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Nutrient, metal and microbial losses in runoff following treated sludge application to an Irish grassland soil

Dara Patrick Peyton, BAgrSc (Hons)

Research supervisors:

Dr. Mark Healy, Civil Engineering NUI Galway

Dr. Owen Fenton, Teagasc, Environment Research Centre, Johnstown Castle, Wexford

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To my wonderful parents, brothers, sisters and girlfriend for your guidance and support.

SUMMARY

Treated sewage sludge, commonly referred to as 'biosolids', is the organic by-product of urban waste water treatment. When spread on grassland or arable land, biosolids may provide an excellent source of nutrients and metals required for plant and crop growth. As biosolids are often considered a waste product, they may be used as a cheap source of organic fertiliser and may provide an excellent opportunity to improve crop profit margins by means of reducing the input costs of chemical fertilisers.

While there are many benefits associated with the use of biosolids as an organic fertilizer amendment, there are currently many concerns associated with their potential to contaminate soil, vegetation and water. In addition, current legislation does not consider the relationship between biosolids application rate and surface runoff. Therefore, the aim of this research was to: (1) undertake a literature review outlining the current situation of biosolids use, legislation, societal issues, various treatments of sewage sludge in Ireland, and advantages and disadvantages associated with their use, (2) produce a lime stabilised biosolid for use in a field scale experiment, (3) undertake a field-scale experiment to assess losses of nutrients (nitrogen and phosphorus), metals (copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), cadmium (Cd), chromium (Cr), microbial matter (total and faecal coliforms) following successive rainfall events on land onto which biosolids had been applied.

As part of this holistic investigation, three biosolids commonly used in Ireland were utilised: anaerobically digested, lime stabilised (LS) and thermally dried (TD). In addition, anaerobically digested biosolids, sourced from the Seventh Framework Programme (FP7) END-O-SLUDGE project, was also utilised and the fifth treatment was an unamended grassland control. For comparison with another commonly spread organic fertiliser using in Ireland, dairy cattle slurry (DCS) was also used in the experiment. Biosolids and DCS were

surface applied in accordance with the legislation in Ireland. A rainfall simulator was used to generate runoff over three successive events (24 hr, 48 hr and 360 hr) after a single application.

Losses from biosolids-amended plots were higher than the study control (soil only) plots, and followed a general trend of highest losses occurring during the first rainfall event and reduced losses in the subsequent events. However, with the exception of total coliforms and some metal parameters (Cu), the greatest losses were from the DCS-amended plots. For example, average losses over the three rainfall events for dissolved reactive phosphorus and ammonium-nitrogen were 4.5 and 11.6 mg L^{-1}, respectively, which were far in excess of the losses from the biosolids plots. Metal losses from DCS-amended plots were higher (Cd, Cr), or of the same magnitude as the biosolids-amended plots (Ni, Pb, Zn).

When compared with slurry treatments, biosolids do not pose a greater risk in terms of losses along the runoff pathway. This finding has important policy implications, as it shows that fears surrounding the reuse of biosolids as a soil fertiliser, mainly concerning contaminant losses upon land application, may be unfounded.

DECLARATION

This dissertation is the result of my own work, except where explicit reference is made to the work of others, and has not been submitted for another qualification to this or any other university.

Dara Patrick Peyton

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ABBREVIATIONS

AD	Anaerobic digestion
ATAD	Autothermal, thermophilic aerobic digestion
ADIRE	Anaerobically digestion Biosolid sourced in Ireland
ADUK	Anaerobically digestion Biosolid sourced in the United Kingdom
APHA	American Public Health Association
BS	British Standards
BSE	Bovine Spongiform Encephalopathy
Ca	Calcium
Cd	Cadmium
Cr	Chromium
Cu	Copper
DAFF	Department of Agriculture, Fisheries and Food
DAFM	Department of Agriculture, Food and The Marine
DCS	Dairy Cattle Slurry
DM	Dry matter
DRP	Dissolved reactive phosphorus
DS	Dried solids
DSC	Dry Solids Content
DUP	Dissolved un-reactive phosphorus
EC	European Commission
EPA	Environment Protection Agency
EU	European Union
Fe	Iron
FC	Faecal Coliforms
FRV	Fertiliser Replacement Value
FSAI	Food Safety Authority of Ireland
FWMC	Flow-weighted mean concentration
hr	Hour
ha	Hectare
Hg	Mercury
K	Potassium

Pb	Lead
LS	Lime stabilised
M3	Mehlich-III P
Mg	Magnesium
Mn	Manganese
N	Nitrogen
Ni	Nickel
OM	Organic matter
OMFs	Organomineral fertilisers
P	Phosphorus
P_2O_5	Phosphorus pentoxide
Pb	Lead
PCPs	Personal Care Products
PDS	Particle size distribution
Pm	Morgan's P
PP	Particulate phosphorus
PBT	Persistence, Bioaccumulation and Toxicity
PPCPs	Pharmaceuticals and Personal Care Products
RS1	Rainfall simulation event 1
RS2	Rainfall simulation event 2
RS3	Rainfall simulation event 3
FP7	Seventh Framework Programme
SS	Suspended sediment
STP	Soil test phosphorus
TC	Total Coliforms
TD	Thermally dried
TS	Total Solids
TDP	Total dissolved phosphorus in water
TN	Total nitrogen
TON	Total oxidized nitrogen
TP	Total phosphorus
U.S.A.	United States of America
USEPA	United States Environment Protection Agency

UC	Christiansen coefficient
WEF	Water Environment Federation
WWTP	Wastewater Treatment Plant
WFD	Water Framework Directive
Zn	Zinc

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

Chapter 1 –INTRODUCTION

1.1. Overview

In the European Union (EU), implementation of directives and other legislative measures in recent decades concerning 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 pressure on WWTPs (European Community (EC), 2014). Consequently, production of untreated sewage sludge in 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 further expected to increase to 13 million tonnes in all EU member states by 2020 (EC, 2010).

The treatment and disposal of sewage sludge presents a major challenge in wastewater management and, consequently, there is a need to find a cost-effective and innovative solution for its disposal (Hall, 2000). In the EU, the drive to reuse sewage sludge has been is a result of various directives, which advocate the re-use of sludge and limit the disposal of biodegradable municipal waste *via* landfill. In addition, the minimisation, recycling and recovering of waste is one of the six key goals outlined by the Environment Protection Agency (EPA). The legislation concerning sewage sludge production has actively prompted those involved in sludge management to find alternative uses for sludge, such as in the production of energy, bio-plastics, polymers, and other potentially useful materials (Healy et al., 2015). Recycling to land is currently considered the most economical and beneficial way for sewage sludge management (Haynes et al., 2009; Peters et al., 2009; Healy et al., 2015). Recycling of biosolids to agricultural land is relatively less expensive compared to incineration and landfill per tonne of raw sludge (DM) (Antille et al., 2013). However, before

this can occur, it must be treated by one or more of the recommended process as set down in the guidelines to prevent harmful effects on soil, vegetation, animals and humans (EC, 2014), after which they may be referred to as 'biosolids'. The term 'biosolids' was formally created in 1991 by the Name Change Task Force of the Water Environment Federation (WEF., 2005) to differentiate raw, untreated sewage sludge from treated and tested sewage sludge that can legally be utilized as a soil amendment and fertiliser.

Although there are many benefits associated with the use of biosolids on agricultural land, biosolids can, and often do, contain other less useful and potentially dangerous constituents such as metals, so-called 'emerging' organic pharmaceutical contaminants and human enteric pathogens, which have been discussed by Lu et al. (2012) and Singh et al. (2008), amongst others. These concerns become more prevalent when losses due to episodic rainfall events following land application are transferred to water bodies *via* direct discharge surface pathways or groundwater discharge. Although much research work has investigated their impact on nutrient, metal and suspended sediment (SS) release, many knowledge gaps still exist surrounding the potential impact arising from the landspreading of biosolids. In addition, the relative impact of different types of biosolids (lime stabilised (LS), anaerobically digested (AD) and thermally dried (TD)), when spread at the same application rate, has not yet been compared on a micro-plot field scale.

The specific objectives of this current runoff study were to:

1) Review the legislation and guidelines governing the application of biosolids to land and to elucidate research to date involving their use.
2) Develop a simple, novel, field-scale micro-plot study to determine the impact of land applications of three types of biosolids (1) AD biosolids from a WWTP in the United Kingdom (ADUK) (2) AD biosolids sourced in Ireland (ADIRE) (3) TD biosolids (4)

LS biosolids, (5) grass-only (the study control), and compare them to a commonly spread organic amendment in Ireland (6) dairy cattle slurry (DCS).

3) Conduct in-field simulated rainfall events (24, 48 and 360 hr after land application of biosolids) to measure incidental losses of nutrients, metals and microbial matter

1.2. Procedure

A literature review examined current legislation governing the landspreading of biosolids and the potential impact that this could have on water quality when spread within (and outside) current guideline limits. The literature review suggested that further investigation was warranted into the potential impact of biosolids on surface runoff following landspreading within current maximum legal application rates in Ireland and, in particular, the impact of surface runoff of so-called 'emerging' organic pharmaceutical and personal care products (PPCPs), sometimes found in biosolids, as they have been shown to have the highest risk ranking of PPCPs based on the factors of persistence, bioaccumulation and toxicity. The relative impact of environmental pollution of different types of biosolids (AD, LS, and TD), when spread at the same application rate, has not been compared at micro-field scale in Ireland. As a result of these knowledge gaps, the experiments were designed accordingly.

Thirty micro-plots, each measuring 0.9 m long and 0.4 m wide, were hydraulically isolated, and the soil was characterised for texture, particle size distribution (% sand/silt/clay), nutrients and metals. Following this, AD and TD biosolids and dewatered sludge cake were collected from a WWTP in Ireland. The dewatered sludge cake was manually lime stabilised under laboratory conditions to create LS biosolids for use in the experiment (this was to ensure the biosolids came from the same source). The biosolids were then characterised for nutrient, microbial and metal content. In addition, AD biosolids, sourced from the Seventh Framework Programme (FP7) END-O-SLUDGE project, was used in the experiment. The maximum permissible application rate under European legislation for the different types of

biosolids was then determined based on the soil test phosphorus (STP) content of the micro-plots, the legal limits for N, P, metal application; DM, nutrient, and metal concentration of the biosolids.

Biosolids were then randomly assigned to the twenty five micro-plots, and three rainfall simulations were conducted over a period of 15 days after land application. Grass samples from each plot were also collected prior to each rainfall simulation event to examine the uptake of metals. In addition, surface runoff from five micro-plots amended with DCS, which is commonly land applied as an organic fertiliser in Ireland, was examined so that a comparison of environmental losses could be made with the biosolids.

