, 2007). The analysis was conducted in three stages: (1) proof of limit of detection (LOD) and limits of quantification (LOQ); (2) proof of concept, method implementation and linearity/recovery; and (3) sample analysis. In stage 1, a mixed standard of TCS and TCC was prepared at a level of 0.5 ppm for each analyte and was analysed by LC-MS-MS using reversed phase chromatography, to determine extraction efficiency, demonstrate linearity and determine probable LOD/LOQ in samples. In stage 2, triplicate extractions of spiked biosolids were performed to determine the accuracy and precision of the method. In stage 3, biosolids and surface runoff water samples were extracted using the established method. Quality control monitoring of the analysis was performed using a spiked reference material, which was analysed every 10 samples.

3.4 Results and Discussion

3.4.1 Metal concentrations

The mean concentrations of the metals in the sewage sludge following treatment in the 16 WWTPs are given in Table 3.1. The concentrations of the metals that are regulated in the EU [expressed as mg kg⁻¹ DS (dried solids)] ranged from 11 mg kg⁻¹ (Cd, anaerobically digested (AD) biosolids) to 1273 mg kg⁻¹ [Zn, AD biosolids] and were well below EU regulatory limits. Of the parameters not regulated in the EU, As (arsenic), Se, Mo and Cr were well below the upper limits of 75, 100, 75 and 1000 mg kg⁻¹, respectively. Of the elements considered bioessential micro-nutrients measured in this study (Se, Fe, Cu and Zn), all were within either EU or international limits (no limits govern Fe).

The biosolids from one WWTP, in which anaerobic digestion was carried out, had an average Pb concentration of 3696 mg kg⁻¹, which is well in excess of the threshold value of 1200 mg kg⁻¹. The average concentrations (across all treatments) of Cu, Pb and Zn were also well above the median values of internationally published results. Lead is among the most hazardous metals that are potentially harmful to human health (Johnson and Bretsch, 2002). Other metals measured in this study, which are also potentially harmful, were Cr, Cd, Sn and Sb. Of these parameters, no international standards exist, to date, for Sb or Sn in biosolids for reuse in agriculture. In the current study, the average concentration of Sb ranged from 17 to 20 mg kg⁻¹, which was substantially higher than recorded elsewhere, e.g. <0.01 to 0.06 mg kg⁻¹ (LeBlanc et al., 2008) and 3.4 mg kg⁻¹ (Eriksson, 2001). As the average concentration of Sb in non-polluted soils is around 0.53 mg kg⁻¹ (Fay et al., 2007) and elevated concentrations in the soil inhibit the early growth of crop plants (Fjällborg and Dave, 2004; Baek et al., 2014), the possibility exists that large quantities of this metal are being applied to land without regulation. Tin, in its inorganic form, is non-toxic; however, a significant portion of sewage sludges may be in a highly toxic, organic form and include compounds such as tributyltin (McBride, 2003). The concentrations of Sn measured in this study ranged from 23 to 55 mg kg⁻¹, which was of the same order as other studies (26 mg kg⁻¹; Eriksson, 2001). Normal ranges of Sn in non-polluted Irish soils are around 1.68 mg kg⁻¹ (Fay et al., 2007). Both metals, Sb and Sn, however, are not considered to be of risk to animals or humans (USEPA, 1995).

3.4.2 Triclosan and triclocarban concentrations

The TCS and TCC concentrations in the biosolids samples are shown in Figure 3.1. There was no trend between the type of treatment carried out and the concentrations measured, nor was there any trend with PE served or geographical location of the WWTP. One of the WWTPs examined (1 and 2 in Figure 3.1) had a history of high concentrations of TCS, with concentrations of 25 µg g⁻¹ being previously reported (EPA, 2013), but the concentrations in this study were below this. Of the previous studies that have carried out testing of TCS and TCC across a number of WWTPs in a given region, the concentrations of both compounds measured in this study rank the lowest. There are currently no limits...
Table 3.1. Mean ± standard deviation (SD) metal concentration (mg kg⁻¹ dry weight) in sludge following anaerobic digestion, lime stabilisation, or thermal drying. n refers to the number of treatments

<table>
<thead>
<tr>
<th>Metal</th>
<th>Anaerobic digestion (n=5)</th>
<th>Lime stabilisation (n=4)</th>
<th>Thermal drying (n=8)</th>
<th>EU regularity upper limits (EU, 1986)</th>
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<tr>
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<td>Mean ± SD</td>
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<tr>
<td>Cu</td>
<td>640 ± 411</td>
<td>491 ± 452</td>
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<td>Ni</td>
<td>25 ± 5</td>
<td>13 ± 2.5</td>
<td>15 ± 7</td>
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<tr>
<td>Pb</td>
<td>791 ± 1625</td>
<td>33 ± 25</td>
<td>54 ± 30</td>
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<td>Cd</td>
<td>11 ± 1</td>
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<td>Zn</td>
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Regulated parameters in EU

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Non-regulated parameters in EU

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<td>55 ± 57</td>
<td>23 ± 4</td>
<td>23 ± 5</td>
<td>5</td>
</tr>
<tr>
<td>Sb</td>
<td>20 ± 5</td>
<td>17 ± 3</td>
<td>17 ± 4</td>
<td>4</td>
</tr>
<tr>
<td>Cr</td>
<td>51 ± 43</td>
<td>25 ± 15</td>
<td>16 ± 12</td>
<td>12</td>
</tr>
<tr>
<td>Fe</td>
<td>32,135 ± 41,717</td>
<td>9654 ± 7264</td>
<td>33,087 ± 43,373</td>
<td>12</td>
</tr>
</tbody>
</table>

*Both anaerobic digestion and thermal drying were carried out in one wastewater treatment plant.

aLOD = 10 ppm.

bLOD = 100 ppm.

Figure 3.1. Triclosan and triclocarban concentrations (µg g⁻¹) in treated sludge from 16 wastewater treatment plants in Ireland, ranging (numerically in ascending order) from a PE of 2.3 million to 6500. Note that two forms of treatment of sludge are carried out in one WWTP: AD (WWTP 1) and thermal drying (WWTP 2). WWTPs with no concentrations shown are WWTPs in which triclosan or triclocarban were below the LOD (TCS, 0.006 µg g⁻¹; TCC, 0.0024 µg g⁻¹).
set for PPCPs in sludge or biosolids (Verlicchi and Zambello, 2015), so the concentrations measured in the current study may only be compared with similar studies. The biosolid samples used in this study were collected in January and February of 2015, so there may be seasonal variation in the concentrations of TCS and TCC that were measured in other studies (Martin et al., 2012).

3.4.3 Environmental policy and management implications

Land application of biosolids is, in the main, determined by the nutrient content of biosolids and not by the metal or PPCP content. Therefore, accumulation of metals and PPCPs in soil following repeated applications of biosolids may be problematic, particularly for those elements that are not regulated and are harmful to human health. Guidelines should aim to govern the maximum allowable concentrations of these elements in biosolids, as well as the land to which they are applied. Handheld XRF analysis is a useful, quick and relatively inexpensive method for determining the metal content of biosolids and should be used frequently to characterise it. In contrast, PPCP testing is extremely expensive, so the costs of routinely testing sludge for all possible contaminants would be prohibitive.

3.5 Conclusions

The concentration of metals detected in 16 WWTPs in Ireland were below the maximum allowable concentrations of metals for use in agriculture in the EU. In addition, they were also within the median levels for biosolids globally. While current EU and international regulations govern certain priority metal pollutants and bioessential elements, other metals that are potentially harmful to human health, such as Se and Sn (both of which were present in the treated sewage sludge in this study), are omitted from the regulations. This means that a number of toxic metals, at levels much higher than their baseline concentrations in soils, are being applied without regulation.

While the metal, TCS and TCC contents of the biosolids in the WWTPs examined were below the concentrations measured elsewhere, there is a possibility that this may increase from one season to the next. In addition, until threshold values, based on human or ecological risk, are set, there is a possibility that the concentrations of TCS and TCC, even at the relatively low concentrations measured in the current study, may be considered to be a risk. Furthermore, the current study only examined two types of PPCPs (albeit the ones that are the most abundant and problematic of the PPCPs worldwide), which are only a small fraction of the total number of contaminants that may be present in biosolids.

To fully characterise all existing known contaminants, as well as emerging contaminants, is cost prohibitive. Therefore, any potential economic and practical gains arising from the recycling of sewage sludge in agriculture need to be considered alongside cost and public health issues. A significant component of the cost of performing such analyses is related to the development and validation of approaches for analyte extraction, detection and quantification. Differences in methodology and lack of data on inter-laboratory comparability also limit confidence in assessing the significance of reported differences in levels of contaminants. Harmonisation/standardisation of methods and development of standards may help to reduce costs and enhance comparability.

It is recommended that, in the first instance, priority elements and TCS and TCC are assessed in sewage sludge in Ireland. Following this, regulations may be extended to cover these parameters.
4 Nutrient, Metal, Microbial, Triclosan and Triclocarban Loss in Surface Runoff Following Application to Grassland Soil

4.1 Overview
Treated municipal sewage sludge (biosolids) may be applied to agricultural land as an organic fertiliser. This study investigates losses of nutrients (N and P), metals (Cu, Ni, Pb, Zn, Cd, Cr), two selected PPCPs (TCC and TCS) and microbial indicators of pollution (total and faecal coliforms) arising from the land application of four types of treated biosolids to field micro-plots. This was done during 60-minute simulated rainfall events at three time intervals (24, 48, 360 hours) after application. To contextualise the results, the plots were also treated with DCS, which was applied at the same rate as the biosolids.

For more technical details on this study, the reader is referred to Peyton et al. (2016).

4.2 Study Site Description
The study site was a 0.6-ha grassland plot located at Teagasc, Johnstown Castle Environment Research Centre, Co. Wexford, Ireland (latitude 52.293415, longitude –6.518497). The area has a cool maritime climate, with an average temperature of 10ºC and a mean annual precipitation of 1002 mm. The site has been used as a grassland sward for over 20 years, with nutrient inputs (organic and inorganic) applied based on routine soil testing. The site has undulating topography with average slopes of 6.7% along the length of the site and 3.6% across the width. Overall, the site is moderately drained with a soil texture gradient of clay loam to sand silt loam, as classified by Brennan et al. (2012). Physico-chemical characterisation of the site is detailed in Peyton et al. (2016).

4.3 Micro-plot Installation and Characterisation
Thirty grassland micro-plots (Figure 4.1), each 0.9 m in length and 0.4 m in width (0.36 m²), were isolated using continuous 2.2 m-long, 100 mm-wide rigid polythene plastic strips, which were pushed to a depth of 50 mm into the soil to isolate three sides of the plot. A 0.6-m polypropylene plastic runoff collection channel was fitted at the end of each plot. Micro-plots were oriented with the longest dimension in the direction of the slope. Once installed, plots were left uncovered to allow natural rainfall to wash away any soil that had been disturbed during their construction. Physico-chemical characterisation of the micro-plots, including textural analysis, soil nutrient status, metal and microbial content, is detailed in Peyton et al. (2016).

4.4 Biosolids Characterisation
Three types of biosolids were examined in this study: two types of AD sludge, one sourced from a WWTP in Ireland [anaerobically digested biosolids sourced in Ireland (ADIRE)] and another used in an EU-funded FP7 (Seventh Framework Programme) project (END-O-SLUDG, 2014) [anaerobically digested biosolids sourced in the UK (ADUK)]; thermally dried (TD) biosolids; and lime stabilised (LS) biosolids. With the
exception of ADUK, all biosolids were sourced from the same WWTP in Ireland. As the Irish WWTP only employed two methods to treat sludge (AD and TD), an untreated, dewatered sewage sludge cake was also collected from the same WWTP, so that it could be manually lime treated. The treated sludge and the dewatered sludge cake were collected in sealed, 50L capacity plastic storage boxes and transported to the Teagasc Environment Research Centre, where they were labelled and stored at 4°C. In accordance with standard methods in Ireland (Fehily Timoney and Company, 1999), lime (calcium oxide) was added to the raw dewatered sewage sludge to raise the pH to greater than 12 and to generate heat. If this had not been done, the sludge would not have been adequately treated and would have carried greater risk. The treated sludge samples (each at \( n = 3 \)) were tested for DM, N (Kjeldahl, 1883), P, K and metal content (Cu, Ni, Pb, Zn, Cd, Cr, Hg) (Peyton et al., 2016). Water extractable P was tested after Kleinman et al. (2007). In addition, the biosolids samples (each at \( n = 3 \)) were tested for TC and FC immediately after collection, using the same methods as for soil (Peyton et al., 2016).

4.5 Slurry Characterisation

Dairy cattle slurry was collected from the dairy farm unit at Teagasc, Johnstown Castle Environmental Research Centre. The storage tanks were agitated and slurry samples were transported to the laboratory in 25-L drums. Slurry samples were stored at 4°C prior to land application. Slurry pH was determined using a pH probe and a 2:1 ratio of deionised water to soil. The DCS was tested for: DM, N (Kjeldahl, 1883), P, K and metal content (Cu, Ni, Pb, Zn, Cd and Cr) (Peyton et al., 2016). In addition, the DCS samples (each at \( n = 3 \)) were also tested for TC and FC immediately after collection using the same methods as for soil (Peyton et al., 2016).

4.6 Rainfall Event Simulation and Application

Amsterdam drip-type rainfall simulators, as described by Bowyer-Bower and Burt (1989), were used to provide rainfall in this study (Figure 4.2). It was designed to form droplets with a median diameter of 2.3 mm, spaced 30 mm apart in a 1000 mm × 500 mm × 8 mm Perspex plate over a 0.5 m² simulator area. The simulator was calibrated to deliver a rainfall intensity of 11 mm h⁻¹. A video of the operation of the rainfall simulator (“Rainfall simulator”) is available online (https://youtube.com/watch?v=JYhsmE8SHvU). Water samples used in the rainfall simulations, were collected over the duration of the three rainfall events; the average concentrations are detailed in Peyton et al. (2016).

The six treatments (four biosolids, one DCS and one soil-only study control) used in this study were assigned to 30 micro-plots by dividing the plots in five blocks (five blocks each containing six micro-plots, so that each treatment was replicated \( n = 5 \) times). As metal content was not limited in soil, DCS or biosolids application to the micro-plots was governed by the P content of the biosolids, and DCS and the P by the index of the soil. For comparable results, all micro-plots were classified into Index 2P soil, which meant that all biosolids and DCS treatments were applied to all plots at a rate of 40 kg Pha⁻¹ (Coulter and Lalor, 2008). As a result of the P content and the DM of each individual biosolid, application rates per designated, individual plot were 96.6 g of TD, 242.2 g of ADIRE, 1063.3 g of LS, 243.9 g of ADUK. The DCS was spread at 2880 g per individual plot.

Prior to application, grass on all plots was cut to 50 mm 48 hours before the first rainfall simulation (RS1). For better control of rainfall simulations and to prevent runoff losses caused by natural rainfall events, individual micro-plots were covered from the time of grass cutting to the end of the last rainfall event by “rainout” shelters (Hoekstra et al., 2014). Surfaces of biosolids were applied to each micro-plot by hand. To ensure even distribution, each micro-plot was divided into four quadrants (each 0.09 m² in area) and a proportionate amount of biosolids was applied in each quadrant. The DCS was applied in rows using a watering can to replicate normal trailing shoe application. The biosolids and DCS were then left for 24 hours on the soil before RS1. The RS1 occurred 24 hours after biosolids and DCS application, so as to demonstrate losses representative of a worst case scenario. The second rainfall simulation event (RS2) was 2 days (48 hours) after initial biosolids/DCS application, which was representative of current legislation, and the third rainfall simulation event (RS3) took place 15 days (360 hours) after initial application.

Volumetric water content of the soil in each plot \( (n=3) \) was measured immediately prior to each rainfall event.
using a time domain reflectometry device (Delta-T Devices Ltd., Cambridge, UK), which was calibrated to measure resistivity in the upper 50 mm of the soil in each plot. Prior to each rainfall event, collection channels from the micro-plots were also rinsed with boiling hot water to decontaminate them.

4.7 Runoff Sample Collection
Surface runoff was judged to occur once 50 mL of water was collected from the runoff collection channel from the start of simulated rainfall to runoff. The collection of the first 50 L \((t=0)\) was used to indicate time to runoff \((TR)\) and was used as part of the microbial analysis. Samples for nutrient and metal analysis were collected every 10 minutes \((t=10, t=20, t=30)\) from TR to allow for the flow-weighted mean concentration \((FWMC)\) to be calculated \((\text{Brennan et al., 2012})\). After this time, another 50 mL of surface runoff water was collected for microbial analysis, so that it could be bulked with the first 50 mL of runoff to create a 100 mL sample for microbial analysis. Immediately after collection, all samples were stored in cool boxes with ice until they were returned to the laboratory for analysis. For TCS and TCC analyses, runoff water samples (from the TD, ADIRE and LS plots) were collected in solvent washed amber glass Pyrex bottles with PTFE-lined lids and were decanted into clean amber bottles to remove any sediment that was present in the samples on returning to the laboratory. Immediately after decanting, 4 mol L\(^{-1}\) of sulfuric acid was added to adjust the water to pH 3 to prevent biodegradation by microorganisms. Samples were then frozen and stored at –20°C until analysis.

4.8 Nutrient and Metal Runoff Analysis
Runoff water samples were filtered through 0.45 μm filters (Sarstedt-Filtropur S 0.45) and a sub-sample was analysed calorimetrically for dissolved reactive phosphorus \((\text{DRP})\), \(\text{NO}_3^-\text{N}\), \(\text{NO}_2^-\text{N}\) and \(\text{NH}_4^-\text{N}\) using a nutrient analyser (Aquachem Labmedics Analytics, Thermo Clinical Labsystems, Finland). A second filtered sub-sample was analysed for total dissolved phosphorus \((\text{TDP})\) using acid persulfate. Unfiltered runoff water samples were analysed for TP with an acid persulfate digestion and total reactive phosphorus using the Aquachem Analyser. Metal analysis was tested on the filtered samples using inductively coupled plasma optical emission spectroscopy. Particulate phosphorus \((\text{PP})\) was calculated by subtracting TDP from TP. The DRP was subtracted from the TDP to give the dissolved

![Figure 4.2. Amsterdam drip-type rainfall simulators used in this study.](image)
unreactive phosphorus (DUP). All samples were tested for P, N and suspended sediment in accordance with the Standard Methods (APHA, 2005) and TCS and TCC were tested in accordance with USEPA Method 1694 (USEPA, 2007).

4.9 Total and Faecal Coliform Analysis

Samples (2 × 50 mL aliquots) of runoff water were collected at the start and towards the end of rainfall simulation experiments, and were stored in cool boxes (4°C) filled with ice until they were returned to the laboratory for analysis. The time interval between the first collection and analysis was always less than 9 hours. Samples were appropriately diluted using sterile isotonic phosphate buffered saline (Osoid, UK) made with water from a Millipore automatic sanitisation module and 100-mL aliquots were apportioned for analysis in accordance with standard methods (APHA, 2005). Total and faecal coliforms were enumerated using the IDEXX Coilsure Quanti Tray/2000 method (IDEXX Laboratories, Westbrook, ME) after incubation at 37 ± 0.5°C degrees for 24 hours. Results were expressed as the most probable number (MPN) of TC and FC per 100 mL.

4.10 Data Analysis

The structure of the data set was a blocked one-way classification (treatments) with repeated measures over time (RS1–RS3). The analysis was conducted using Proc Mixed in SAS software (SAS, 2013) with the inclusion of a covariance model to estimate the correlation between rainfall events. Details of the statistical treatments employed are in Peyton et al. (2016).

4.11 Results

4.11.1 Nutrient losses in surface runoff

the average FWMC of TP, comprising DUP, PP and DRP, for all treatments and rainfall events is shown in Figure 4.3. The application of TD and ADIRE biosolids and DCS significantly increased the average FWMC of DRP in RS1 and RS2 compared to the study control, but this highly mobile P fraction was low for the other biosolids treatments. The highest median FWMC of DRP in the biosolids treatments (0.86 mg L⁻¹) was measured during RS1 for TD-amended plots and this decreased significantly (p = 0.02) over subsequent rainfall events to 0.14 mg L⁻¹ for RS3. In comparison, the median FWMC of DRP from the ADIRE treatment was highest for RS2 (0.78 mg L⁻¹), although results for the three events were not significantly different. However, losses for DRP from biosolids treatments were low compared to the DCS. Dissolved reactive phosphorus loss for DCS during RS1 was 7.0 mg L⁻¹ and this remained higher than any of the biosolids treatment losses during all simulation events.

Losses of PP were detected across all treatments, including the study control. Particulate P comprised >45% of TP losses for ADUK, ADIRE and LS biosolids, and the study control. PP losses comprised only 14% of TD biosolids, due to the high proportion of DRP losses, and were much lower than the losses from the DCS plots.

The average FWMC of total nitrogen (TN) across all treatments is shown in Figure 4.3. There was a significant interaction between treatment and the rainfall simulation for NH₄-N. The application of all biosolids treatments increased the average FWMC of NH₄-N for RS1, compared to the study control, and while there was a downwards trend between RS1 and RS3 for all treatments except the control, the decrease was not significant for LS. The ADUK-amended plots had the highest FWMC of surface runoff of NH₄-N for all biosolids treatments in RS1 (15.3 mg L⁻¹). TD and ADIRE treatments had the next highest FWMCs of NH₄-N, but these were not significantly different from each other or from the LS runoff during RS1.

4.11.2 Metal losses in surface runoff

The average FWMC of metals (Cu, Ni, Pb, Zn, Cd, Cr) in runoff are shown in Figure 4.4. All runoff samples, except the surface runoff of Cu from the LS plots, were below their respective surface water standards intended for human consumption (S.L.549.21). With the exception of Cu, the surface runoff of all the parameters measured was of the same order, or much less than, the DCS. For Cu, the LS-amended plots had significantly higher FWMCs than all other treatments (p < 0.001), with the highest median concentration of 202 µg L⁻¹ measured during RS1. There was a decreasing trend in Ni concentrations across all treatments from RS1 to RS3, except for the study control, but there were no significant differences within treatments. All Ni concentrations were elevated in comparison to the study...
control. The highest median FWMC for Pb (1.5 \( \mu g L^{-1} \)) was measured during RS3 for the DCS and the second highest was 0.82 \( \mu g L^{-1} \) during RS1 for TD-amended plots. However, there was no significant difference between the treatments and the study control. The highest median FWMC of Zn (30.8 \( \mu g L^{-1} \)) was during RS1 for DCS-amended plots, but there were no significant differences across treatments or events.

4.11.3 Microbial losses in surface runoff

The average losses of TC and FC are shown in Figure 4.5. The ADUK-amended plots produced runoff with the lowest number of TC (averaged over the three rainfall simulations), but produced the highest average number of FC: \( 7.1 \times 10^3 \) MPN per 100mL during RS1 and RS2. While median losses from the TD-amended
plots increased with successive rainfall events from $1.9 \times 10^5$ MPN per 100 mL during RS1 to $1.0 \times 10^6$ MPN per 100 mL during RS3, there were no significant differences within treatments. Overall losses from DCS ($3.1 \times 10^2$ MPN) were greatest and significantly greater than LS, ADIRE and the study control. ADUK losses ($1.7 \times 10^2$ MPN) were not statistically different from DCS, but they were significantly greater than the control ($p = 0.009$). The highest median count of TC and FC measured in LS biosolid-amended plots was $5.6 \times 10^5$ and $1.5 \times 10^6$ MPN per 100 mL, respectively. The highest median loss of TC for DCS-amended plots was $1.5 \times 10^6$ MPN per 100 mL.

Figure 4.4. Flow-weighted mean concentrations of (a) Cd, (b) Cr, (c) Cu, (d) Ni, (e) Pb and (f) Zn in the runoff over three successive rainfall events at 24 hours (RS1), 48 hours (RS2) and 360 hours (RS3) after application to grassland.
4.11.4 Triclosan and triclocarban losses in surface runoff

The surface runoff concentrations of TCS and TCC were below the LOD in all cases, with the exception of TD biosolids at 360 hours (0.01 µg L⁻¹) and LS biosolids (0.02 µg L⁻¹) one day after application (Table 4.1). These TCS and TCC concentrations in the surface runoff were lower than values observed in similar studies and below the concentrations at which biota are considered to probably be or are known to be potentially impacted.