1.3. Structure of dissertation

Chapter 2 presents the literature review that was conducted in this study. Chapter 3 discusses importance of rainfall simulators in agricultural research and the design of the rainfall similar used in this study. Chapter 3 also describes the rainfall simulator and how it was calibrated. Chapter 4 describes a bench-scale test used to determine the incorporation of lime (calcium oxide, CaO) into dewatered sludge and to create LS biosolids. Chapter 5 describes the field rainfall simulator study and presents the surface runoff results for biosolids and DCS. Finally, Chapter 6 presents the conclusions and recommendations of the study.

1.4 Study outcomes to date

Book chapter:

Healy, M.G., Clarke, R., Peyton, D., Cummins, E., Moynihan, E.L., Martins, A., Beraud, P., Fenton, O. 2015. Resource Recovery from sludge. p. 139 - 162. In K. Konstantinos, K.P.

Tsagarakis (Eds.) Sewage treatment plants: economic evaluation of innovative technologies for energy efficiency. IWA, London.

Journal:

Peyton, D.P, Healy, M.G, Fleming, G.T.A., Grant, J., Wall, D., Morrison, L., Cormican, M., Fenton, O. Nutrient, metal, microbial and persona care product losses in runoff following treated sludge application to an Irish grassland soil: a rainfall simulation study. Submitted to Science of the Total Environment.

Conferences:

Healy, M.G., Morrison, L., Forrestal, P.J., Peyton, D., Fleming, G.T.A., Danaher, M., Wall, D., Cormican, M., Fenton, O. 2015. Characterisation of metal concentrations in treated municipal sludge in Ireland and impacts on runoff water quality following land application. International Conference on Solid Wastes 2015: Knowledge Transfer for Sustainable Resource Management. Hong Kong SAR, China. 19 – 23 May, 2015

Healy, M.G., Peyton, D., Fleming, G., Danaher, M., Morrison, L., Wall, D., Grant, J., Cormican, M., Fenton, O. 2014. Measurement of surface runoff of mixed contaminants arising from the landspreading of treated sewage sludge. ASA, CSSA, and SSSA International Annual Meeting. Nov 2 – 5, Long Beach, CA.

Peyton, D.P, Healy, M.G, Fleming, G.T.A., Grant, J., Wall, D., Morrison, L., Cormican, M., Fenton, O. 2015. Nutrient, metal, microbial and persona care product losses in runoff following treated sludge application to an Irish grassland soil: a rainfall simulation study. 25th Annual SETAC Europe Conference Meeting 3-7 May Barcelona, Spain 2015 (*poster presentation*)

Chapter 2 - LITERATURE REVIEW

2.1. Overview

This chapter reviews the use of biosolids in agriculture, and investigates their potential impact on surface and groundwater quality.

2.2. Introduction

Sewage sludge is the inevitable organic by-product of urban waste water treatment (Fehily, Timoney and Company, 1999), and is formed when wastewater undergoes various physical, chemical and biological processes to separate water from solids. Following appropriate treatment of sewage sludge by one of more the recommended process, treated sewage sludge, hereby referred to as "biosolids", may be successfully recycled and applied to agricultural land as an organic fertiliser (USEPA, 2012). When spread on tillage or grassland, they offer an excellent source of nutrient and metals required for plant and crop growth (Lucid et al, 2014). As demands for food and energy are expected to increase from a growing population (FAO, 2009), the demands for nitrogen (N), phosphorus (P), and potassium (K) are also expected to increase at an average rate of 2.5% per year to 2020 (Heffer et al., 2013), and as a result, the price of chemical fertiliser is also expected to increase (Heffer et al., 2013).

As biosolids are often considered a waste product, they may be used as a cheap source of fertiliser and may provide an excellent opportunity to improve crop profit margins by means of reducing the input costs of chemical fertilisers. The recycling of biosolids to agricultural land is also seen as a means to reduce dependence on phosphate rock (Antille et al., 2013). Although there are many benefits associated with the land application of biosolids on

agricultural land, environmental pollution as a result of losses of nutrients and, in particular, other less useful and potentially dangerous constituents such as metals, human enteric pathogens, and so-called 'emerging' PPCP contaminants, following an episodic rainfall event, may result in the limitation of biosolids as a fertiliser. It is therefore essential to investigate the many knowledge gaps currently associated with the landspreading of biosolids, so that any potential nutrient recovery from biosolids is considered against possible adverse impacts on the environment are minimised.

2.3. Sewage Sludge as a Resource

Biosolids may be used as an agricultural fertiliser, as they contain organic matter (OM) and inorganic elements (Girovich, 1996). The recycling of biosolids to agriculture as a source of the fundamental nutrients and metals required for plant growth is going to be essential for future sustainable development, as it is estimated that there are only reserves of 50-100 years of P depending on future demand (Cordell et al., 2009). Evans (2009) highlighted that up to 95% of P can be recovered from wastewaters and concentrated into the raw sludge. As P is a limited resource, any recovery and utilisation is a significant step in reducing the rate of depletion. When spread on arable or grassland, and provided that it is treated to the approved standards, biosolids may offer an excellent source of nutrients and metals required for plant and crop growth (Jeng et al., 2006). Biosolids may also contribute to improving soil physical and chemical characteristics (Mondini et al., 2008). It increases water absorbency and tilth, and may reduce the possibility of soil erosion (Meyer et al., 2001).

Land application of biosolids to agricultural land can be relatively inexpensive in countries in which it is considered to be a waste material. An alternative, but costly, option in such countries is to pay tipping fees for its disposal (Sonon et al., 2009). However, in some countries sewage sludge is seen not as a waste but instead as a product containing valuable

nutrients (e.g. the U.K and Ireland) with an associated fertiliser replacement value (FRV) and cost for its usage.

As the world population increases, pressure on natural resources, especially food, oil and water, will increase. Inorganic fertilizer prices are tied to crude oil prices globally and demand (Bremer, 2009): when prices of oil are high, inorganic fertilizer prices also climb. For instance, in Ireland, the cost of inorganic fertilisers has continually increased, with the cost of a mean kg of N, P and K rising from €0.41, 1.06 and 0.23 in 1980 to €103, 203, 105 in 2011 (Fig. 2.1). Similar price increases of 13% were seen in the U.K. in 2010 (Tasker, 2010). Recent fertiliser increases since 2008 can be attributed to increases in both energy costs and global demand for fertilisers. Increased prices and volatility are important considerations, as they lead to volatility in farm input costs and profit margins, and make farm planning more difficult and risky (Lalor et al., 2012).

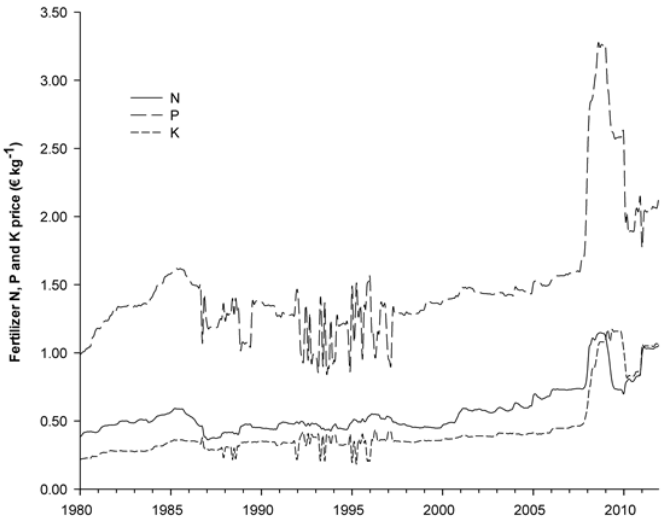


Figure 2.1. Trends in unit cost of nitrogen (N), phosphorus (P) and potassium (K) in chemical fertilisers in Ireland from 1980 to 2011 (Lalor et al. 2012).

Nutrient price equivalents of sewage sludge will depend on the nutrient availability and the FRV of the nutrients in the sludge. The FRV of nutrients in cattle slurry over time was calculated in Lalor et al. (2012) assuming a total N, P and K content in slurry of 3.6, 0.6 and 4.3 kg m^{-3}, respectively, and an assumption of respective FRV of 25%, 100% and 100% (Coulter., 2004). Of course in biosolids, as in other nutrient streams, micronutrients used by the plant give added value to the product. In addition, factors such as transport and land application costs would also need to be considered in an overall assessment. It is therefore essential that such data are known for biosolids.

There is a good body of literature that has examined its fertilisation potential (Smith et al., 2002; Epstein, 2003; Singh et al., 2008). Siddique et al. (2004) mixed AD-treated sewage sludge, poultry litter, cattle slurry and an inorganic P fertiliser with five soil types at rates equivalent to 100 mg P kg^{-1} soil and, following incubation at 25^{o}C for 100 d, found that AD-biosolids and poultry litter had a slower rate of P release compared with cattle slurry and inorganic P fertiliser. This may indicate that it may have good long-term fertilisation potential.

2.4. Legislation governing disposal of biosolids

The drive to recycle biosolids to agricultural land has been accelerated by, amongst other legislation, the Landfill Directive, 1999/31/EC (EC, 1999), the Urban Wastewater Treatment Directive 91/271/EEC (EC 1991), the Waste Framework Directive (2008/98/EC; EC 2008), and the Renewable Energy Directive (2009/28/EC; EC 2009), which places an increased emphasis on the production of biomass-derived energy. However, one of the main pieces of legislation governing the use of biosolids in agriculture in Ireland and the EU is the Sewage Sludge Directive 86/278/EEC (EEC, 1986), which seeks to encourage the use of sewage sludge in agriculture and to regulate its use in such a way as to prevent harmful effects on

soil, vegetation, animals and man. In Ireland, the directive is enacted in the "Codes of Good Practice for the Use of Biosolids in Agriculture" (Fehily, Timoney and Company, 1999) which set out limits for metal application (Table 2.1), and S.I. No. 610 of 2010, which sets out nutrient limits for various crops grown in Ireland.

The Directive 86/278/EEC and the Codes of Good Practice specifies rules for the sampling and analysis of sludge and soils. It also sets out requirements for the keeping up to date records on the quantities of sludge produced by each EU member state, the quantities used in agriculture, the composition and properties of the sludge, the type of sludge treatment, and the sites where the sludge is used and disposed. It also sets out requirements on the concentrations of metals in biosolids intended for agricultural use and in biosolids-treated soils (Table 2.1). The Directive 86/278/EEC and the Codes of Good Practice also specifies rules which detail issues such as constraints on grazing for animals and cultivation of crops following land application of biosolids, the types of crops and lands on which the biosolids may be spread, the times of the year when the land application of biosolids is prohibited, and safe spreading distances from entities such as watercourses. As a result of the legislation, land application of biosolids in the EU is typically based on its nutrient and metal content, although individual member states often have more stringent limits than the Directive (EC 2010; Milieu et al. 2013a,b,c). Generally, when applying biosolids based on these guidelines and depending on the nutrient and metal content of the biosolids, P in the majority of cases becomes the limiting factor. However, while the guidelines aim to prevent harmful effects, they do not consider the relationship between biosolids application rate, nutrient availability, and surface runoff of nutrients, microbes and metals.