4.12 Discussion

4.12.1 Incidental nutrient losses

With the exception of LS biosolids, FWMCs of TP and DRP across all treatments were significantly higher than the study control and, in some cases, were in breach of maximum admissible concentrations for surface water. The volumetric water content of all study micro-plots was approximately 40% and the runoff ratio (the volume of runoff as a percentage of the volume of water applied...
Table 4.1. Concentrations of triclosan and triclocarban in applied biosolids to field plots (“Influent”) and average concentrations of triclosan and triclocarban in surface runoff (µg L⁻¹) from field plots. LOD = 0.09 µg L⁻¹ (TCS) and LOD = 0.006 µg L⁻¹ (TCC) in this study

<table>
<thead>
<tr>
<th>Triclosan</th>
<th>24-hour</th>
<th>48-hour</th>
<th>360-hour</th>
<th>Triclocarban</th>
<th>24-hour</th>
<th>48-hour</th>
<th>360-hour</th>
</tr>
</thead>
<tbody>
<tr>
<td>Influent</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.05</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.01</td>
</tr>
<tr>
<td>TD</td>
<td>4.9</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td></td>
<td>0.02</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
<tr>
<td>LS</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td></td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
<tr>
<td>ADIRE</td>
<td>0.27</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td></td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
</tbody>
</table>

...to each micro-plot) was broadly similar across treatments (data not shown). Therefore, the nutrient load from each micro-plot was proportional to the FWMCs.

The FWMCs of TP and TN generally decreased across successive rainfall events. This trend was similar to several studies that have examined runoff of nutrients resulting from the land application of different types of biosolids and DCS (Rostagno and Sosebee, 2001; Penn and Sims, 2002; Ojeda et al., 2006; Eldridge et al., 2009; Lucid et al., 2014). The DRP losses measured in the current study were proportional to the water-extractable phosphorus (WEP) of the biosolids. Several studies have shown that WEP is an effective quantitative indicator of dissolved P losses from surface applied biosolids (Kleinman et al., 2002, 2007; Elliot et al., 2005). Thermally dried and ADIRE biosolids, which also had high WEPs, had the highest losses of dissolved P from their respective plots.

All biosolids treatments had elevated FWMCs of NH₄-N in runoff compared to the study control across all rainfall simulations, whereas the study control and biosolid-amended plots had the same NO₃-N concentrations. Ammonium can be volatilised (or rapidly mobilised by runoff and leaching) after OM spreading (Quilbé et al., 2005). ADUK biosolids, which had the highest initial NH₄-N concentration of the biosolids at the time of application (3846 mg kg⁻¹ DM; Peyton et al., 2016), also had the highest FWMC of NH₄-N in runoff compared to biosolids treatments during RS1. Similar trends were noted for the ADIRE and LS biosolids. However, the initial concentration of NH₄-N in TD biosolids before application (573 mg kg⁻¹) was lower than the ADIRE biosolids (3428 mg kg⁻¹), but had similar losses of NH₄-N in surface runoff during RS1. These types of anomalies may be due to the consistency of the biosolids, which means that different types of biosolids will have varying surface area exposure to rainfall. Therefore, TD biosolids could possibly be more easily diluted and transported in the runoff compared to the ADIRE, ADUK and LS biosolids, due to their finer particle granulated consistency. This is also the reason for the high proportion of runoff measured for the DCS; it had the highest FWMC of NH₄-N and DRP. A possible reason for this is that the DCS had a DM composition of 8% and was highly mobile following an episodic rainfall event. This study showed that biosolids, although having a higher DM than DCS, are not as easily mobilised.

4.12.2 Incidental metal losses

The concentrations of metals, excluding Cu for LS biosolids, in runoff were below surface water standards intended for human consumption (S.L. 549.21, 2002). Similar results have been reported for several runoff studies using different types of biosolids at higher application rates than the current study (Dowdy et al., 1991; Joshua et al., 1998; Eldridge et al., 2009; Lucid et al., 2013). This shows that the codes of good practice for the use of biosolids in agriculture (Fehily Timoney and Company, 1999) are appropriate in limiting metal application and, therefore, losses to water bodies. The metal content in the biosolids was not the limiting factor for the spreading rate and the soil metal content was also below maximum permissible guidelines (Fehily Timoney and Company, 1999). The soil pH and clay content were within the recommended guidelines set out in the code of good practice (Fehily Timoney and Company, 1999).

While there was generally low FWMC of metals across all rainfall simulations, the LS biosolid-amended plots released the highest quantity of Cu, Ni and Zn compared to other plots. One possible explanation for this is that Cu, Ni and Zn are more soluble metals (Joshua et al., 1998), and as LS biosolids consist of larger sized particles of a more compact consistency, time to runoff increased (results not shown). This gave these metals more contact time to dissolve and to subsequently be...
released compared to the other biosolids treatments. Metal concentration was low in DCS in comparison to the biosolids before application and, as a result, did not cause excessive losses of metals in runoff. However, the FWMCs of Cd and Cr in DCS-amended plots were higher than any of the biosolids plots, with peak concentrations of 1.68 µg L⁻¹ during RS3 for Cd and 3.89 µg L⁻¹ during RS1 for Cr.

4.12.3 Incidental triclosan and triclocarban losses

The low concentrations of TCS and TCC in surface runoff may have been a function of the low TCS and TCC concentrations in the biosolids applied to land relative to similar studies, but were probably either due to their degradation or transformation to other compounds or due to the soil characteristics at the study site. This suggests that TCS and TCC may build up in the soil over continuous applications and may be incorporated into crop biomass (Mathews et al., 2014). The relationship between persistence of PPCPs and the composition and physico-chemical properties of soil is well established in the literature (Verlicchi and Zambello, 2015). As reported in other studies (e.g. Wu et al., 2009), the high soil OM content in the micro-plots of the current study (8.1–9.0%) may have adsorbed some of the TCS and TCC.

4.12.4 Incidental losses of indicator microorganisms

Understanding the environmental persistence and fate of the introduction of enteric microorganisms following land application of biosolids and organic amendments is necessary, as it provides a sound scientific basis for management practices designed to mitigate the potential microbiological health risks associated with spreading on agricultural land (Lang et al., 2007). The risk associated with biosolid-derived microorganisms and other organic-amendment microorganisms is largely determined by their ability to survive and maintain viability in the soil environment after land spreading. In general, enteric pathogens are poorly adapted for survival in a soil environment and pathogens that are land applied from biosolids are influenced by climatic and agronomic variables (Lang et al., 2003). Others have reported the persistence of enteric microorganisms in soils (Brennan et al. 2010). When biosolids are incorporated into the soil, microorganism survival is affected by factors such as pH, OM, soil texture, temperature, moisture content and competition with other microorganisms (Lang et al., 2007). These factors were reviewed by Erickson et al. (2014). However, when biosolids are surface applied, as in the current study, desiccation and UV light are the key factors in the decay of pathogens (Lu et al., 2012). Desiccation of pathogens is influenced by the soil and biosolids moisture content. In the current study, soil moisture remained constant at approximately 40%, which was unlikely to affect pathogen survival or regrowth. However, as the rainfall simulator provided moisture to the biosolids, there may have been regrowth of the FC in the ADIRE and LS biosolids between RS1 and RS2. Similar FC regrowth in AD biosolids was also reported by Zaleski et al. (2005). All TC and FC in biosolids had decayed by RS3, which was most likely due to the desiccation of microorganisms rather than the influence of UV, as all plots were covered by the rainout shelter, which prevented natural rainfall between RS2 and RS3.

ADUK biosolids had significantly higher concentrations of FC in runoff during RS1 and RS2 compared to other treatments. At the start of the experiment, the ADUK biosolids were above the recommended standards of 1 × 10⁶ MPN g⁻¹ (Fehily Timoney and Company, 1999), and, as a result, were equivalent to Class B microbial matter under the USEPA Part 503 Regulations (USEPA, 1993), which allows detectable levels of FC up to 2 × 10⁴ MPN g⁻¹ DS. All the Irish biosolids were some 10-fold below the Class A Irish standard (Peyton et al., 2016).

It is important to evaluate the risks arising from the application of biosolids to land in relation to other common agricultural practices, such as the application of animal waste on land (Vinten et al., 2004), which is commonly spread as an organic fertiliser. Hubbs (2002) reported that land application of DCS as a fertiliser had FC concentrations in surface runoff of up to 1.2 × 10⁵ cfu (colony-forming units) per 100 mL 2 days after application. Furthermore, after five rainfall events over 30 days, the mean FC concentrations in runoff, although decreasing, remained at high levels compared to the biosolids in the same study (4.0 × 10⁴ cfu per 100 mL). This was also observed in the current study, as the DCS had the second highest FC during RS1 and RS2; however, it was the highest by RS3, showing that FC survive for a longer period in DCS compared to biosolids. This may result in loss of microorganisms to water bodies for a longer period following application. Moreover, Payment
et al. (2001) found that the pathogen concentration was lower in untreated sludge ($3 \times 10^2$ to $6 \times 10^2$ cfu g$^{-1}$) compared to fresh and stored cattle slurries ($7.5 \times 10^4$ to $2.6 \times 10^8$ cfu g$^{-1}$) (Hutchison et al., 2004). When considered within this context, the risk of infectious diseases arising from the land application of biosolids appears to be low in magnitude. This study also did not provide buffering capacity to the runoff samples and overland flow was not sampled at delivery end of the transfer continuum; therefore, the bacterial results represent a worst case scenario.

While this and many other studies focus on the TC group as an indicator of the presence of pathogens, relying on them does have the drawback that they are a poor indicator of the presence of viruses and parasitic protozoa, which may survive for much longer periods (NHMRC, 2003).

### 4.13 Conclusions

The results of this plot-scale study show that there were elevated losses of nutrients, TC and FC from biosolid-amended plots compared to unamended plots. However, nutrient and microbial losses were higher from DCS-amended plots. The metal concentrations in surface runoff of Cr, Ni, Pb and Zn were below their respective surface water limits for both biosolids and DCS, and TCS and TCC concentrations were below the limits of detection (90 ng L$^{-1}$ for TCS, 6 ng L$^{-1}$ for TCC). The surface runoff concentrations measured in this study represented a worst case scenario for potential losses, as further buffering (e.g. in open ditch networks with high P sequestration capacity) may be possible further down the transfer continuum. This study was conducted at a micro-plot scale, but the results should be verified at field-scale. In addition, future work should also be carried out to assess other emerging contaminants that may be present in biosolids and to assess the comparative impact of biosolids and cattle slurry on environmental levels of specific associated pathogens, such as verotoxigenic *Escherichia coli* (*E. coli*), *Cryptosporidium parvum* and *Norovirus*. Notwithstanding these caveats, these results are significant as they show that fears over elevated losses of nutrients, TCS, TCC, metals and microorganisms may be unfounded.
5 Metal Concentrations in Ryegrass Following a Single Application of Lime Stabilised, Thermally Dried and Anaerobically Digested Sludge

5.1 Overview

This chapter measures the uptake of metals by ryegrass following land application of biosolids. For more technical details on this study, the reader is referred to Healy et al. (2016b).

5.2 Introduction

While numerous studies have examined issues surrounding surface runoff water quality following biosolids application to land (Quilbé et al., 2005; Ojeda et al., 2006; Eldridge et al., 2009; Lucid et al., 2014) and the subsequent uptake of metals by plants (Dijkshoorn et al., 1981; Antoniadis et al., 2008), to our knowledge no study has examined the relative impact of the land application of various types of biosolids (originating from the same WWTP) on metal uptake by Lolium perenne (ryegrass), a common grass species generally used for cattle grazing. As different types of treatments are used prior to land application, such a study may inform policy on land spreading and on appropriate crop harvesting and cattle grazing exclusion times.

Regulations governing the land application of biosolids frequently place constraints on crop harvesting and cattle grazing subsequent to land application of biosolids. In Ireland, for example, no animal fodder, including kale, fodder beet or silage, may be harvested until at least 3 weeks after application of biosolids and cattle should not be turned out onto a pasture that has been fertilised with biosolids for 3 to 6 weeks after the date of application (Fehily Timoney and Company, 1999).

The aims of this study were to examine (1) if biosolids application to ryegrass at the legal rate in Ireland increases the metal content of the plant biomass; (2) if the method used to generate the biosolids results in different metal bioavailability and subsequent uptake and bioaccumulation rates in ryegrass; and (3) if the metal content of the ryegrass following the application of biosolids reduces as the ryegrass plants grow. To address these aims, biosolids were applied to ryegrass at the legal permissible rate in Ireland and ryegrass samples were collected and tested for metal content at time intervals of up to 18 weeks from the time of application.

5.3 Materials and Methods

5.3.1 Study site and instrumentation of micro-plots

Twenty-five grassland micro-plots, each 0.9 m in length and 0.4 m in width (0.36 m²), were isolated using continuous 2.2 m-long, 100 mm-wide rigid polythene plastic strips, which were pushed to a depth of 50 mm into the soil to isolate three sides of each micro-plot. A 0.6-m polypropylene plastic runoff collection channel was fitted at the end of each micro-plot to facilitate surface runoff. Micro-plots were orientated with the longest dimension in the direction of the slope.

5.3.2 Biosolids application to plots

Three types of biosolids were examined in this study: two types of AD sludge, one sourced from a WWTP in Ireland (AD) and another used in an EU-funded FP7 project (ADUK) (END-O-SLUDG, 2014), and TD and LS biosolids. With the exception of ADUK, all biosolids were sourced from the same WWTP in Ireland.

5.3.3 Rainfall event simulation and application

Legislation governing the application of any type of organic or inorganic waste in Ireland states that there should be a period of 48 hours between the land application and the first rainfall event (S.I. 610 of 2010). As the greatest amount of organic, nutrient, metal and microbial matter is released from the soil surface in rainfall events close to the time of waste application, an Amsterdam drip-type rainfall simulator (as discussed in Chapter 4) was used in this experiment. This way the rainfall events, intensity and durations could be controlled for the first 360 hours after biosolids application.
The five treatments (four biosolids and one soil-only study control) used in this study were assigned to 25 micro-plots by dividing the plots in five blocks (each block containing five micro-plots on which each of the five treatments were examined). Biosolids application to the micro-plots commenced in January 2014 (winter time in Ireland). The biosolids were applied to each micro-plot at a rate of 40kg P ha\(^{-1}\) (0.04 tonne P ha\(^{-1}\)) and the rate of application was governed by the P content of the biosolids and the P concentration (agronomic soil P index) of the soil.

Prior to application, grass on all micro-plots was cut uniformly to 50mm using plastic hand shears, 48 hours before the first rainfall simulation. Biosolids were applied to the surface of each micro-plot by hand and to ensure even distribution, each micro-plot was divided into four quadrants (each 0.09 m\(^2\) in area) and a proportionate amount of biosolids was applied in each quadrant. The biosolids were then left on the soil for 24 hours before the first rainfall simulation. The first rainfall simulation event occurred 24 hours after the application of biosolids, which was less than the 48 hours legal limit; however, this allowed a worst case surface runoff scenario to be evaluated. The second rainfall event took place 2 days (48 hours) after initial biosolids application, which was representative of current legislation, and the third took place 15 days (360 hours) after initial biosolids application. During the period between these three rainfall applications, a “rainout” shelter was used (Hoekstra et al., 2014). A rainout shelter is a large, plastic shelter on a steel frame that protects the soil from direct rainfall, while allowing air to circulate over the soil surface. After 360 hours (the time of the third rainfall simulation), the micro-plots were exposed to normal weather conditions.

### 5.3.4 Collection of grass samples

Three grass samples, each comprising a composite of six to eight blades or shoots of ryegrass, were cut at the soil surface in each of the 25 micro-plots immediately prior to the second (at 48 hours after application) and third (at 360 hours after application) rainfall simulations, and finally at a time varying between 55 and 130 days from the time of the application of biosolids. This variation was due to the staggered nature of the initial application of biosolids to the micro-plots and the fact that the final collection of grass blades occurred on the same day in June 2014. Nitrile gloves were used to collect the grass samples and the gloves were changed between plots to avoid cross-contamination. Each grass sample was thoroughly washed with ultrapure water (18.3 mΩ, Milli-Q Element System, Merck Millipore, Cork, Ireland) to remove any particulate or adhered material, before being placed into separate, clean, sealed bags and frozen at –20ºC, after which they were transported to the laboratory.

### 5.3.5 Preparation and analysis of ryegrass samples

The ryegrass was freeze-dried at –52ºC (Freezone 12, Labconco, Kansas City, MO, USA). Approximately 0.1g of the sample was digested with an optimised microwave digestion procedure (Anton Paar Multiwave 3000, Graz, Austria) using 3L of trace metal grade 67–69% HNO\(_3\) (ROMIL-Spa™, USA) and 3L of 30% H\(_2\)O\(_2\) (TraceSELECT® Ultra ≥30%, SIGMA-ALDRICH, USA) (Morrison et al., 2008). The digested samples were transferred into trace metal-free centrifuge tubes (Labcon, Petaluma, CA, USA). The determination of Cr, Cu, Pb, Ni, Zn, Cd, B (boron), Co (cobalt), Fe, Mn (manganese), Mo, Al (aluminium), V (vanadium), As, Nb (niobium), Sb, Ba (barium), and W (tungsten) in the ryegrass digests were performed on a PerkinElmer ELAN DRCe (Perkin Elmer, Waltham, USA) inductively coupled plasma mass spectrometer using both standard and dynamic reaction cell mode (Staunton et al., 2014; Ratcliff et al., 2016) in a class1000 (ISO class 6) clean room. A CRM of ryegrass (ERM® – CD281 RYE GRASS, European Commission, Joint Research Centre, Institute for Reference Materials and Measurements, Belgium) was used with method blanks and duplicate samples for method validation and quality assurance purposes.

### 5.3.6 Statistical analysis

The data were assessed using analysis of variance (ANOVA). Minitab statistical software (Pennsylvania, USA) was used for the analysis, with a two-sided confidence interval level set at 95%. Variances were not assumed to be equal across treatments and control, and consequently the Welch’s ANOVA test was utilised in the Minitab software. The same approach was used to test the statistical significance of the observed reduction in metal content with test program duration across all treatments and control.
5.4 Results and Discussion

The average concentrations of metals that are subject to legislation in Ireland in the ryegrass for each treatment over the duration of the study are displayed in Figure 5.1. The reader is referred to Healy et al. (2016b) for graphs of unlegislated metal concentrations. For most metals examined, there was no statistically significant difference between the biosolids treatments and the study control except for Cu (ADUK biosolids; from 360 hours onwards) and Ni (LS biosolids; at 360 hours).

As the application rate of the biosolids to each micro-plot was determined by their P content, and not by their metal content, application of metals was different between treatments. As a result, for example, the application rate of Cu was much higher for LS and AD biosolid-amended micro-plots (119 and 183 mg per micro-plot, respectively) than the other micro-plots (70 mg for ADUK-amended micro-plots and 49 mg for TD-amended micro-plots). Over the 18-week duration of the study, and with the exception of Ni, Cu and Al,

![Figure 5.1](image-url)

Figure 5.1. Measured metal content of legislated metals in Ireland (Ni, Zn, Cr, Pb, Cu) in ryegrass up to 3120 hours (130 days) after a single application of either ADUK or AD originating from Ireland (AD), LS or TD biosolids. Cadmium is not displayed as it was below the LOD. Control refers to ryegrass that did not receive any biosolids application.
there was no statistically significant difference in metal uptake by the ryegrass between treatments.

In general, the element concentration of the ryegrass followed the temporal fluctuation of the study control. On the basis of these findings, biosolids application at the legal rate in Ireland (which is currently based on the metal content of biosolids and the available P content of the soil) did not increase the metal content of the plant biomass. Moreover, the method used to generate the biosolids (AD, LS, TD) did not result in different metal bioavailability and subsequent uptake and bioaccumulation rates in ryegrass. While the maximum average concentrations of As, Cr, Fe and V were at, or above, the ranges of trace elements in Irish pastures, no measured metals were within or above the concentration at which phytotoxicity of ryegrass occurs. The metals were also below the maximum levels specified for animal feeds (EU, 2002).

There was a statistically significant downwards trend with time from around 360 hours onwards in the concentration of all metals in all treatments, including the study control. Concentration of some metals such as As, Nb and Sb, reduced from low initial concentrations (average initial concentrations for each were 0.2, 0.07 and 0.01 mg kg$^{-1}$, respectively) to below the LOD of the instrumental technique over the duration of the study, indicating the impact of metal dilution by shoot growth. There was a residual metal concentration in the ryegrass (prior to biosolids application) at the start of the growing season, but this became more diluted as the growing season progressed. When the metal uptake in the biosolid-amended micro-plots were taken away from the metal uptake in the control micro-plots at each of the time intervals sampled, the metal content of the shoots reduced from the time of the first sampling (at 48 hours) through all subsequent sampling times.

5.5 Conclusions

This study found that, in general, there was no statistically significant difference in the shoot metal concentration of ryegrass cultivated on biosolid-amended plots and unamended plots when the biosolids were spread at the maximum permissible rate in Ireland. Over a period of 18 weeks after land application of biosolids, a downwards trend in the concentration of metals in the shoots of ryegrass was observed, which was attributed to a dilution effect as the ryegrass grew. On the basis of the parameters measured in this study, it would appear that the legislation governing livestock exclusion rates from land after biosolids application are overly strict. However, a short period of withdrawal (e.g. 3 weeks) seems reasonable to reduce the risk of biosolids ingestion by the animals; this is also the case with cattle slurries. Any further restrictions may be overly strict for a single application to land at compliant application rates.
6 Hazard Identification

6.1 Overview

The aim of this chapter is to identify potential chemical and biological hazards present in biosolids that are capable of causing adverse human health effects through surface water. A suite of 16 contaminants identified in the literature review were further analysed in a risk-ranking model to include a health-based risk endpoint. A probabilistic model was constructed in Microsoft Excel (incorporating @Risk 6.0) to estimate human exposure to organic contaminants that are contained within biosolids destined for land application. Probability density distributions were used to take account of uncertainty and variability in the model inputs. A database of inputs was collated from international and European peer-reviewed journals, online chemical databases (Chemfinder, CAS and, ECHA, ESIS), regulatory agency data and internet search engines.

6.2 Contaminants of Concern

The contaminants analysed in this study comprise a group of organic contaminants belonging to various categories and were chosen based on the following risk factors: persistence, bioaccumulation and toxicity (PBT). The selected contaminants include: POPs [polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxin furans (PCDD/Fs) and polybrominated diphenyl ethers (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 bisphenol A (BPA). The aforementioned contaminants show that the compounds that are expected to remain in the soil at the origin of application [Log Koc (soil adsorption coefficient) >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). Recent toxicological reports have shown that TCC has the potential to disrupt excitation–contraction coupling in skeletal and cardiac muscles in humans (Gautam et al., 2014). Emerging persistent organic pollutants, such as PFOS and PFOA, have frequently been detected in drinking water and have become a significant concern to human health at a concentration of 40 ng L⁻¹ or higher (Xiao et al., 2013). 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 accumulate in the environmental compartments that are considered high in organic content, such as sewage sludge (Soares et al., 2008).

6.3 Multimedia Risk Assessment Model Tools

Environmental exposure modelling has been developed in an effort to quantify human exposure to chemicals via contact with the surrounding natural environment and to evaluate the safe use of biosolids (Morais et al., 2013). Biosolids risk assessment has mainly focused on risks from microbials and infectious diseases in biosolids (Cummins and Adkin, 2007; Gottschall et al., 2013). However, with an increasing number of reports in the literature regarding identification of contaminants in biosolids, risk assessment models have been devised to assess the chemical fate from sewage treatment to biosolids application. The identification of emerging contaminants in biosolids has provoked academics and policymakers alike to evaluate the risks associated with land spreading of biosolids and contaminant exposure by humans through the food chain. This has spawned the development of environmental risk assessment models.