Table 2.1. Limit values for metal concentrations in sludge and soil (taken from Lucid et al., 2013).

Limit values	Copper (Cu)	Nickel (Ni)	Lead (Pb)	Zinc (Zn)	Cadmium (Cd)	Chromium (Cr)	Mercury (Hg)
	---------- mg kg^{-1} ----------						
European Union[a]							
For concentrations of heavy metals in soil	50 - 140	30 - 75	50 - 300	150 – 300	1 - 3	-	1 - 1.5
For heavy metal concentrations in sludge for use in agriculture	1,000 - 1,750	300 -400	750 - 1,200	2,500 - 4,000	20 - 40	-	16 - 25
	---------- kg $ha^{-1}y^{-1}$ ----------						
For amount of heavy metal that may be applied annually to soil	12.0	3.0	15.0	30.0	0.15	-	0.1
Ireland							
For average annual rate of addition of metal (over a 10 yr period)[b]	7.5	3.0	4.0	7.5	0.05	3.5	0.1

[a] Limit values taken from Directive 86/278/EEC (EEC, 1986).

[b] Limit values taken from (Fehily, Timoney and Company, 1999).

While the Directive 86/278/EEC and the Codes of Good Practice share many of the regulations, there are a number of exemptions and provisions in the current regulation which should be removed or amended with the Codes of Good Practice, as it will give rise to further food safety concerns. For example, the Codes of Good Practice states that untreated wastewaters sludge should not be landspread or injected into soil. However, 86/278/EEC states the latter provided that it has been injected or incorporated into the soil.

2.5. Wastewater treatment

The purpose of wastewater treatment is to remove contaminants from wastewater, including household sewage and runoff, while producing an environmentally safe fluid waste stream (treated effluent) and a solid waste (treated sludge) suitable for disposal or reuse as a farm fertiliser. Wastewater treatment involves the physical, chemical, and biological treatment or a combination of these processes depending on the nature of the inflow influent wastewater, together the water quality objectives of the receiving bodies (Grey, 2002). The treatment process is classified into five main stages: preliminary, primary secondary, tertiary and sludge treatment. A simplified wastewater treatment process is illustrated in Fig. 2.2 comprising the five main treatment processes.

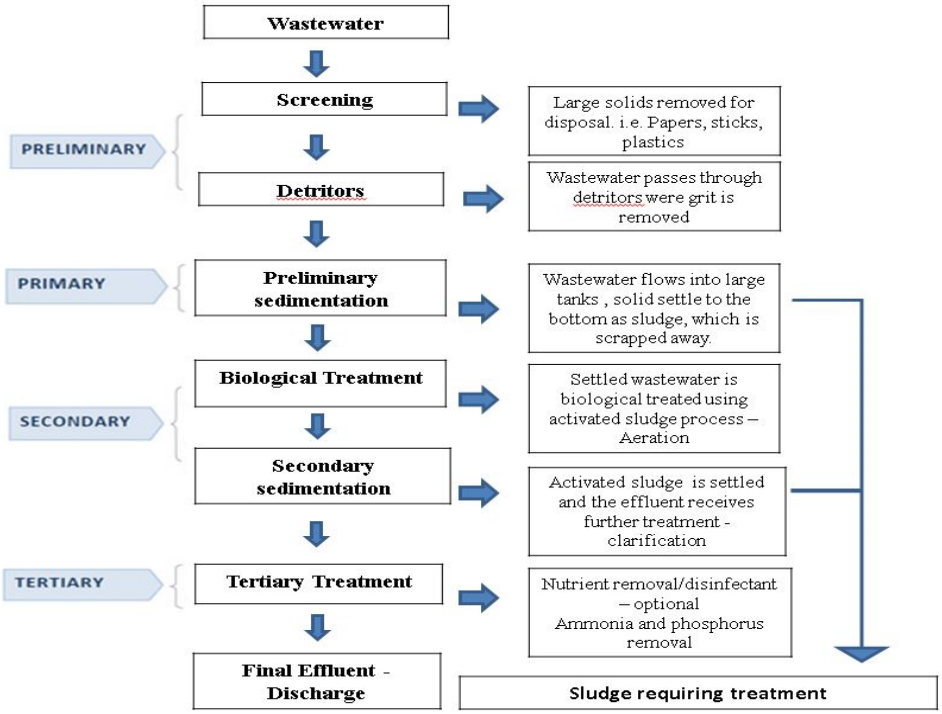


Figure 2.2. Illustration of a simplified wastewater treatment process (adapted from Antille et al., 2011; Metcalf et al., 2003).

2.5.1. Preliminary treatment

The purpose of preliminary treatment is to protect the operation of the wastewater treatment plant. Preliminary wastewater treatment involves the removal of coarse solids and other large materials that may cause operational and maintenance of subsequent treatment units (FAO, 2014). Solids that may be removed during this process may consist of pieces of wood, plastics, paper, cloth, together with some faecal matter. Heavy inorganic solids such as sand and gravel, as well as metals or glass, are removed at this stage, and finally excessive amounts of oils or greases in the influent wastewater are also removed. Flow equalization of the inflow is also controlled at this stage (EPA, 1997).

2.5.2. Primary treatment

The purpose of primary treatment is to remove organic and inorganic solids by sedimentation. There are two main methods employed during this stage. The first is physical settlement, which involves the removal of settleable solids from base of the tank (removed as primary sludge). The second method is chemical coagulation and flocculation. This involves the addition of chemical coagulants to the influent, where the coagulant encourages insoluble material to form flocks. Following further settlement, these are removed as primary sludge. Primary treatment can reduce the biochemical oxygen demand (BOD) by 30 - 40%, SS by 40 - 70%, and faecal coliforms (FC) by up to 50% (Grey, 2002).

2.5.3. Secondary treatment

Secondary treatment involves the removal of biodegradable dissolved and colloidal OM. This treatment process uses microorganisms to convert soluble and colloidal OM into carbon dioxide, water and new cells (Lehany, 2003). Secondary settlement tanks are used to separate microorganisms from the treated wastewater to produce clarified secondary effluent. The biological solids removed during secondary treatment are combined with the primary sludge for sludge treatment (FAO, 2014). Secondary sludge is composed mainly of biological cells, in contrast to the primary sludge, which is composed mainly of faecal solids (Lehany, 2003). There are several secondary biological treatment processes available, which include trickling filters or biofilters, fixed film reactors, activated sludge systems, and stabilisation ponds (FAO, 2014; Gray, 2002; Metcalf et al., 2003;).

2.4.4.Tertiary treatment

Tertiary treatment further removes BOD, SS, bacteria, potentially toxic element and nutrients (Lehany, 2003). Tertiary treatment is needed when wastewater is discharged to sensitive water bodies (Antille, 2011). A number of systems are available for tertiary treatment, and a

detailed overview of them is given in Metcalf et al. (2003). These include: prolonged settlement in lagoons or irrigation onto grasslands or percolation areas, wetlands, disinfection by either of the two main methods - ultraviolet (UV) treatment or chlorination, chemical precipitation (e.g. ferric chloride, aluminium sulphate or lime), which react with the soluble phosphate to produce an insoluble precipitate.

2.5.5. Sludge treatment

Sludge is the organic by product arising from the treatment of wastewater, which requires further treatment (i.e. to produce biosolids) for their safe use as an agricultural fertiliser. One of the main objectives of sludge treatment is reduced water content (dewatering) prior to disposal. Primary and secondary sludge typically comprise 97 to 99.5% water (Ruiz-Hernandoet et al., 2013) and as a result, dewatering is necessary to reduce the total sludge volume as well increasing its handling characteristics. Sludge dewatering is completed by gravity thickening, using a belt filter press or by using drying beds. Dewatering using these methods will leave a composition of solids in treated sewage sludge of between 12 and 30%, and between 80 and 90% for sludge treated by thermal drying (Lehany, 2003).

An important objective of sludge treatment is the reduction or removal of pathogens to an acceptable level and reduction of attractiveness of sewage sludge to vectors. As untreated sewage sludge contains high levels of pathogens (e.g., bacteria, viruses, protozoa, helminths) (Sidhu et al., 2009), if applied to agricultural lands, they will have the potential to contaminate soil, vegetation and water. Due to the lack of well-developed methods for the detection and enumeration of pathogens (Sidhu et al., 2009), the use of indicator organisms such as FC along with *Salmonella* species, are used to evaluate the microbiological contamination of biosolids. The microbiological standards used are derived from the USEPA

part 503 biosolids rule (USEPA, 1993) (Table 2.2). Class A biosolids are treated to a higher standard than Class B biosolids and also require less or no restrictions on buffer requirements, public access, or crop harvesting restriction (USEPA, 1993), while these are required for virtually all forms of Class B biosolids (USEPA, 2012). While Class A biosolids are treated to a higher standard than Class B biosolids, the long-term application of Class B biosolids to land is still regarded as sustainable, with the risk of pathogens posing a low threat to human health (Pepper, 2008). In Ireland, the microbiological standards are defined under the code of good practice for the use of biosolids in agriculture (Fehily, Timoney and Company., 1999), and are equivalent to Class A biosolids.

Table 2.2. Microbiological standards used to classify biosolids as Class A or Class B biosolids (Fehily, Timoney and Company., 1999; USEPA, 1993)

Type of biosolids	Microbiological standards used to classify biosolids
Class A	Either the density of faecal coliforms in the biosolids must be less than 1 x 10^3 most probable number (MPN) per gram total solids (DM), or the density of *Salmonella* species bacteria in the biosolids must be less than 3 MPN per 4 g of total solids (DM) and time of use or disposal.
Class B	Class B biosolids are treated by the same process as Class A biosolids, but can contain detectible levels of faecal coliforms up to 2 x 10^6 MPN g^{-1} DS. For this reason, Class B biosolids are required to have site restriction, preventing crop harvesting, animal grazing and access to the public for specific period of time following application until pathogen levels have further reduced (USEPA, 1993).

2.6. Types of treated biosolids

Stabilisation is designed to control potential putrefaction process, odour releases and vector attraction. A variety of sewage sludge treatment technologies can be employed and are implemented according to regulations. As can be seen from Table 2.3, significant differences in sewage sludge treatment exist. At present, in Ireland, there are five main methods adopted

for the treatment of biosolids before land application: AD, TD, composting, LS and autothermal, thermophilic aerobic digestion (Fehily, Timoney and Company, 2007).

Table 2.3. Global municipal sewage sludge treatment processes

	Denmark[a]	France[a]	Germany[a]	Greece[a,b]	Ireland[a]	Italy[a]	Spain[a]	Sweden[a]	UK[a]	Czech Rep.[a]	Poland[a]	USA[c]	Portugal[d]
Stabilisation													
Aerobic	✓	✓		✓✓	✓	✓	✓	✓	✓	✓	✓✓	✓	✓
Anaerobic	✓	✓	✓	✓	✓	✓✓	✓✓	✓	✓✓	✓✓	✓	✓	✓
Lime	✓	✓		✓	✓	✓	✓	✓	✓		✓	✓	✓
Composting	✓	✓		✓	✓	✓	✓	✓	✓	✓✓	✓	✓	✓
Conditioning													
Lime	✓					✓							
Inorganics						✓		✓					
Polymers				✓									
Thermal			✓			✓		✓			✓		
Drying belts				✓									
								✓					
Dewatering													
Filter press		✓		✓	✓	✓			✓		✓		
Centrifuges		✓						✓			✓		✓
Belt filter press	✓			✓✓	✓	✓		✓	✓		✓		✓
Others													
Thermal		✓	✓✓	✓	✓	✓	✓	✓	✓			✓	
Solar drying	✓	✓				✓	✓						
Pasteurisation												✓	
Long-term storage				✓	✓	✓	✓	✓	✓		✓		
Cold fermentation bag filling											✓		

✓ Common use ✓✓ most common use

[a] Kelessidis at al., (2012); [b] Tsagarakis et al. (1999) [c] Lu et al., (2012)

2.6.1. Anaerobic digestion

Anaerobic digestion is a common method for the treatment of sewage sludge prior to land application. It involves the incubation of sludge under anaerobic conditions for a mean

retention time of at least 12 days of primary digestion at the mesophilic temperature range of 35°C, or of at least 20 days of primary digestion at a temperature of 25°C, or the thermophilic temperature of 55^{o}C for a mean retention time of 48-72 hr. The AD process works by the stabilising the organic material and reducing the pathogenic content by utilising certain microbes that thrive in an environment that lacks oxygen. During the process, organic material is converted to methane, carbon dioxide and digestate. Anaerobic digestion produces Class A biosolids at thermophilic temperatures and Class B biosolids at mesophilic temperatures (Epstein, 2002). It is required that the AD process undergo a pasteurisation phase in which there is a retention period of at least 1 hr for a temperature of 70^{o}C and 2 hr for a temperature of 55^{o}C (Fehily, Timoney and Company, 1999). Anaerobically digested biosolids are shown in Fig. 2.3. The advantages of AD are that the methane gas can be subsequently used as an energy source (Epsten, 2002), the mass and volume of the sludge are reduced, a low running cost and high loading rates (Grey, 2002). The disadvantages include long start up times due to slow growth rate of anaerobic bacteria, the highly polluted supernatant arising from thickening and dewatering and the sensitivity to chemicals, pH variation and toxic overloads (Spinosa et al., 2001).

Figure 2.3. A 50 g sample of anaerobically digested biosolids (ADIRE)

2.6.2. Thermal drying

Thermal drying technology is based on the removal of water from dewatered solids by evaporation of water, which dramatically reduces achieves both volume and weight. The result is a Class A product with a DM content of approximately 90% (Fig. 2.4). The high temperatures used in the production of TD biosolids ensure a sufficient reduction in pathogen numbers and the temperatures used, while high, are generally low enough to prevent oxidation of OM. In recent years, TD biosolids pellets have been mixed with urea, potash and other substance to created organomineral fertilisers (OMFs) (Antille et al., 2013). The advantages of thermal drying is that approximately 90-95% DM can be achieved, and that the sludge generated is stable, odourless and amenable to long-term storage, and that spreading techniques are similar to those used for mineral fertilizers. The disadvantage is the high capital investment and on-going operational cost (Lehaney et al., 2003)

Figure 2.4. A 50 g sample of thermally dried biosolids (TD)

2.6.3. Lime Stabilisation

Lime stabilisation, commonly known as alkaline stabilisation, raises the pH level of the sludge, thus making conditions unfavourable for the growth of organisms. Lime stabilisation is increasingly used in countries because it is a cost effective way of stabilisation municipal sludge (Krach et al., 2008). Materials that may be used for alkaline stabilization include hydrated lime, CaO, commonly known as quicklime or burnt lime; fly ash, lime and cement kiln dust, and carbide lime (USEPA, 2000). However, CaO is commonly used because it has a high heat of hydrolysis, which can significant enhance pathogen destruction (USEPA, 2000). The high temperature and pH inhibits biological action, therefore inactivating pathogens in the treated biosolids product (Joyce et al., 2014). The rate at which lime is added to achieve these regulations is dependent on dry solid content content of the sludge produced (Andreadakis, 2000). In addition, the extent of heat generated is also dependent on the lime dose rate (Smith et al., 1998).

The effectiveness of the lime stabilisation process for pathogen reduction and odour is dependent on significant lime addition and incorporation (Burns et al., 2007). Uniform lime incorporation is critical to the lime stabilisation process, as it is important to eliminate regions with low pH within the lime-sludge mix. Poor lime incorporation will result in inadequately stabilised regions, leading to microbial regrowth, driving further pH reduction and causing increased odour (Burns et al., 2007). Lime stabilised biosolids also offer the benefit of a substitute for agricultural lime (Jacobs et al., 2003). Lime biosolids are shown in Fig. 2.5.

Figure 2.5. **A 50 g sample of lime stabilised biosolids (LS).**

2.6.4. Composting

Composting is the biological degradation of OM, resulting in the formation of a stable end product. Composting of sludge produces a humus-rich material that can be applied directly to land to provide a nutrient benefit, or to add organic content and improve the tilth of a soil (Fehily, Timoney and Company, 2007). However, composted biosolids generally spread as a

soil improver rather than fertiliser, as their fertilising capability is a function of time and maturity of the material (Fehily, Timoney and Company, 2007).

The method of composting is wide ranging and varied, but the two main methods involve windrows or aerated static piles. Due to the waterr content and the fine particle size of sludge, it needs to be mixed with an amendment material to provide further bulking and space for the passage of air through the material. The amendment material is generally shredded green waste, woodchip and, in some cases, shredded tires (USEPA, 2002). Composted biosolids may also be mixed with other materials e.g. household or commercial food waste. The 'Codes of Good Practice for the Use of Biosolids in Agriculture' gives time – temperature recommendations for the sanitisation of material when composting biosolids. Windrow composting must be held at 55°C for at least 15 days, during which time the material must be turned 5 times. In-vessel or static pile composting requires maintaining a temperature of 55°C for a minimum of 3 days. However, the beneficial reuse of compost as an organic fertiliser can be limited, as Sidhu (2001) highlighted that composted biosolids have a *Salmonella* re-growth potential. As a result, long-term storage is not recommended

2.6.5. Autothermal Thermophililic Aerobic Digestion

Autothermal thermophilic aerobic digestion (ATAD) is a biological sludge treatment process that converts soluble organics to lower energy forms through fermentative anaerobic and aerobic processes. Autothermal TAD is an exothermic process where sludge is subjected to temperatures > 55 °C and a hydraulic retention time of 6–15 days (Layden et al., 2007). Organic solids are degraded and the heat released during the microbial degradation maintains thermophilic temperatures. Autothermal TAD can produce a biologically stable product while reducing both sludge mass and volume (Bernard et al., 2000). Minimum concentrations of total volatile solids of 3 – 4% and total solids (TS) of 5-6% are also typical requirements

(Fehily, Timoney and Company, 2007). At present, there is only one plant in Ireland producing Class A biosolids by ATAD, which is located in Killarney, County Kerry.

2.7. Existing and emerging issues concerning the use of biosolids on agricultural land

2.7.1. Nutrient and metal losses

Phosphorus and reactive N losses to a surface waterbody originates 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 waterbody occur *via* primary drainage systems (end of pipe discharges, open drain networks) (Ibrahim et al., 2013), runoff and/or groundwater discharges. Application of biosolids to soils may also contribute to STP build-up in soils, thereby contributing to chronic losses of P, metal and pathogen losses in runoff (Gerba et al., 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 (Domagalski et al., 2011; 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; EEC, 1986). However, guidelines governing the application of treated sewage sludge to land (e.g. Fehily Timoney and Company 1999) mean that 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). In countries such as the USA, where treated sewage sludge is land applied in the majority of states (e.g. exclude Maryland) based on the N requirement of the crop being grown and not on a soil-based test (McDonald et al., 2011), excessive metal losses may potentially occur.

2.7.2. Behaviour of metals in the soil/Uptake by plants

The potential of biosolids to contaminating soils with heavy metals has caused great concern about their application on agricultural land (Wuana et al., 2011). Heavy metals most commonly found in biosolids are lead (Pb), nickel (Ni), cadmium (Cd), chromium (Cr), copper (Cu), and zinc (Zn), and the metal concentrations are governed by the nature and the intensity of the industrial activity, as well as the type waste water treatment employed (Silveira et al., 2003). As a result, emphasis concerning land application of biosolids has been placed on these heavy metals. In Ireland, the code of good practice for the use of biosolids in agriculture places maximum concentrations limits on the loadings of these metals to agricultural land (Table 2.1). However, many other factors, including, application rate, pH and other soil characteristic such as OM content and redox potential, affect the accumulation of the metals in biosolid-amended soils (Hue et al., 1994; Singh et al., 2008; Smith, 2009).

As the application of biosolids to land may pose a risk to soil contamination due to metal accumulation, an understanding of behaviour of metals in the soil is essential for assessing environmental risks. One of the main concerns with the possible accumulation of metals in soil is the possibly of them being incorporated into plants. As a consequence, the consumption of plants containing high levels of metals may pose a serious threat to human health *via* the food chain (Silvera et al., 2003). Studies examining the uptake of metals by plants have focused on metals behaviour and fate in soils; and on three hypotheses, plateau, time bomb, and soil-plant barrier (Lu et al., 2012). The plateau hypotheses considers that metal bioavailability is greatly reduced as they are so tightly held by the OM and clay content of the soil, that they are retained in the soils surface horizon or plow layer, instead of leaching down the soil profile (Lu et al., 2012). Therefore, metal concentrations in plant tissues will reach a plateau as biosolids loading increases and will remain at this plateau even after land spreading has stopped (Ross, 1994). Time bomb theory suggest that metal concentration

bound to biosolids could be realised to soluble forms over time, therefore, becoming toxic to plants, as a time bomb (Lu et al., 2012). The soil-plant barrier theory indicates that plants play an important role in protecting the food chain, since transfer of metals to the edible part of the crop is under the physiological control of the plant (McBride, 2002). In addition, metals are tightly bonded to soil, limiting their transfer to the roots of a plant (McBride, 2002).

2.7.3. The microbial risk associated with the landspreading of biosolids

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, AD, or temperature, may substantially reduce pathogens, complete sterilisation is difficult to achieve (Sidhu et al., 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 (Sidhu et al., 2009). There can be also 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-culturable, 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 et al., 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, temperature, UV radiation, nutrient and oxygen availability, and

land management regimes (van Elsas et al., 2011), whereas survival in sludge is primarily related to temperature, pH, water content (of treated sewage sludge), and sunlight (Sidhu et al., 2009). Pertinent biotic interactions include antagonism from indigenous microorganisms, 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 which can lead to human infection. First, pathogens may be transported *via* overland or sub-surface flow to surface and ground waters, and infection may arise *via* ingestion of contaminated water or accidental ingestion of contaminated recreational water (Jaimeson et al., 2002; Tyrrel et al., 2003). Alternatively, 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 sanitization (Itoh et al., 1998; Solomon et al., 2002). In this case, a person may become infected if they consume the contaminated produce. Therefore, it is critical to accurately determine the pathogen risk associated with land application of biosolids to fully understand the potential for environmental loss and, consequently, human transmission.