The evaluation of the removal efficiency and emission of a chemical from a WWTP requires an effective model. SimpleTreat 3.1 was developed for use under the Dutch Chemical Substances Act as a spreadsheet model for use as a tool for undertaking chemical risk assessment. SimpleTreat 3.1 is now a recommended model in the EU for environmental risk assessment. The SimpleTreat computational model is widely used to quickly calculate the relevant fate of a substance and
the exposure concentrations in the effluent, sludge and air directly surrounding the plant (van Egmond et al., 2013).

The European Union System for the Evaluation of Substances (EUSES) is a decision support instrument that enables government authorities, research institutes and chemical companies to carry out rapid and efficient assessments of the general risks posed by chemical substances to people and the environment (EC, 2016). As an assessment tool to calculate the predicted environmental concentration (PEC), EUSES has been applied as the standard risk assessment model in the EU for several years. The system is based on the EU Technical Guidance Document on Risk Assessment for new notified substances, existing substances and biocides. It represents a level III-type multimedia fate model that works with numerous QSARs, regression and default parameters in order to make predictions of steady state exposure concentrations based on a minimum set of input parameters (Wind, 2004).

The Risk Assessment Identification and Ranking (RAIDAR) model is a screening level risk assessment model that brings together information on chemical partitioning, reactivity, environmental fate and transport, bioaccumulation, exposure, critical objective or effect levels, and emission rates in a coherent system for assessing risk (CCEMC, 2007). The RAIDAR and EUSES models are conceptually similar in many respects: however, there are differences in the treatment of key processes. For example, the food web bioaccumulation models in RAIDAR allow for biotransformation rate estimates to be included, whereas the default food web bioaccumulation models in EUSES do not allow for this (Arnot et al., 2010).

Quantitative structure–activity relationship models are mathematical models that approximate the, often complex, link between chemical properties and biological activities of a compound (de Tilleghem and Govaerts, 2007). QSAR models enable prediction of physical, chemical and biological properties of non-assessed compounds by comparing structurally and/or quantitatively similar assessed compounds based on the structure and composition of the molecule (Sanderson et al., 2003). With regard to toxicity, QSARs have been successful in predicting well defined endpoints with a similar mode of action, e.g. baseline toxicity (narcosis) (Öberg and Iqbal, 2012).

The Pharmaceutical Assessment and Transport Evaluation (PhATE) model was developed by the Pharmaceutical Research and Manufacturers of America. This model was designed to offer a PEC of pharmaceuticals discharged into surface waters through WWTP (Anderson et al., 2012). Implementation of PhATE provides the advantage of evaluating the potential effect on human health associated with an active pharmaceutical ingredient (API) concentration that is below detection limits in surface waters. Predicted environmental concentrations generated by PhATE are based on average per capita human use of an API in the US. This assumption would cause PhATE to underestimate exposure in areas where per capita use is higher than the national average (Schwab et al., 2005).

6.4 Probabilistic Model

A probabilistic model was constructed to estimate and rank human exposure to organic contaminants that are contained within biosolids destined for land application. The model includes four major compartments: concentration in soil (PEC$_{\text{soil}}$), surface runoff (PEC$_{\text{runoff}}$), groundwater (PEC$_{\text{groundwater}}$) and final human risk [consisting of the level of chemical intake or human exposure (HE) and the chemical toxicity ratio (RR)]. The risk of organic contaminants leaching into surface water, groundwater or lakes from land spreading of biosolids can be estimated by the PEC$_{\text{soil}}$ PEC$_{\text{runoff}}$ and PEC$_{\text{groundwater}}$ model adopted from Trevisan et al. (2009), which was designed for the PECs of pesticides in ground and surface water. The model has been modified for use in Irish conditions. Toxicological indicator LC$_{50}$ (median lethal concentration) was used to assess the effect of the contaminants on human health. The 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 it was run for 10,000 iterations.

6.5 Results and Discussion

The results of the PEC$_{\text{soil}}$ indicate that from the contaminants analysed, the contaminants NP, NP1EO and NP2EO ranked the highest with mean PEC$_{\text{soil}}$ values of 5.69 mg kg$^{-1}$, 1.72 mg kg$^{-1}$ and 1.44 mg kg$^{-1}$ (95th percentiles 13.69 mg kg$^{-1}$, 5.14 mg kg$^{-1}$ and 2.88 mg kg$^{-1}$, respectively). This was attributed to the initial high concentrations of NP, NP1EO and NP2EO in the biosolids.
(mean values 103.8, 83.3 and 25.7 mg kg⁻¹, respectively), with NP and NP1EO exceeding the critical levels of 50 mg kg⁻¹ DM as suggested by the EU Working Document on Sludge 3rd Draft (Ömeroğlu et al., 2015). NPs are also restricted under the 2003/53/EC Directive, which restricts the use and marketing of products and product formulations that contain more than 0.1% NP or NPE in Europe (EC, 2002). The results for PEC runoff revealed that the highest values obtained were from NP, NP1EO and NP2EO with mean values of $1.17 \times 10^{-2} \mu g L^{-1}$, $4.13 \times 10^{-3} \mu g L^{-1}$ and $3.36 \times 10^{-3} \mu g L^{-1}$ (95th percentiles $4.08 \times 10^{-2} \mu g L^{-1}$ and $9.99 \times 10^{-3} \mu g L^{-1}$, respectively). NP, NP1EO and NP2EO ranked highest for PEC groundwater with mean values of $2.22 \times 10^{-1} \mu g L^{-1}$, $1.84 \times 10^{-2} \mu g L^{-1}$ and $9.11 \times 10^{-2} \mu g L^{-1}$ (95th percentiles $1.27 \mu g L^{-1}$, $5.5 \times 10^{-1} \mu g L^{-1}$ and $3.8 \times 10^{-1} \mu g L^{-1}$, respectively). The contaminants that follow NP, NP1EO and NP2EO in the risk ranking are the microbials, TCC and TCS. These microbials have only recently been regulated by the EU. Recent toxicological reports have shown that TCC has the potential to disrupt excitation–contraction coupling in skeletal and cardiac muscles in humans (Gautam et al., 2014). TCC has been linked to endocrine disruption in skeletal and cardiac muscles in humans (Gautam et al., 2014). TCC has been linked to endocrine disruption in skeletal and cardiac muscles in humans (Gautam et al., 2014). TCC has been linked to endocrine disruption in skeletal and cardiac muscles in humans (Gautam et al., 2014). TCC has been linked to endocrine disruption in skeletal and cardiac muscles in humans (Gautam et al., 2014). TCC has been linked to endocrine disruption in skeletal and cardiac muscles in humans (Gautam et al., 2014).

The results of this study suggest that the spreading of biosolids on agricultural land can result in moderate concentrations in the environment. Although the predicted concentrations in runoff and groundwater are low, it is imperative to note that these contaminants are continuously released into the environment. The top ranking contaminants for PEC runoff (NP, NP1EO and NP2EO) are below the Water Framework Directive (WFD) threshold value (0.3 µg L⁻¹) for annual average and the maximum allowable concentrations (2 µg L⁻¹) for surface waters (Lepom et al., 2009).

Human exposure through consumption of drinking water (combined with body weight and PEC runoff) showed that the contaminants that ranked the highest for adult and child consumption were NP, NP1EO and NP2EO, with mean values of $8.5 \times 10^{-5}$, $3.0 \times 10^{-5}$ and $2.8 \times 10^{-5} \mu g kg^{-1} bw d^{-1}$, respectively, for adult consumption, and $9.7 \times 10^{-5}$, $3.4 \times 10^{-5}$ and $2.2 \times 10^{-5} \mu g kg^{-1} bw d^{-1}$, respectively, for child consumption. HE and PEC groundwater showed that NP, NP1EO, and NP2EO ranked the highest

<table>
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<th>Contaminant</th>
<th>(RR) PEC runoff</th>
<th>Rank</th>
<th>(RR) PEC groundwater</th>
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</table>
for adult and child consumption, with mean values of $1.5 \times 10^{-3}$, $1.4 \times 10^{-3}$ and $7.2 \times 10^{-4}$ µg kg$^{-1}$ bw d$^{-1}$, respectively, for adult consumption, and $2.2 \times 10^{-3}$, $1.6 \times 10^{-3}$ and $5.1 \times 10^{-4}$ µg kg$^{-1}$ bw d$^{-1}$, respectively, for child consumption. The daily intake values of NP are much lower than the tolerable daily intake value of $5$ µg kg$^{-1}$ bw d$^{-1}$ of NP proposed by the Danish Institute of Safety and Toxicity (Ademollo et al., 2008). No European tolerable daily intake value has yet been established.

According to the LC$_{50}$ risk-ranking results for human exposure (LC$_{50}$ RR; PEC$_{\text{runoff}}$), the top ranked contaminants considered to be a risk to human health include NP, NP1EO and NP2EO, with mean LC$_{50}$ RR values of $1.10 \times 10^{-4}$, $3.94 \times 10^{-5}$ and $1.35 \times 10^{-5}$, respectively. Similarly, the results of the PEC$_{\text{groundwater}}$ also included NP, NP1EO and NP2EO. Mean RR values for PEC$_{\text{groundwater}}$ were $2.40 \times 10^{-3}$, $1.64 \times 10^{-3}$ and $3.66 \times 10^{-4}$ for NP1EO, NP and triclocarban, respectively (Table 6.1).

### 6.6 Sensitivity Analysis

A sensitivity analysis based on rank order correlation coefficient was conducted for NP as this contaminant ranked the highest across all of the environmental compartments. Results revealed that the Koc and soil organic carbon (SOC) were the most important parameters (correlation values of $-0.89$ and $-0.30$, respectively) that affected the variance in model predictions (Figure 6.1). This highlights the importance of the soil and site conditions [SOC, DT$_{50}$-soil (degradation time for 50% of a compound)] that influence the runoff and leaching of the contaminants through the soil matrix and the importance of contaminant properties (Koc) in influencing risk estimates. Furthermore, the sensitivity analysis results also showed that precipitation amount and water table level are other parameters of importance in the human health risk model.

### 6.7 Conclusion

A probabilistic model was developed to rank “classic” and emerging contaminants according to PEC$_{\text{soil}}$, PEC$_{\text{runoff}}$, PEC$_{\text{groundwater}}$ and resulting human health risk (RR). The highest rank obtained for PEC$_{\text{soil}}$, PEC$_{\text{runoff}}$ and PEC$_{\text{groundwater}}$ was the surfactant NP. To compare toxicity endpoints, the LC$_{50}$ combined with PEC$_{\text{runoff}}$ and PEC$_{\text{groundwater}}$ revealed that NP and NP1EO ranked the highest. A sensitivity analysis revealed that Koc and SOC were the most important parameters that affected model variance. This indicates that the type of soil that biosolids are spread on and its chemical properties are critical in controlling human health risk. Although the NPs ranked highest in this study, it is important to note that these contaminants have either been restricted or banned in Europe since 2005; therefore, these contaminants will reduce in risk. However, they still persist in the environment. The contaminants that ranked lower than the NPs, such as TCC and TCS, can be considered of greater interest as these contaminants are emerging

Figure 6.1. Sensitivity analysis for input parameters and the contaminant NP.
and have only recently been restricted within the EU, despite ongoing toxicological reports. The model developed in this study is of importance for risk managers as it provides a ranking of potential chemical hazards resulting from the spreading of biosolids on agricultural land, while it also highlights some emerging contaminants that will require vigilance in the future.
7 Exposure Assessment and Characterisation for Metals

7.1 Overview
An exposure assessment model was developed for metals. The model considered exposure to metals through surface water that was abstracted for drinking; it takes account of surface runoff amount, dilution and water treatment effects. Data from the field trials (Chapter 3) were used to model the initial level of metals in surface runoff.

7.2 Metal Accumulation in Soil
Long-term application of biosolids to agricultural land has led to concerns regarding the potential accumulation of metals in soil, the subsequent runoff into surface waters and the potential risk to human health through drinking water consumption. Previous studies have shown that overland transport from fields amended with biosolids can impact the quality of surface waters through runoff of heavy metals (Topp et al., 2008). The long-term use of biosolids can cause heavy metal accumulation in the soil (Elkhatib and Moharem, 2015). Furthermore, accumulation of these metals in the soils can have toxic effects on microorganisms and plants, and ultimately also on animals and human health via the food chain (Wang et al., 2005). Severe effects include reduced growth and development, cancer, organ damage, nervous system damage and, in extreme cases, death (Akpor and Muchie, 2010).