However, survival patterns of sludge-derived pathogens in the environment are complex, and a lack of a standardised approach to pathogen measurement makes it difficult to quantify their impact. For example, Avery et al. (2005) spiked treated and untreated sludge samples with a known concentration of *E. coli* to quantify the time taken to achieve a reduction. The pathogen response was variable and ranged from 3 to 22 days, depending on sludge properties. Lang et al. (2007) investigated indigenous *E. coli* survival in dewatered, mesosphilic anaerobically digested (DMAD) sludge, and in different soil types post DMAD sludge application. Again, decimal reduction times proved variable, ranging from 100 days

when applied to air-dried sandy loam, to 200 days in air-dried, silty clay textured soil. This time decreased to 20 days for both soil types when field moist soil was used, demonstrating the importance of water content in regulating survival behaviour.

Therefore, in order to quantify pathogen risk in a relevant, site-specific manner, it is necessary to incorporate both soil and treated sewage sludge characteristics in risk assessment modelling. This has been done previously by conducting soil, sludge and animal slurry incubation studies, where pathogens are often spiked to generate a survival response (Vinten et al., 2004; Lang et al., 2007; Moynihan et al., 2013). Pathogen decay rate is then calculated based on decimal reduction times, or a first-order exponential decay model previously described by Vinten et al. (2004), and has been shown to be highly contingent on soil type and sludge or slurry combinations. Currently, the Safe Sludge Matrix provides a legal framework for grazing animals and harvesting crops following landspreading of treated sewage sludge, and stipulates that a time interval of three weeks and 10 months should be enforced to ensure safe practice, respectively (ADAS, 2001). However, further work is required to determine if these regulations are overly stringent, particularly in light of the comparatively higher pathogen concentrations reported for animal manures and slurries. For example, *E. coli* concentrations ranged from $3x10^2$ to $6x10^4$ colony forming units (CFU) g^{-1} in sludge (Payment et al., 2001), compared to $2.6x10^8$ to $7.5x10^4$ CFU g^{-1} in fresh and stored cattle slurry, respectively (Hutchison et al., 2004). Therefore, environmental losses associated with treated sewage sludge application may not be as extensive as previously thought and further comparisons on pathogen risk should form the basis of future research.

2.7.4. Pharmaceutical and personal care products

Pharmaceuticals comprise a diverse collection of thousands of chemical substances, including prescription and over-the-counter therapeutic drugs and veterinary drugs (USEPA, 2012).

Pharmaceuticals are specifically designed to alter both biochemical and physiological functions of biological systems in humans and animals (Walters et al., 2010). Pharmaceuticals are referred to as 'pseudo-persistent' contaminants (i.e. high transformation/removal rates are compensated by their continuous introduction into the environment) (Barceló et al., 2007). Pharmaceuticals are likely to be found in any body of water influenced by raw or treated waste water, including river, lakes, streams and groundwater, many of which are used as a drinking water source (Yang et al., 2011). Between 30 and 90% of an administered dose of many pharmaceuticals ingested by humans is excreted in the urine as the active substance (Cooper et al., 2008). In a survey conducted by the US Environmental Agency (McClellan et al., 2010), the mean concentration of 72 pharmaceuticals and personal care products were determined in 110 treated sewage sludge samples. Composite samples of archived treated sewage sludge, collected at 94 U.S. wastewater treatment plants from 32 states and the District of Columbia were analysed by liquid chromatography tandem mass spectrometry using EPA Method 1694. The two most abundant contaminants found in the survey were the disinfectants triclocarban and triclosan. The second most abundant class of pharmaceuticals found were antibiotics, particularly ciprofloxain, ofloxacin, 4-epitetra-cycline, tetracycline, minocycline, doxycycline and azithromycin (McClellan et al., 2010). It was concluded that the recycling of biosolids was a mechanism for the release of pharmaceuticals in the environment.

Pharmaceuticals have received increasing attention by the scientific community in recent years, due to the frequent occurrence in the environment and associated health risks (Chen et al., 2013). In 2007, the European Medicines Agency (EMEA) issued a guidance document (ERApharm) on environmental risk assessment of human medicinal products. It relies on the risk quotient approach used in the EU and is also used for industrial chemicals and biocides, where the predicted environmental concentration is compared to the predicted no-effect

concentration. The overall objective of ERApharm is to improve and complement existing knowledge and procedures for environmental risk of human and veterinary pharmaceuticals. The project covers fate and exposure assessment, effects assessment and environmental risk assessment (Lienert et al., 2007). A considerable amount of work focused on three case studies. Two of the case studies focused on human pharmaceuticals, β-blocker atenolol and the anti-depressant fluoxetine, and the third on a veterinary parasiticide ivermectin. Atenolol did not reveal any unacceptable risk to the environment but cannot be representative for other β-blockers, some of which show significantly different physiochemical characteristics and varying toxicological profiles in mammalian studies (Knacker et al., 2010). Although found in trace levels (several nanograms per litre), some therapeutic compounds such as synthetic sex hormones and antibiotics, have been found to cause adverse effects on aquatic organisms (Chen et al., 2013). Therefore, understanding their environmental behaviour and impact has recently become a topic of interest for many researchers.

2.7.5. Public perception of the land spreading of biosolids

Managing municipal and industrial biosolids by recycling to land application is currently a strategic policy directive in the EU. Management and treatment capacity of land application, as well as the economic benefits, makes recycling biosolids to agricultural land appealing. Although recycling biosolids to land is seen as a plausible management option, it is a contentious issue that has cause much public opposition and concern (Beecher et al., 2005). Concerns have been raised over potential health, safety, quality of life and environmental impacts that land spreading of biosolids may have (USEPA, 2002). While governmental bodies have laws, restriction and recommendation in place, public acceptance of land application of biosolids still remain mixed. There are a number of reasons that can be attributed to this and while traditional ways of addressing concerns has been through

scientific research (Tyson, 2002), many questions still remain. In addition to this, mistrust of the opinions of politicians and technical advisors, resulting from failed past environmental industrial incidence (Giusti, 2009), has created a sceptical public when it comes to new technology.

As public perception can be critical in influencing the choice of options used for biosolids management (USEPA, 2002), scepticism and mistrust of authorises and published science has led to the banning or restriction of land application in some countries. Although the quality of biosolids have improved over time with advancement in treatment technology (Robinson et al., 2012), public concerns remains about the long-term health effect of exposure to substances present in biosolids – especially through pathways such as food and soil. Food scares worldwide in recent decades such as the bovine spongiform encephalopathy (BSE), foot and mouth and, more recently, the Ecoli Cumcumber scares (BBC, 2011) and horsemeat scandel in Europe (EC, 2014), have had a detrimental effect on public confidence and farming practices. Although none of these presented cases are attrutued to the use of biosolids, the fact that diseases could be contracted by humans *via* the direct food chain resulting from farming practices has lead many people concerned about the use of biosolids in agriculture. This concern has seen the introduction of "sludge free labels" being added to packaging of food in some countries, mainly because the use of sludge is not considered to be acceptable for products with a high-quality image (EC, 2010). In surveys undertaken on public attitudes towards the land applcation of biosolids, interviewees are not enthusiastic about recycling biosolids into food growing land (Tanto et al., 2010). Surveys also showed that communities perceived greater health risk associated with exposure to biosolids than animal manure due to the presence of pathogens (Robinson, et al., 2012). This perception could be, in part, due to the fact that biosolids are heavily regulated or the fact that animal manure is more commonly seen and used.

As biosolids are brought to forefront of the general public's mind through increased land application, knowledge and awareness has been heightened (Robinson et al., 2012). As awareness, plays a key role in public perception of risk (Robinson et al., 2012), the public at large are now beginning to assess for themselves whether or not this activity is safe. While governments and environmental authorities have tried to manage and reduce risk associated with the reuse of treated sludge, effective management strategies should be to make sure that the public is aware of the risks associated their reuse (Robinson et al., 2012). People's acceptance of risk is often subjective and depends in part on their basic values and beliefs, as well as their training and experience (Harrison et al., 1999). In the past, waste management programs have tried to improve acceptability by explaining the risk factors, where lack of knowledge was deemed the primary issue (Nancarrow et al., 2008). While programs to increase public knowledge have helped in educating with facts, little attention has been given to addressing the values and beliefs driving the public's perception (Robinson et al., 2012). Research on public perception have shown that it is not often the overall concern with biosolids, but rather the associated factors such as the increase in vehicle movements, odour, or noise of machinery and equipment (Tyson, 2002; Beecher et al. 2004). Research has also shown that the public are far more likely to be tolerant and, in some cases, supportive if they have had their questions and concerns addressed (Tyson, 2002; Beecher et. al 2004). Norway is a prime example of a country that has gained the trust of public acceptance with more than 90% its sludge used as a soil improvement product on land (EC, 2010).

2.8. Summary

The use of organic biosolids as a replacement for inorganic (i.e. chemical) fertilisers has potential, provided they are spread within guideline limits. Where legislation is followed, land application of biosolids should not pose any greater risk to the environment than other organic fertilisers in terms of nutrient, metal and microbe losses. However, further research

will have to be carried out on emerging PPCPs to ensure their safe long-term use. At present, public perception is one of the major stumbling blocks surrounding their use as an organic amendment. However, further data on their potential impact on surface runoff of nutrients, microbes and metals will address some of these concerns. These will be addressed in the following chapters.

Chapter 3 - DESIGN OF A RAINFALL SIMULATOR

3.1 Overview

This chapter outlines the design, calibration and operation of the outdoor rainfall simulator used in this study.

3.2 Rainfall simulators and their importance in agricultural research

Rainfall simulators are an important tool in arable and grassland agricultural research and have been widely used for the assessment of soil hydrologic properties (Mohanty et al., 1996; Loch et al., 1987), soil erosion (Iserloh et al., 2012; Sukhanovskii, 2007), infiltration and runoff generation, and the movement of nutrients, metals or polluting agents in field and laboratory conditions (Brennan et al., 2012; Fernandez - Galvez et al., 2008; Kramers et al., 2009; Kurz et al., 2006; Lucid et al., 2013; Regan, 2012).

While natural rainfall is desirable, data collection can be slow, as precipitation characteristics such as intensity, spatial and temporal frequency and duration of natural rainfall cannot be controlled (Humphry et al., 2002). The use of rainfall simulators provide the opportunity for increased experimental control over the variables that govern natural rainfall (Júnior et al., 2011). As rainfall simulators provide this control, they allow for quick, specific and reproducible rainfall events (Iserloh et al., 2012), and therefore dependable data.