7.3 Quantitative Drinking Water Treatment Model
A quantitative drinking water treatment model that is capable of predicting probable human exposure and resulting risk from six metals (Cu, Cd, Cr, Pb, Ni and Zn) present in treated drinking water was developed. The model was created in Microsoft Excel 2010 with the add-on package @Risk. Data from peer-reviewed scientific literature was incorporated at various steps of the drinking water treatment (i.e. coagulation, flocculation, sedimentation and disinfection). Distributions were used to account for uncertainty in the data. The mean and standard deviation were calculated for all surface runoff results and a normal distribution was assigned to account for uncertainty in the data. Most drinking water in Ireland is sourced from surface waters. When there is a lack of data, a standard dilution factor of 10 was used, as proposed by the EU Technical Guidance Document on Risk Assessment Part II (EC, 2003), and applied to the data to take account of dilution effects in the predicted environmental concentrations.

7.4 Water Treatment Effects
Three stages of drinking water treatment were used based on the Irish Environmental Protection Agency’s (EPA) best practice guidelines for drinking water treatment manuals (EPA, 1995, 2002, 2011). The first stage, primary treatment, considers the screening, storage, pre-conditioning and pre-chlorination of the water. Primary treatment is typically used to screen out large objects such as sticks, stones and other debris that may restrict flow. In this instance the effect of primary treatment was deemed negligible in terms of contaminant removal.

As a worst case scenario, the model assumes a 90% probability of coagulation and flocculation operating at an optimum stable run (Copt) and a 5% probability for both sub-optimal (CS-opt) and failure (C-fail). When operating optimally, the model assumes a removal rate (metal specific). When operating sub-optimally, the model assumes a removal of 50% of the optimal removal rate and zero removal during failure events.

The filtration process in a conventional drinking water treatment plant consists of slow or rapid sand filtration. The filter run time is not only an indicator of the effectiveness of prior treatment (i.e. the ability of the coagulation and sedimentation steps to remove suspended solids), but it also plays a role in the effectiveness of the filter itself (WHO, 2004). In keeping with the EPA's filtration manual guidelines, the filtration process of rapid gravity filtration was considered in the model. As a worst case scenario the model assumes a 90% probability of filtration operating at an optimum stable run (Fopt) and a 10% probability for sub-optimal run (Fsub). When operating optimally, the model assumes a removal rate
that is metal specific. When operating sub-optimally, the model assumes a removal of 50% of the optimal removal rate.

Worldwide, chlorine is the most commonly used disinfection in drinking water treatment, although other alternatives are being increasingly introduced such as ozonation, UV irradiation, ultrasonic vibration, ultra-filtration, silver, bromide and iodine, membrane filtration and granular activated carbon. Chlorination does not remove metals (O’Connor and O’Connor, 2001) and therefore no removal distribution was assigned.

7.5 Human Exposure

The water consumption in Ireland was modelled using a log-normal distribution with a mean daily consumption of 0.564 L and a standard deviation of 0.617 L according to a survey on adult nutrition conducted by the Irish Universities Nutrition Alliance (IUNA), which was based on 1274 consumers (IUNA, 2011). A normal distribution with a mean value of 78 kg and standard deviation of 16.5 kg was used to model the variation in body weight for adults. A normal distribution with a mean value of 33 kg and standard deviation of 11.3 kg was used to model variation in body weight for children (IUNA, 2005). To evaluate the human health risk exposure, the lifetime average daily dose (LADD) (mg kg⁻¹) and the hazard quotient (HQ) were used as toxicity endpoints in the model and were metal specific. A Monte Carlo simulation technique was applied to sample from the input distributions to create an output distribution for metal concentration in drinking water, LADD and the HQ.

7.6 Results and Discussion

The environmental fate of selected metals (Cd, Cr, Cu, Ni, Pb, Zn) was modelled from biosolids application at three different time periods, 24 hours (RS1), 48 hours (RS2) and 360 hours (RS3), to drinking water treatment and subsequent human exposure. The model resulted in several output distributions post-drinking water treatments (Pₓₛₜ), LADDs and HQs that can be used to compare the concentration of metals that were detected in surface runoff and their potential risk to human health. The model predicted that surface runoff arising from the land spreading of LS biosolids produced the highest concentrations of Cu and Zn in drinking water. The modelled mean Cu concentration in drinking water (Figure 7.1) was highest when the surface runoff concentrations from the LS biosolids at each rainfall simulation time (24, 48 and 360 hours) were used as input into the model (mean concentration values of 2.45, 1.78 and 1.2 µg L⁻¹, respectively). This was followed by Zn, which had mean concentrations of 1.25, 5.14 × 10⁻¹ and 6.16 × 10⁻¹ µg L⁻¹ for each rainfall event. All metal concentrations were below the metal threshold values of the EU and the World Health Organization (WHO, 2011a; S.I. No. 122 of 2014).

The results for the exposure assessment (LADD) show that child exposure concentrations were highest for
the metal Cu and for all three time frames RS1, RS2 and RS3 (mean values $2.07\times10^{-2}$, $2.07\times10^{-2}$ and $1.18\times10^{-2}\,\mu g\,kg^{-1}\,bw\,d^{-1}$) and LS treatment. This was followed by adult Cu exposure concentrations (mean value $1.80\times10^{-2}$, $1.31\times10^{-2}$ and $9.21\times10^{-3}\,\mu g\,kg^{-1}\,bw\,d^{-1}$, for all three time frames RS1, RS2 and RS3, respectively).

All LADD values were below the recommended provisional maximum tolerable daily intake (PMTDI) values for Cd, Cu, Ni and Zn ($7, 500, 5$ and $100\,\mu g\,kg^{-1}\,bw\,d^{-1}$, respectively) as proposed by the Joint FAO/WHO Expert Committee on Food Additives (JECFA). A PMTDI has not been established for Cr and the PMTDI for Pb was withdrawn in 2010 as it could no longer be considered health protective (WHO, 2011a).

The HQ is calculated by dividing an estimate of an exposure concentration by the threshold toxicity reference value such as a reference dose. A HQ value of $<0.01$ indicates no existing risk. The risk is low for HQ $0.1-1.0$ and moderate for HQ $1.1-10$; a HQ value of $>10$ indicates a high risk (Lemly, 1996). The results for the HQ showed that of all the scenarios considered, the metal Cu and the biosolids treatment LS were the highest for children for all three time frames (RS1, RS2 and RS3) with mean child HQ values of $5.59\times10^{-4}$, $4.09\times10^{-4}$ and $3.18\times10^{-4}$, respectively, followed by adult Cu concentrations (mean adult HQ values of $4.87\times10^{-4}$, $3.54\times10^{-4}$ and $2.49\times10^{-4}$, respectively). However, these were still below the threshold value of risk (HQ$<0.01$; no existing risk) (Figure 7.2).

### 7.7 Sensitivity Analysis

A sensitivity analysis based on rank order correlation was carried out to assess the ways in which the model's predictions are dependent on variability and uncertainty in the model input parameters. A sensitivity analysis was conducted for Cu corresponding with the LS biosolids treatment, as this metal and treatment had the highest concentration in both risk endpoints (LADD and HQ). Results revealed that tap water intake and filtration reduction were the most important parameters (correlation coefficient values of 0.67 and −0.54, respectively) that affected the variance in model predictions. This highlights, of all the inputs assessed, the efficiency of the filtration system as one of the most important parameters influencing the final risk assessment. The effectiveness of the filtration is reliant on the efficiency of the coagulation/flocculation and sedimentation (correlation coefficient −0.35) stage of the process, as this stage can help to remove a majority of the metals in the water. Body weight (correlation coefficient −0.15) was also an important parameter, as when the body weight is reduced, the risk increases. The initial concentration in runoff was also an important parameter (correlation coefficient 0.12), which highlighted the importance of having the initial concentration of metals in sludge as low as possible (Figure 7.3).

### 7.8 Conclusions

Six metals were assessed to estimate the total concentration in surface runoff following land application of four...
types of biosolids with a soil only control, following drink-
ing water treatment and subsequent HE. A quantitative
model was developed in Excel, using the @Risk add-on
software, to follow the fate of all six metals. The model
incorporated dilution of the runoff concentrations in a
stream and it was assumed that water was extracted
for drinking water treatment. Drinking water treatment
including the primary, secondary and tertiary stages.
The outputs of the model included metal concentration
in drinking water post-treatment (P_{dw}) and exposure
endpoints, LADD and the HQ. Results showed that the
heavy metal Cu had the greatest concentration in drink-
ing water post-secondary and tertiary treatments. The
results for HE (LADD) showed that Cu had the greatest
concentration, especially for child consumption; how-
ever, it was still below the World Health Organization
(WHO) tolerable daily dose. The results for the HQ
showed that of all the scenarios considered, the metal
Cu was the highest risk, particularly for children, and
those corresponding to the lime treated biosolids.
However, these were still below the threshold value of
risk (HQ<0.01: no existing risk). Under the conditions
monitored, metal concentrations in the four biosolids
were not considered a risk to human health. Although
metal accumulation in soil was not considered in this
study, it is worth considering for future research.
8 Exposure Assessment and Characterisation for E. coli

8.1 Faecal Coliforms and the Sewage Sludge Directive (86/278/EC)

Faecal coliforms are used as indicator organisms; high levels of these bacteria indicate the potential presence of pathogens that cause waterborne diseases (Selvaratnam and Kunberger, 2004). Faecal coliforms include E. coli, among other coliforms. In the EU, sewage sludge production is regulated by the Sewage Sludge Directive 86/287/EC. It does not specify limits for pathogen populations but specifies general land use and harvesting and grazing limits to provide protection against the risk of infection (Sobrados-Bernardos and Smith, 2012). A revision of the Sewage Sludge Directive (Working Document 3rd Draft) states that “the use of microbial indicators to evaluate the hygienisation of treated sludge is based on fulfilling the limits of E. coli to achieve a 99.9% reduction and to less than 1 × 10³ cfu/g dry weight, produce a sludge containing < 3 × 10³ spores of Clostridium perfringens/g (dry weight) and absence of Salmonella. spp in 50 g (wet weight)” (EEC, 2000). Furthermore, the Working Document also states that sludge produced by conventional treatment shall achieve at least a 2 log₁₀ reduction of E. coli (Mininni et al., 2014). In their Guidelines for Drinking Water Quality, the WHO have developed a risk classification to prioritise interventions, as higher levels of indicator organisms are generally indicative of greater levels of faecal contamination. The risk classification is based on the number of indicator organisms in a 100-mL sample, which includes < 1 “very low risk”, 1–10 “low risk”, 10–100 “medium risk”, > 100 “high risk” or “very high risk” (WHO, 2011b).

8.2 Quantitative Drinking Water Treatment Model (E. coli)

A quantitative drinking water treatment model that is capable of predicting probable human exposure and resulting risk from E. coli present in drinking water was developed. The model was created in Microsoft Excel 2010 with the add-on package @Risk. Data from peer-reviewed scientific literature were incorporated at various steps of the drinking water treatment (i.e. coagulation, flocculation, sedimentation and disinfection). Distributions were used to account for uncertainty in the data. The data for biosolids runoff post-application were generated by project partners (NUI Galway, Teagasc). The mean and standard deviation were calculated for all surface runoff levels and a log-normal distribution was assigned to account for uncertainty in the data. Most drinking water in Ireland is sourced from surface waters and therefore a standard dilution factor of 10 (EC, 2003) was applied to the data to take dilution effects into account. The model simulates the surface water die-off rate using Chick’s law first-order decay equation Nₜ = N₀ e⁻kt, where Nₜ is the number of organisms at time (t), N₀ is the initial number of organisms, K is the first-order inactivation constant (day⁻¹) and t is the time in the stream (day⁻¹). To account for the uncertainty in the data, a uniform distribution using K values (minimum 0.7, maximum 1.5) (Schueler, 2000) was assigned. “K” values in this range mean that about 90% of the bacteria present will disappear from the water within 2 to 5 days. Therefore, it was assumed that water was abstracted from the stream to a nearby water treatment plant between 0 and 5 days. To account for uncertainty, the time in stream “t” was fitted with a uniform distribution (minimum 0, maximum 5 days).