As stated in Bowyer-Bower et al. (1989), types and design of rainfall simulators have been developed since the first attempts by Dudley et al. (1932). Design characteristics must take into account operation requirements, drop sizes to be replicated, plot size, water usage, portability, ease of use and cost. However, designs are often centralised around two established dispersal methods: ‘spray type’ simulators using water sprayed from an irrigation sprinkle nozzle, or ‘drop forming’ simulators, which drip water from a suitable apparatus

(Bowyer-Bower et al., 1989). A more detailed synopses of these rainfall simulators can be found in Bubenzer et al. (1979), Agassi et al. (1999) and Bowyer-Bower et al. (1989).

Desirable rainfall characteristics should include drop size distribution similar to natural rainfall, rainfall intensity in the range of the requirement of the research program, uniformity over the study area, accurate reproduction of rainfall events, fall velocity, kinetic energy similar to natural rainfall and portability if for use *in situ* (Tossell et al., 1987; Humphry et al., 2002; Pall et al., 1983; Bowyer-Bower et al., 1989).

While rainfall simulators are a useful tool, there is no standardisation of rainfall simulator design, which may impede on drawing comparisons between results (Iserloh et al., 2012). In addition, their overall performance can be limiting (Humphry et al., 2002). Renard (1985) listed some of the disadvantages associated with the use of rainfall simulators, including the fact that areas simulated are typically small, ranging from less than a square metre up to several hundred square metres, depending on the design used. Most simulators do not produce drop size distribution similar to natural rainfall. Terminal velocities of natural rainfall is not produced by some simulators and as a result, the kinetic energy produced may only be 40 – 50% of natural rainfall in some nozzle drop formers or free falling dropper simulators. Although there are many disadvantages to the use of rainfall simulators, the key factor is whether the advantages outweigh the disadvantages (Neff, 1979). In many instances, simulated rainfall is the only effective way to obtain results in reasonable time frame and under controlled conditions (ASCE, 1996). Furthermore, the acquisition of data provides fundamental information on the cause/effect relationship of many agricultural research questions, as well as improving decision making on environmental protection (Iserloh et al., 2012). In addition, the cost associated with simulated rainfall is relatively inexpensive when

compared to long-term hydrologic experiments that rely solely on natural rain events (Foster, 2005).

3.3 Rainfall simulator in the current study

In the current study, an Amsterdam drip-type rainfall simulator, as described by Bowyer-Bower et al. (1989), was used to provide rainfall for the runoff experiment (Chapter 5). This type of rainfall simulator has been successfully used in micro-plot runoff experiments (Holden et al., 2002; Kurz et al. 2006; Brennan et al. 2012; Mohr et al. 2013). The simulator is driven solely by gravity, with raindrops falling from heights of one or two metres, which makes achievement of terminal velocity difficult (Bowyer-Bower et al., 1989). However, their accuracy in replicating rainfall between experimental sites is an advantage (Bowyer-Bower et al., 1989). In addition, these simulators are cheap and easily transported, making them potentially advantageous over their spray-type counterparts, which can be time consuming to construct and transport (Bowyer-Bower et al., 1989). In addition, these simulators allow for simple, small-scale side-by-side rainfall simulations to be conducted on different treatments.

3.4 Rainfall simulator construction

The simulator used in the current study was designed to form droplets of median diameter 2.3 mm, spaced 30 mm apart in a 1000 mm × 500 mm × 8 mm Perspex plate over a 0.5 m^2 simulator area. The principal components are shown in Fig. 3.1. Drops are formed by controlling flow though Tygon tubing of 2.3 mm outside diameter (OD) and 0.7 mm internal diameter (ID). The ID tubing determines the rate of water drop formation, which is then further slowed down by lengths of fishing line inserted into each tube. The Perspex plate of drop formers contained 420 drop formers arranged in a 14 × 30 matrix. The water reservoir consisted of two 25 L water tanks mounted above the Perspex plate. The pressure head in the

two tanks are maintained at a target level of ± 10 mm. An adaptation of the rainfall simulator as used by Brennan et al. (2012), was the addition of gate flow pressure values, inside of manometer board, which were used to control flow rate to the Perspex plate, which, in turn, controls the rainfall intensity.

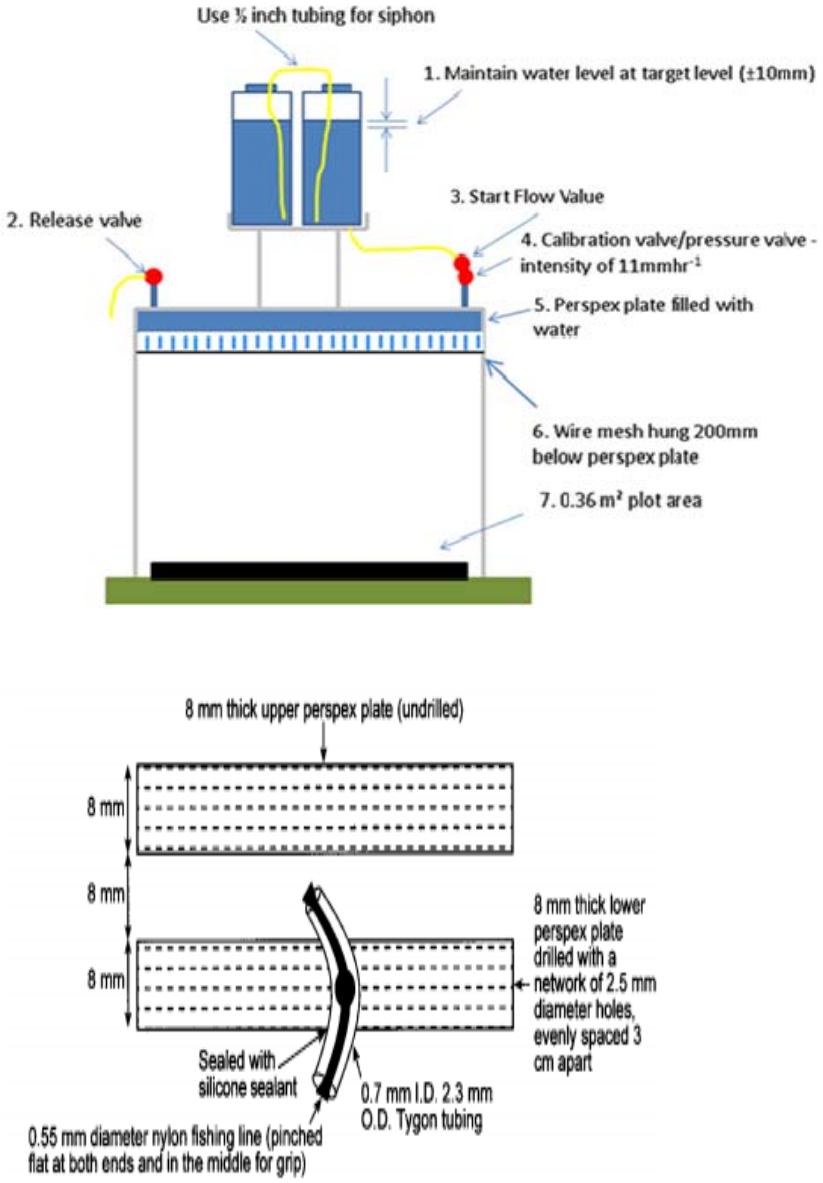


Figure 3.1. Principal components of the rainfall simulator (top) and of Perspex plate (bottom)

The simulator was supported by a metal frame, which was fitted with adjustable legs so that the simulators could be levelled. This ensured that water droplets fell from a level surface. The frames of the simulators were also fitted with plastic sheets so that simulated rainfall was protected from wind effects (Fig. 3.2). A wire mesh was hung 200 mm below the Perspex plate so that water droplets could be the intercepted, coagulating them and dispersing others to create drop sizes, similar to that of natural rainfall. A fall height of 2 m was achieved with this rainfall simulator. The simulator was calibrated to achieve a target rainfall intensity of 11 mm hr^{-1}, which is not uncommon hourly rate for a short term rainfall event in Ireland (Met Eireann, 2015). The rainfall simulators can be seen in use in this YouTube video: https://www.youtube.com/watch?v=JYhsmE8SHvU.

3.5 Areal uniformity and intensity calibration

Uniformity of the areal distribution of rainfall is an important measurement of a rainfall simulator's performance, as it reflects the ability of the simulator to evenly distribute rainfall over a surface area being examined. Rainfall intensity measurement is also important, as variations in intensity can influence the experimental results. Performance tests for intensity and uniformity were conducted in accordance with Tossell et al. (1987). The target area for simulated rainfall was 0.36 m^2 (0.9 × 0.4 m). The simulation area was reduced so that the effect of edge effects could be eliminated. The intensity of simulated rain was determined by collecting a volume of water during a known period of rainfall. Large trays of known area were placed underneath the rainfall simulator in the target area to take a representative sample of simulated rainfall. This enabled the calculation of rainfall intensity:

$$\frac{\text{Volume of water collected (L) x 60 min}}{\text{Length (m) x width (m) x 60 min}} \qquad [3.1]$$

Figure 3.2. Amsterdam-styled drip-type rainfall simulators fitted with wind shield in use in field.

Uniformity was determined by positioning 15 collection containers (68 mm ID and 75mm deep) under the rainfall simulator (Fig. 3.3) during an experimental run. They were then weighed. The uniformity of application could then be determined after Christiansen (1942). The Christiansen uniformity coefficient (UC) is a measure of the spatial distribution of simulated rain falling over a defined area, and is calculated by:

$$CU = 100\left(1 - \left(\sum_{i=1}^{i=n} \frac{x_i}{x} - \bar{x}\right)\right) \quad [3.2]$$

where X is the mean rainfall intensity (mm hr^{-1}), n is the number of observations, and X_i (i = 1, 2, 3, . .,n) are the individual observations. As the number and size of the collection containers will affect the results of uniformity trails, greater theoretical accuracy may be achieved by increasing the number of collection containers. However, this can be extremely time consuming, and the amount of information collected is offset by the time involved (Tossell et al., 1987). Conversely, few large gauges covering the entire plot can be misleading; therefore, it is more informative to use small collection containers spaced evenly over the plot.

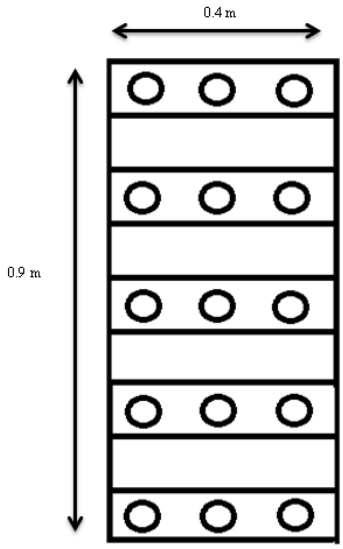


Figure 3.3. Calibration area and positions of collection containers

3.6 Summary

This chapter gives a brief explanation of the importance of rainfall simulators as a tool in arable and grassland agricultural research. The chapter also gives a brief explanation and guide on how to preform one of the most importance steps in using a rainfall similar i.e. calibration. The rainfall simulator described in this chapter was a drop-forming rainfall

simulator and will be used in the surface runoff experiment in Chapter 5. The simulator provided everything required for this research in terms of portability, ease of use and cost, but more importantly, it produced desirable rainfall characteristics.