8.3 Water Treatment Effects

Three stages of drinking water treatment were used based on the EPA’s best practice guidelines for drinking water treatment manual (EPA, 2011). The first stage (primary treatment) considers the screening, storage, pre-conditioning and pre-chlorination of the water. The second stage (secondary treatment) considers coagulation, flocculation and sedimentation. Flocculation, sedimentation and removal of E. coli were reported by Pritchard et al. (2010); the authors compared the efficacy of aluminium sulfate to more natural coagulants and reported E. coli reductions of 89% using 30–50 mgL⁻¹ of aluminium sulfate. Bulson et al. (1984) reported removal rates of E. coli of 99.99% following a dose of 15mgL⁻¹ of aluminium sulfate. Rapid mixing for a few seconds is important once a coagulant is added to ensure uniform dispersion. Subsequent and prolonged mixing aids in the formation of flocs. The flocs settle by gravity and can be removed via filtration. Therefore, a uniform distribution was assigned to the
percentage reduction in bacteria during a stable optimal run (minimum 0.89, maximum 0.99 reduction). As a worst case scenario, the model assumes a 90% probability of coagulation/flocculation and sedimentation at an optimum stable run (Copt), a 5% probability for sub optimal (CS-opt) and a 5% probability for failure (Cfail). The model assumes a bacterial reduction at a rate of 50% of the optimal removal rate during a suboptimal run and a rate of 0% reduction during a failed run.

Rapid sand filtration provides fast and efficient removal of relatively large suspended particles. It is a relatively erudite process, which usually requires power-operated pumps, regular backwashing or cleaning and flow control of the filter outlet. In keeping with the EPA’s filtration manual guidelines, the process of rapid gravity filtration was considered in the model. As a worst case scenario, the model assumes a 90% probability of filtration operating at an optimum stable run (Fopt) and a 10% probability for sub-optimal run (Fsub). When operating optimally, the model assumes a uniform log removal rate (minimum 74%, maximum 99%). When operating sub-optimally, the model assumes a removal of 50% of the optimal removal rate.

The third stage (tertiary) involves the disinfection of the effluent. Chlorination is the most popular tertiary treatment in Ireland; it has been found to remove between 97% and 99% of E. coli (O’Connor and O’Connor, 2001). To account for uncertainty in the data, a uniform distribution (minimum 0.97, maximum 0.99) was assigned to the disinfection process of the model.

8.4 Human Exposure

Two dose response models were considered for E. coli exposure [healthy population and immunocompromised (IC) populations]. The probability for illness was calculated using a negative exponential model for E. coli O157 as proposed by Gale (2005): $P_r = 1 - e^{-r}$, where $r$ is the risk of illness from ingestion of a single bacterial cell. To account for susceptibility in IC populations an “$r$” value of 0.01 was used, while for healthy populations an “$r$” value of 0.0000005 was used. As a worst case scenario, the illness model was parameterised with the assumption that the virulence of the pathogen is similar to E. coli O157:H7. The E. coli O157:H7 strain is a particular serotype of the group referred to as verocytotoxigenic E. coli (VTEC). VTEC produces verotoxins or shiga-like toxins that are closely related to the toxin produced by Shigella dysenteriae (Cassin et al., 1998).

The USEPA have proposed a drinking water limit of $10^{-4}$ per person per year for Shigella (Grant et al., 2012). The entire model was constructed in Microsoft Excel 2010 with the @Risk add-on using Monte Carlo simulation techniques and it was run for 10,000 iterations.

8.5 Results and Discussion

The environmental fate of E. coli was modelled from biosolids application at three different time periods, 24 hours (RS1), 48 hours (RS2) and 360 hours (RS3), to drinking water treatment and subsequent drinking water consumption. The model resulted in several output distributions that can be used to compare the coliforms that were detected in surface runoff and their potential risk to human health. Outputs from the model include viable E. coli consumption and the probability of illness (both for healthy and IC individuals). The results for viable E. coli consumed show that the biosolids treatment ADUK was highest for RS1 and RS2, with exposure mean values of $5.20 \times 10^{-4}$ and $2.34 \times 10^{-1}$ MPN d$^{-1}$, respectively (Figure 8.1). The WHO states that any treated water should have no E. coli detection per 100 mL. A consequence of variable susceptibility to pathogens is that exposure to drinking water of a particular quality may lead to health problems in different populations (WHO, 2011b), particularly in the very young and IC individuals.

The results for the probability of illness for healthy and IC populations show that, among IC populations, the biosolid treatment ADUK (RS1 and RS2) had the greatest probability of illness/day with mean probability values of $3.68 \times 10^{-3}$ and $2.1 \times 10^{-3}$, respectively (Figure 8.2). The probability of illness over a period of a year was also analysed and this showed that the biosolids treatment ADUK combined with IC populations had the greatest probability of illness in time frames RS1 and RS2, with mean values of $2.0 \times 10^{-1}$ and $1.7 \times 10^{-1}$, respectively.

8.6 Sensitivity Analysis

A sensitivity analysis based on rank order correlation was carried out to assess the ways in which the model’s predictions are dependent on variability and uncertainty in the model input parameters. A sensitivity analysis was conducted for ADUK biosolids treatment as this treatment had the highest concentration in both dose-response models. Results revealed that from all the inputs considered, the time in stream, concentration in
surface runoff, tap water intake and *E. coli* inactivation were the most important parameters that affected the variance in model predictions (correlation coefficient values of –0.55, 0.42, 0.30 and –0.20, respectively) (Figure 8.3).

Ideally, water intended for human consumption should be pathogen free; however, this is an unachievable goal in practice. The risk of illness for IC individuals was greater than the acceptable level of risk of illness per year ($1 \times 10^{-4}$), as set by the USEPA. It is noted in such cases that vulnerable groups may take specific precautions with respect to the intake of food and water, which, as a result, controls the vulnerable nature of this sub-population. The biosolids treatment ADUK was persistently high throughout the model [VCC and probability of illness/day (RS1 and RS2)]. Anaerobic digestion is the most popular method of stabilisation of sewage waste in Europe. A key factor in pathogen removal is the sludge retention time, as was reported by Chen et al. (2012). It is unknown how long the ADUK sludge was digested for, but it may explain the consistency of *E. coli* throughout the model. The disinfection process used (i.e. chlorination) is in line with the recommended disinfection treatment commonly used in Ireland. The efficacy of chlorine in relation to inactivation of bacteria such as *E. coli* is high (between 97% and 99%). Other factors, such as die-off in stream, also reduced the colony counts by a factor of 2. Under the conditions monitored, *E. coli* concentrations in the four biosolids were not considered a risk to healthy individuals but, as always, caution is required with IC individuals.
A quantitative risk assessment model was developed to assess the probability of human exposure to *E. coli* following land application of various biosolids treatments to agricultural land. The model inputs included application to agricultural land, dilution in stream, die-off in stream, drinking water treatment processes (coagulation, flocculation, sedimentation and disinfection) and a dose–response model to calculate human health risk. Outputs from the model include viable *E. coli* consumed and probability of illness for healthy and IC populations.

Results from the model looking at viable *E. coli* consumed show that the biosolids treatment ADUK had the highest concentration of *E. coli* for RS1 and RS2 time frames, while biosolids treatment ADIRE showed the greatest concentration of *E. coli* for RS3 time frame.

Results for HE via a pessimistic dose–response model (i.e. *E. coli* O157:H7) for the probability of illness/day showed that the biosolids treatment ADUK had the greatest probability of illness risk for all time frames except RS3. ADUK was also greater for the probability of illness/year (RS1 and RS2). The risk of illness for IC individuals was greater than the acceptable level of risk of illness/year ($1 \times 10^{-4}$), as set by the USEPA, which highlights the vulnerable nature of this sub-population.

A sensitivity analysis revealed that the inputs that influenced the model’s variance were: the time in stream, concentration in surface runoff, tap water intake and *E. coli* inactivation. The results of the model showed that the concentrations of *E. coli* in the final effluent could be a hazard for IC populations when assuming a worst case scenario.
9 Conclusions and Recommendations

9.1 Overview

The objective of this study was to (1) characterise the concentrations of metal, TCS and TCC in biosolids from a selection of wastewater treatment plants in Ireland; (2) determine the impact on surface runoff following land application of three types of biosolids (AD, LS and TD) and compare them to another commonly spread organic fertiliser, DCS; (3) measure the uptake of metals by ryegrass for a period of time after the application of biosolids; and (4) model and conduct a risk assessment of potential hazards of human health concern based on the experimental data. To achieve this, a simple, novel, field-scale micro-plot study was designed and conducted, which examined the possible impacts arising from the land application of these treatments on surface runoff water and soil properties. Sections 9.2 and 9.3 present the main conclusions and recommendations arising from this study.

9.2 Conclusions

9.2.1 Land application

1. The impact of biosolids spread at the maximum application rate on grassland had no adverse impact on surface water quality compared to DCS in terms of nutrients and metal losses in surface runoff. The concentrations of TCS and TCC were not compared to DCS as they were below the LOD of the instrumentation used in this analysis.

2. With the exception of Cu in runoff from LS biosolids plots, all runoff samples from the biosolid-amended field plots were below their respective surface water standards.

3. Surface runoff losses of the water quality parameters analysed from biosolid-amended plots were higher than the study control (soil only) plots, and followed a general trend of highest losses occurring during RS1 and reduced losses in the subsequent events.

4. With the exception of total coliforms and some metal parameters, the greatest losses were from the DCS-amended plots over the time studied (15 days). This means that with respect to the parameters studied, biosolids do not pose a greater risk in terms of runoff losses following land application over a 15-day period from the time of first application.

5. The concentrations of metal of the treated sludge from the WWTPs examined as part of this study were below the maximum allowable concentrations of metals for use in agriculture in the EU.

6. TCS and TCC, for which no regulatory standards for sewage sludge exist, were present in the treated sludge from the WWTPs examined. They were below the concentrations measured in other studies; the concentrations of these parameters may, however, vary throughout the year.

7. This study found that, in general, there was no statistically significant difference in the shoot metal concentration of ryegrass cultivated on biosolid-amended plots and those cultivated on unamended plots, when the biosolids were spread at the maximum permissible rate in Ireland.

9.2.2 Health issues

1. Under the conditions and for the parameters monitored, there was no measurable risk to human health. However, further testing in a larger field-scale experiment is needed to verify the findings of this study.

2. A risk-ranking model highlighted the importance of soil type (viz. SOC) and chemical properties (viz. Koc) in influencing the human health risk.

3. For both the metal and E. coli models, the initial concentration in the biosolids and filter reduction were the key parameters influencing the final risk estimates.

Under the conditions monitored, metal and E. coli concentrations in the four biosolids were not considered to pose a measurable risk to human health when spread on land, although caution is warranted when it comes to...
IC individuals. However, such individuals would follow strict food and water intake guidelines, thus reducing the risk of illness.

9.3 Recommendations

1. While current EU and international regulations govern certain priority metal pollutants and bio-essential elements, other emerging contaminants that are potentially harmful to human health are omitted from the regulations. This means that, potentially, a number of emerging contaminants are being applied to land without regulation. As metals are relatively easy to measure using the techniques detailed in this study, it is recommended that the regulations governing the values for metal concentrations in biosolids for recycling in agriculture are extended to cover more metals. The measurement of pharmaceuticals is more problematic as it is very costly to measure these numerous parameters. In the first instance, it is recommended to test biosolids for TCS and TCC, as these parameters are of the greatest concern internationally. WWTPs may also be upgraded or retrofitted to include treatment of these emerging contaminants, thereby negating the issue of their potential land application.

2. On the basis of the parameters measured in this study, it would appear that the legislation governing livestock exclusion rates from land after biosolids application are overly strict. However, a short period of withdrawal (e.g. 3 weeks) seems reasonable to reduce the risk of biosolids ingestion by the animals (as would be the case with cattle slurries). Any further restrictions may be overly strict for a single application to land at compliant application rates.