Chapter 4 - METHODOLOGY TO INCORPORATE CALCIUM OXIDE INTO DEWATERED SLUDGE

4.1. Overview

In this chapter, a bench-scale test was used to incorporate calcium oxide (CaO) into dewatered sludge under laboratory conditions. This created lime-adjusted biosolids for use in the micro-plot scale experiment described in Chapter 5.

4.2. Introduction

Lime stabilisation, commonly known as alkaline stabilisation, is an internationally recognised method used by WWTPs for the treatment of sewage sludge. Alkaline stabilisation of sludge works by raising the pH level of the sludge, thus making unfavourable conditions for the growth of organisms. The process of alkaline stabilisation is increasingly used in countries, as it is a cost-effective way of stabilisation sewage sludge (Krach et al., 2008). Materials that may be used for alkaline stabilization include hydrated lime, CaO, fly ash, lime and cement kiln dust, and carbide lime (USEPA, 2000). However, CaO is commonly used because it has a high heat of hydrolysis, which can significantly enhance pathogen destruction (USEPA, 2000), creating a better stabilised sludge product.

In accordance with Irish regulation set out under the "Code of Good Practice for the Use of Biosolids in Agriculture", the quantity of lime added must increase the pH of the lime-sludge mix to ≥12 and the temperature to 70˚C for 30 minutes, or to increase the pH above 12 for 72 hr and maintain a temperature of ≥ 52˚C for 12 hr, or greater (Fehily, Timoney and Company, 1999). The high temperature and pH inhibits biological action, therefore inactivating pathogens in the treated biosolids product (Joyce et al., 2014). The rate at which lime is

added to achieve these regulations is dependent on the DS content of the sludge (Andreadakis, 2000). In addition, the extent of heat generated is also dependent on the lime dose (Smith et al., 1998). The exothermic reaction is shown in the following hydration reaction:

$$CaO \text{ (quicklime)} + H_2O \text{ (water)} = Ca(OH)_2 \text{ hydrated lime} + \text{ Heat} \quad [4.1]$$

The effectiveness of the lime stabilisation is dependent on the achievement and maintenance of pH ≥12, which is dependent on lime addition and significant lime incorporation (Burns et al., 2007). Uniform lime incorporation is critical to the lime stabilisation process, as poor lime incorporation will result in inadequately stabilised regions, leading to microbial regrowth, driving further pH reduction and causing increased odour (Burns et al., 2007).

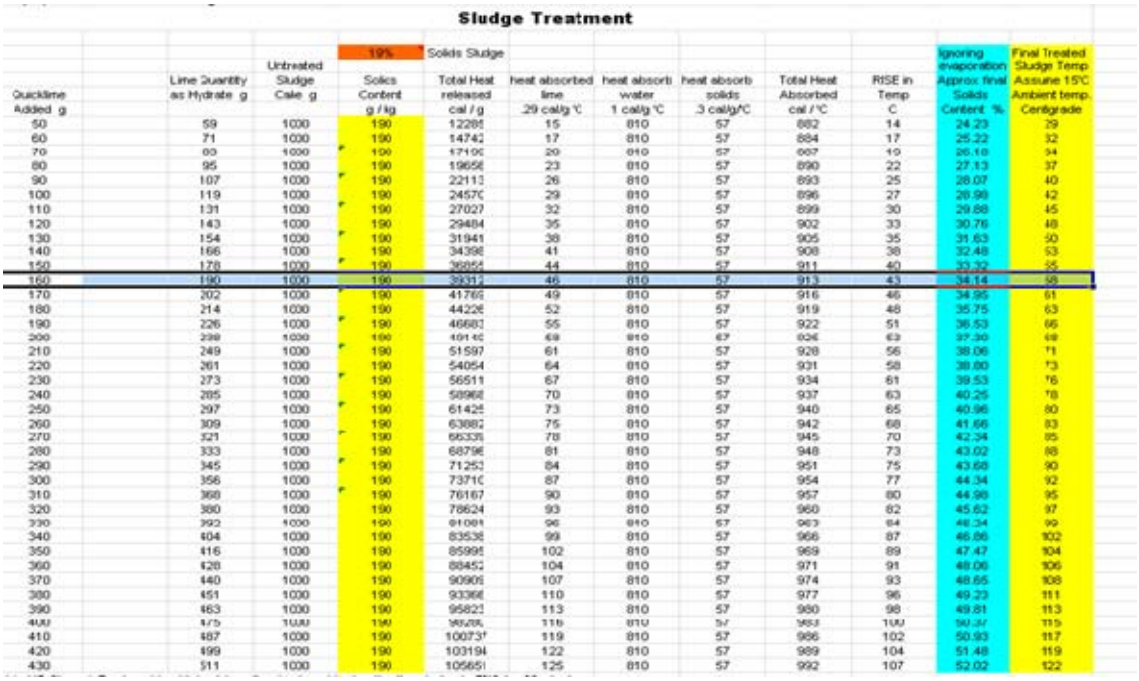
Therefore, the objectives of the bench-scale test was to created lime-adjusted biosolids for use in the micro-plot scale experiment described in Chapter 5, while following the protocols for pathogen kill and heat requirement currently in place in Ireland.

4.3. Materials and Methods

4.3.1. Sample collection and analysis

Dewatered sludge cake was collected in sealed 50 L-capacity plastic storage boxes from a WWTP in Ireland and transported to Teagasc, Environment Research Centre, Johnstown Castle, Co Wexford, where it was labelled and stored at 4^{o}C until lime was added. To determine the amount of CaO that needed to be added to the mixture, the dry solid content content of the dewatered sludge cake was determined by drying eight representative 50 g samples at 105^{o}C for 24 hr. The dry solid content content of the sludge cake was determined to be 19±0.64%. The CaO was obtained from Clogrennane Lime, Co. Carlow, Ireland - a major provider of CaO to WWTP facilities in Ireland. In addition, an in-house spread sheet,

provided by Clogrennane Lime, was used to calculate the amount of lime to add to this dry solid content to comply with the current regulations (Fig. 4.1).



Sludge Treatment

Quicklime Added g	Lime Quantity as Hydrate g	Untreated Sludge Cake g	19% Solids Content g / kg	Total Heat released cal / g	heat absorbed lime .29 cal/g °C	heat absorbi water 1 cal/g °C	heat absorb solids .3 cal/g/°C	Total Heat Absorbed cal / °C	RISE in Temp C	Ignoring evaporation Approx final Solids Content %	Final Treated Sludge Temp Assume 15°C Ambient temp. Centigrade
50	59	1000	190	12285	15	810	57	882	14	24.23	29
60	71	1000	190	14742	17	810	57	884	17	25.22	32
70	83	1000	190	17199	20	810	57	887	19	26.19	34
80	95	1000	190	19656	23	810	57	890	22	27.13	37
90	107	1000	190	22113	26	810	57	893	25	28.07	40
100	119	1000	190	24570	29	810	57	896	27	28.98	42
110	131	1000	190	27027	32	810	57	899	30	29.88	45
120	143	1000	190	29484	35	810	57	902	33	30.76	48
130	154	1000	190	31941	38	810	57	905	35	31.63	50
140	166	1000	190	34398	41	810	57	908	38	32.48	53
150	178	1000	190	36855	44	810	57	911	40	33.32	55
160	190	1000	190	39312	46	810	57	913	43	34.14	58
170	202	1000	190	41769	49	810	57	916	46	34.95	61
180	214	1000	190	44226	52	810	57	919	48	35.75	63
190	226	1000	190	46683	55	810	57	922	51	36.53	66
200	238	1000	190	49140	58	810	57	925	53	37.30	68
210	249	1000	190	51597	61	810	57	928	56	38.06	71
220	261	1000	190	54054	64	810	57	931	58	38.80	73
230	273	1000	190	56511	67	810	57	934	61	39.53	76
240	285	1000	190	58968	70	810	57	937	63	40.25	78
250	297	1000	190	61425	73	810	57	940	65	40.96	80
260	309	1000	190	63882	75	810	57	942	68	41.66	83
270	321	1000	190	66339	78	810	57	945	70	42.34	85
280	333	1000	190	68796	81	810	57	948	73	43.02	88
290	345	1000	190	71253	84	810	57	951	75	43.68	90
300	356	1000	190	73710	87	810	57	954	77	44.34	92
310	368	1000	190	76167	90	810	57	957	80	44.98	95
320	380	1000	190	78624	93	810	57	960	82	45.62	97
330	392	1000	190	81081	96	810	57	963	84	46.24	99
340	404	1000	190	83538	99	810	57	966	87	46.86	102
350	416	1000	190	85995	102	810	57	969	89	47.47	104
360	428	1000	190	88452	104	810	57	971	91	48.06	106
370	440	1000	190	90909	107	810	57	974	93	48.65	108
380	451	1000	190	93366	110	810	57	977	96	49.23	111
390	463	1000	190	95823	113	810	57	980	98	49.81	113
400	475	1000	190	98280	116	810	57	983	100	50.37	115
410	487	1000	190	100737	119	810	57	986	102	50.93	117
420	499	1000	190	103194	122	810	57	989	104	51.48	119
430	511	1000	190	105651	125	810	57	992	107	52.02	122

Figure 4.1. The spreadsheet provided by Clogrennane Lime, which shows a 19% dry solid content with the amount of CaO required, highlighted, to get the required heat.

4.3.2. Monitoring of pH and temperature and microbes

To determine the pH of the sludge-lime mixture (Section 4.3.3 and 4.3.4), 10 g of sludge-lime mixed was added to 20 mL of deionized water (1:2 ratio sludge-lime mixture:water). The mix was then shaken for 5 min using an adapted New Brunswick Scientific Gyrotory Shaker, before allowing to stand for 5 min. The pH was then measured using a Jenway 3510 pH meter, and temperature was measured using a Testo 925 Thermometer probe and a temperature probe attached to the Jenway 3510.

4.3.3. Test 1 (preliminary test)

First, a single replication of 100 g of dewatered sludge cake was mixed with CaO at 12%, 13% and 16 %, based on the wet weight of the sludge. This was equivalent to 12 g, 13 g and 16 g of CaO, respectively. As dewatered sludge cake had been stored in the cold room at 4^{o}C and only removed earlier that day, samples were too cold to generate the heat required for stabilisation (≥52^{o}C). A larger volume of sludge and lime addition, coupled with a warmer room temperature equivalent, was needed. Therefore, a single replication of 200 g dewatered sludge cake (which was allowed to stabilise to room temperature) was mixed with CaO at 17%, 20% and 25% based on the wet weight of the sludge (Fig. 4.2). This was equivalent to 34 g, 40 g and 50 g of CaO, respectively.