3. Currently, there is a knowledge gap concerning the effectiveness of LS in adhering to the pH and temperature requirements of the Codes of Good Practice. There is a need for research into the LS process and its effectiveness to minimise food safety concerns. This research should result in the introduction of mandatory standards governing LS methodologies.
References


CCEMC (Canadian Centre for Environmental Modelling and Chemistry), 2007. RAIDAR Model. Available online: http://www.trentu.ca/academic/aminss/envmodel/models/RAIDAR100.html


Magnusson, K. and Noren, F., 2014. Screening of microplastic particles in and downstream a wastewater treatment plant. IVL Swedish Environmental Research Institute, Stockholm.


USFDA (United States Food and Drug Authority), 2015. FDA taking closer look at “antibacterial” soap. Available online: http://www.fda.gov/ForConsumers/ConsumerUpdates/ucm378393.htm


# Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Definition</th>
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<tr>
<td>AD</td>
<td>Anaerobically digested</td>
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<tr>
<td>ADIRE</td>
<td>Anaerobically digested biosolids sourced in Ireland</td>
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<tr>
<td>ADUK</td>
<td>Anaerobically digested biosolids sourced in the United Kingdom</td>
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<td>AI</td>
<td>Aluminium</td>
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<td>ANOVA</td>
<td>Analysis of variance</td>
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<td>API</td>
<td>Active pharmaceutical ingredient</td>
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<td>Biological oxygen demand</td>
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<td>Bisphenol A</td>
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<td>Copper</td>
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<td>DCS</td>
<td>Dairy cattle slurry</td>
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<td>DM</td>
<td>Dry matter</td>
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<tr>
<td>DRP</td>
<td>Dissolved reactive phosphorus</td>
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<td>Dried solids</td>
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<tr>
<td>DUP</td>
<td>Dissolved unreactive phosphorus</td>
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<td>Faecal coliforms</td>
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<td>Fe</td>
<td>Iron</td>
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<td>FP7</td>
<td>Seventh Framework Programme</td>
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<td>FWMC</td>
<td>Flow-weighted mean concentration</td>
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<td>Hectare</td>
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<td>Human exposure</td>
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<td>Mercury</td>
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<td>Hazard quotient</td>
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<td>LC_{50}</td>
<td>Median lethal concentration</td>
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<td>Limits of quantification</td>
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<td>Nitrogen</td>
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<td>Nb</td>
<td>Niobium</td>
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<td>Nickel</td>
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<td>Nitrate-nitrogen</td>
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<td>P_{2}O_{5}</td>
<td>Phosphorus pentoxide</td>
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<tr>
<td>PBDEs</td>
<td>Polychlorinated dibenzyl ethers</td>
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<td>PBT</td>
<td>Persistence, bioaccumulation and toxicity</td>
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<td>Polychlorinated biphenyls</td>
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<td>Personal care products</td>
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<td>Provisional maximum tolerable daily intake</td>
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<td>RAIDAR</td>
<td>Risk Assessment Identification and Ranking (model)</td>
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<td>Description</td>
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<td>Triclocarban</td>
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<td>Total nitrogen</td>
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<td>Total phosphorus</td>
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<td>Time to runoff</td>
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<td>United Nations Environment Programme</td>
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<td>USEPA</td>
<td>United States Environmental Protection</td>
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<td>Water-extractable phosphorus</td>
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<td>X-ray fluorescence</td>
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<td>Zn</td>
<td>Zinc</td>
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AN GHÍNÍOMHRAECHT UAM CHAOMHÍNÚ COMHSHAOL

Tá an Ghníomhaireacht um Chaomhínú Comhshaol (GCC) freagrach as an gcomhshaol a chaomhún agus a héabhsú mar shochrann lauchmhór do mhuintir na hÉireann. Táimid tiomanta do dhhuine agus don chomhshaol a chosaint ó éifeachtai diobhálacha na radaithe agus an truaillithe.

Is féidir obair na Gníomhraechta a roinnt ina tri fhriomhréimese:

**Ríolú:** Déanaimid córais éifeachtachta rialaithe agus comhlionta comhshaol a chur i bhfeidhm chun tourthait maithte comhshaol a sholáthar agus chun diriú orthu stáid na gclú leis na córais sin.

**Eolas:** Soláthairmid sonrait, fáisnéis agus measúnú comhshaol atá ar ardaighdeán, spriochaithe agus tráthnú chun bonn eolaí a chur faoin gcintneoirceacht ar gach leibhéil.

**Tacaochta:** Bimid ag saothrú i gcomhar le grúpaí eile chun táiliú le comhshaol agus an tráthnú i bhfeidhm leis an gcomhshaol.

**Ár bhFheagrachtai****

Ceadúnú Déanaimid na gniomhaochtaí seo a leanas a rialú ionsach nach ndéanann siad dochar do sháithiú an phobail ná don chomhshaol:

- saoráid draimhiala (m.sh. láthairín, lúsan talún, leisceóirí, stáisiúin astúití draimhrialta); gniomhaochtaí tionsclaíochta ar slán mór (m.sh. déanntaíocht cóigísíochta, déántaíocht stróighne, stáisiúin chumhacht). D'fhéadfadh siad na gniomhaochtaí a dhéanamh ar leasúcháin a chur le thráthnú.
- an diantalmhailhaochta (m.sh. mórca, éanlaith); úsáid shraithanta agus scáilteadh rialaithe Orgánaí Gníomhbaolaithe (OGM).
- foinse radaiteacha i an t-áthasúnaíocht (m.sh. trealamh x-gha agus radaitheiripe, foinse tionsclaíochta); áiseanna móra stórála peitril.
- scardadh draimhuisce.
- gniomhaochtaí dumpála ar farrage.

Forfhleidhmíu Náisiúnta in leith Cúrsaí Comhshaol

- Clár náisiúnta inicí aici agus círeacairtaí a dhéanann gach bhaili an shaoráid a bhfuil céad uile don nGníomhaireacht acu.
- Mairesceachtaí a dhéanann ar fhreagrachtai cosanta comhshaol na n-údaráis áitiúil.
- Caighdeán na bhfuil domhain faoi th对接he, go mór, mar gheall ar shaothar agus an rialtaíocht.
- Cur i bhfeidhm rialachán agus dhábháis, le léiríochtaí sa radaíocht a bheith níos iomlán air féin.
- An dá a chuir orthu siúd a bhíonna dli an chomhshaol agus a dhéanann dochar don chomhshaol.

Bainistíocht Uisce

- Monatóireacht, Análisis agus Tuairiscíú ar an gComhshaol

- Monatóireacht a dhéanann ar cháiliocht an aer agus Treoir an AE maidir le hAer Gan don Eoraip (CAFÉ) a chur chun féidhme.
- Tuairiscíú neamhspleách le cabhrú le ñíos mótháiní ar dhal éifeacht a bhainistiú.

Ríolú Astaíochta na nGás Ceaptha Teasa in Éirinn

- Fardail agus reamh-mheastacháin chun teasa na n-údaráis a taitneamh.
- An Treoir maidir le Trádáil Astaíochta a chur chun féidhme i gcomhair breis agus 100 de na táirgí leis an gcoisialtaíocht an gcomhshaoil.

Caipíní Raideolaioch

- Monatóireacht a dhéanann ar leibhéil raideolaiochta, meascúnacht a dhéanamh ar nochtadh mhuintir na hÉireann do chomhshaol iomhamhacht.
- Cabhrú le rialaíocht an Írisce a tharla le ghlacadh éagdaíodh ag eascairt as taismí núicléacha.
- Monatóireacht a dhéanann ar forbartha i mBearn agus ar pholaitiúl a bhainte agus ar leasúcháin a chur le thráthnú.
- Sainseirbhísí cosanta ar an radaíocht a sholáthar, nó máoisíóidí a dhéanann don chomhshaol.

Measúnacht, Treoirí, Fíorpháirtíochtaí agus Treoirí

- Monatóireacht a dhéanamh ar gceartacht aithneach a chosc agus a bhainistiú.
- Monatóireacht a dhéanamh ar leasúcháin agus le teaghlaigh a bhainte.

Treoir, Saoirse, Oideachas, a dhéanann dochar don chomhshaol.

- Forbhreithint, Fáintíochta agus Oideachas.
- Treoir maidir le Trádáil Astaíochta a chur chun feidhme i gcomhair seirbhísí chumhachtachta.

Forghníomhaireacht Thoirseal Comhshaol

- Monatóireacht a dhéanamh ar gceartacht aithneach a chosc agus a bhainistiú.
- Monatóireacht a dhéanamh ar leasúcháin agus le teaghlaigh a bhainte.

Múscaill, Feasachta agus Astaíochta

- Forphrionnisiacht.
- Treoir maidir le Thrádáil Astaíochta a chur chun feidhme.

Bainistíochta agus Forphréaictíocht

- Treoir agus saoráidí a thugtar air féin.
- Treoir agus saoráidí a thugtar air féin.

**Ceadúnú**

Déanaimid na gniomhaochtaí seo a leanas a rialú ionas nach ndéanann siad dochar do sháithiú an phobail ná don chomhshaol:

- saoráid draimhiala (m.sh. láthairín, lúsan talún, leisceóirí, stáisiúin astúití draimhrialta);
- gniomhaochtaí tionsclaíochta ar slán mór (m.sh. déántaíocht cóigísíochta, déántaíocht stróighne, stáisiúin chumhacht);
- an diantalmhailhaochta (m.sh. mórca, éanlaith);
- úsáid shraithanta agus scáileadh rialaithe Orgánaí Gníomhbaolaithe (OGM);
- foinse radaiteacha i an t-áthasúnaíocht (m.sh. trealamh x-gha agus radaitheiripe, foinse tionsclaíochta);
- áiseanna móra stórála peitril;
- scardadh draimhuisce;
- gniomhaochtaí dumpála ar farrage.

Forfhleidhmíu Náisiúnta in leith Cúrsaí Comhshaol

- Clár náisiúnta inicí aici agus círeacairtaí a dhéanann gach bhaili an shaoráid a bhfuil céad uile don nGníomhaireacht acu.
- Mairesceachtaí a dhéanann ar fhreagrachtai cosanta comhshaol na n-údaráis áitiúil.
- Caighdeán na bhfuil domhain faoi th对接he, go mór, mar gheall ar shaothar agus an radaíocht.
- Cur i bhfeidhm rialachán agus dhábháis, le léiríochtaí sa radaíocht a bheith níos iomlán air féin.
- An dá a chuir orthu siúd a bhíonna dli an chomhshaol agus a dhéanann dochar don chomhshaol.
The aims of this study were to: (1) undertake a thorough literature review of the spreading of treated sewage sludge (biosolids) on land to include analysis of potential impacts on environmental and human health; (2) examine, under controlled conditions in the laboratory and field, the impact of the landspreading of biosolids (on grassland) on surface runoff/subsurface drainage/shallow groundwater of nutrients, solids, metals, pathogens and some specified emerging contaminants identified in the literature review, when spread based on N and P application rates; and (3) to model and conduct a risk assessment of potential hazards of human health concern.

Identifying Risks

Implementation of European Union Directives in recent decades concerning the collection, treatment and discharge of wastewater, as well as technological advances in the upgrading and development of wastewater treatment plants, has resulted in an increase in the number of households connected to sewers and an increase in the production of sewage sludge (the by-product of wastewater treatment plants). Recycling to land is currently considered the most economical and beneficial way for municipal sewage sludge management. However, despite the many potential benefits of recycling municipal sewage sludge to land, there are many risks, which include the presence of emerging contaminants in the sewage sludge that may enter the food chain, and the potential for surface runoff of contaminants into receiving waters. This project found that although the application of biosolids poses no greater threat to surface water quality than the land application of dairy cattle slurry, there is a possibility that many non-priority elements and emerging contaminants, for which no legislation currently exists, may be applied to land without regulation, and may accumulate in the soils and enter the food chain.

Informing Policy

Current legislation governing the land application of municipal sewage sludge to land considers certain priority pollutants and bio-essential elements. However, other emerging contaminants may be inadvertently applied to land. Regulations should be extended to cover non-priority elements, pharmaceuticals and personal care products (PPCPs). Non-priority elements are relatively inexpensive to measure, but PPCPs are prohibitively expensive as well as being continuously evolving. Wastewater treatment plants may be upgraded to include treatment of emerging contaminants, but the potential presence of known, as well as currently unknown parameters, raises concerns over the continued application of biosolids to land in Ireland.