Figure 4.2. The experimental setup for the preliminary test. A) – temperature monitoring of 2 kg sludge cake and B) – a temperature probe close up.

Another test was conducted to test the theory that a greater quantity of dewatered sludge cake would be a more crucial factor in increasing and maintaining the sludge-lime mix at the recommend temperature. This test mixed 2 kg of dewatered sludge cake with 20% or 400g of CaO. Sludge and lime were hand mixed for 5 min until the lime was sufficiently incorporated into the sludge.

4.3.4. Test 2 (Full-scale bench test)

Results from the preliminary tests allowed the following setup to be justified. A 16% lime-sludge mix was used in this test. 15 kg of dewatered cake was mixed with 2.4 kg of CaO (Fig. 4.2). Sludge and lime were hand mixed together until lime was sufficiently incorporated in the plastic container that had been used for collection and storage of sludge. Similar to the preliminary tests (Section 4.3.3), the mixing time was 5 min. To ensure proper lime incorporation to the sludge, CaO was added in stages and not at once. After mixing, two temperature probes were inserted into the sludge-lime mixture and the box lid was closed. This provided better insulation to the mixture and helped maintain temperature (Fig. 4.3).

Figure 4.3. The measurement of lime and sludge, and sealed container mixture with temperature probes inserted.

Temperature was measured with two temperature probes every 10 min after the mixing time had stopped for the first 3 hr and then every hour after that for 12 hr. Four pH measurements were taken daily for 72 hr, three representative samples of 10 g each, with the fourth a composite samples of five 2 g samples pooled together. While the lime–sludge mixture was being tested daily for pH, it was stored outdoors for 48 hr, stirring once a day with a spade to allow for the further reduction of water content by air drying (Fig. 4.4). The final lime-sludge is shown in Fig. 4.4.

Figure 4.4. Shows final lime-sludge mixture and storage outside for 48 hours

4.4. Results

4.4.1. Test 1 (preliminary test)

This first test using 100 g of dewatered sludge cake mixed together with CaO at 12%, 13% and 16% recorded peak temperatures, after 10 min, of 23^{o}C, 26^{o}C and 29.1^{o}C, respectively. The sludge was at pH 5.88 before any addition of lime, and increased over the recommend pH of 12 for all three lime mixes, and had pH readings ranging from 12.4 - 12.6 (Table 4.1).

For the second test (200 g), the subsample removed from the cold room had an initial temperature of between 12.9^{o}C – 13.5^{o}C. After mixing, the temperature was 34^{o}C, 41.2^{o}C and 44^{o}C, respectively, for the 17%, 20% and 25% CaO mixes. Temperature decreased to 29.3^{o}C, 39.1^{o}C and 41^{o}C, respectively, after 30 min. Temperature of the 25% reduced to 22.4^{o}C after 90 min. A pH test for the 25% mixture was 12.4 (Table 4.1).

For the third test (2000 g), temperature ranged from 15.2^{o}C – 15.6^{o}C before the addition of lime to the sludge. The recommended ≥52^{o}C was exceeded; temperatures of 57.7^{o}C and 56.5^{o}C were reached after half hour (Table 4.1). However, these temperatures fell below 52^{o}C after 30 min to an average temperature of 41^{o}C.

Table 4.1. Results for 100 g, 200 g and 2000 g of dewatered sludge mixed with varying percentages of quicklime (CaO).

Weight of sludge w/w	Weight of Mix used d/w	Percentage Mix	Temperature (◦C)		pH	
g	g	%	After mix	½ hr after	After mix	24 hrs
100	12	12	23	-	12.6	-
100	13	13	26.1	-	12.6	-
100	16	16	29.1	-	12.4	12.4
200	34	17	34	29.3	-	-
200	40	20	41.2	39.1	-	-
200	50	25	44	41	12.4	-
2000	400	20	≥52	41	-	-

4.4.2. Test 2 (Full–scale Bench test)

When 16% CaO was added to the dewatered sludge cake, the recommended ≥52^{o}C was observed in temperature gauge B after 50 min and remained above ≥52^{o}C for 90 min. The peak temperature observed in gauge B was 53.6^{o}C. For gauge A, the peak temperature observed was 51.4^{o}C after 60 min before declining exponentially. The average temperature for both gauges showed that temperature reached ≥52^{o}C after 50 min, but fell below after 90 min. Fig. 4.5 shows a graph of both temperature gauges, with the average temperature of both A and B gauges over the 12-hour observation period. The pH was measured in the sample over a 72-hr period, and showed that pH remained above the recommend value of 12 as recommend in the legislation (Table 4.2). In addition, the average TC and FC (±std. dev.) biosolids also proved to be a Class A standard (Table 5.4)

Table 4.2 Sample pH of lime - sludge mix at 24, 48 and 72 hr periods.

Sample	Day 1 (24 hours)	Day 2 (48 hours)	Day 3 (72 hours)
	pH		
1	12.73	12.66	12.70
2	12.72	12.70	12.62

3	12.74	12.71	12.59
4*	12.74	12.66	12.58

*Composite sample

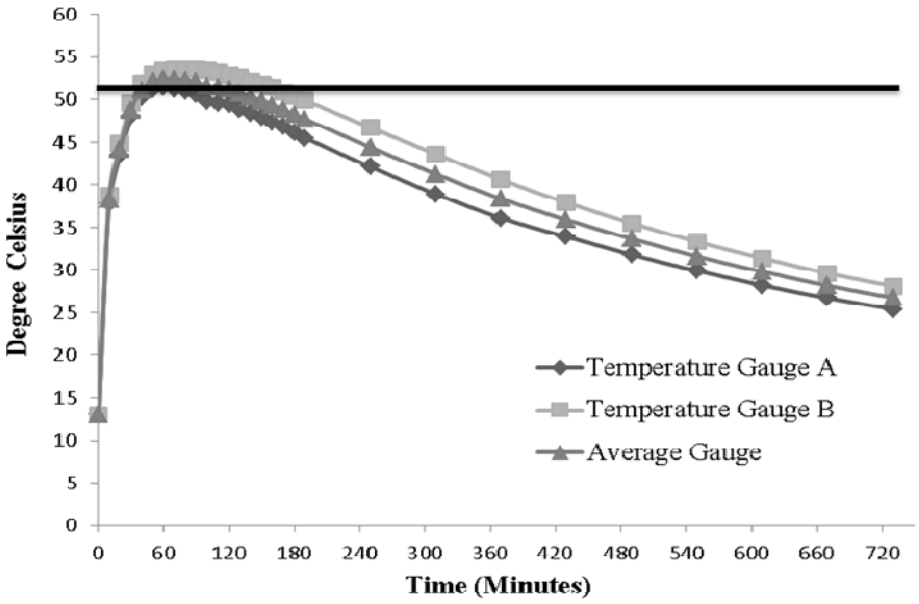


Figure 4.5. Temperature vs. time over 12 hr. Horizontal bar indicates 52°C, which is the temperature guideline (Fehily, Timoney and Company, 1999)

4.5 Discussion

4.5.1. Preliminary test

Obtaining the guideline temperature for the required time period, for achievement of a lime adjusted sludge treatment sample proved difficult. No sample during the 100 g or 200 g tests reached the recommended guideline temperature; even with the increase in lime to 25% wet weight. This was also the case for the greater quantity of dewatered sludge cake used in the third preliminary test. Although the temperature target of ≥52°C was obtained using a 20% lime mix in the 2 kg test, preliminary test results suggested that obtaining and maintaining the required temperature guideline was quantity dependent (i.e. the amount of sludge

incorporated into the mix) rather than an excessive lime dose. Dewatered sludge, when mixed on a bigger scale such as at a WWTP, will have increased insulation in storage mounds. Fig. 4.6 shows a typically lime biosolid processing at a WWTP, which combines all processed sludge together on a truck trailer before removal to a bigger storage mound before application. The mixing process is better illustrated in Fig. 4.7

Figure 4.6. A) Standard lime-sludge mixing apparatus at a WWTP, B) Pugmill Augers, C) completion of sludge and lime mixture on transfer belt, D) truck collection.

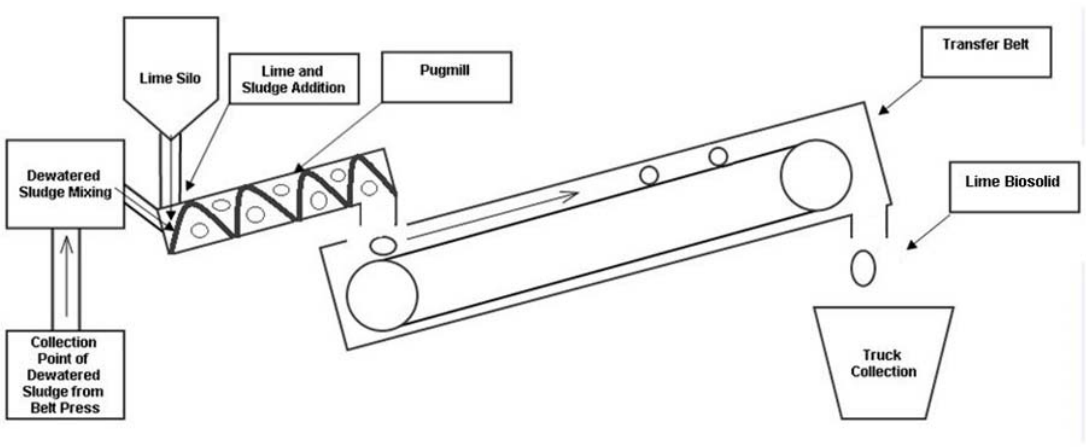


Figure 4.7. Illustration of a standard mixing apparatus at a WWTP

As a result, quantity dependence proved to be the case when comparing the 20% lime mix at 200 g with 2000 g in the mini-scale test. It was concluded from the mini-scale test that a 20% amount, although reaching the target, would not be a representative liming amount used by the wastewater treatment industry to stabilise sludge. It was felt that for economic reasons, a 20% liming amount to sludge would not be cost-effective, but as treatment plants deal with higher volumes of sludge, the amount required would not have to be the same to reach the recommended heating requirement. It was concluded that with a larger amount of sludge, such as the amount which was used in the full-scale test, the lime dosage could be smaller due to greater heating capacity and insulation of heat in a mound. It was for this reason and with consultation of the spread sheet provided by Clogrennane lime, a 16% liming requirement based on wet weight was used in the full-scale test. Although slightly higher than that recommended by Clogrennane Lime, whose recommendation is 14%, it was felt that the increase in lime would ensure that temperature was reached, while at the same time not overdosing the dewatered sludge on the smaller laboratory scale study. A 16% liming requirement is also in line with recommendation used by the European Lime Association,

whose recommendations for typical CaO addition for advanced treatment of dewatered sludge is 50-90% CaO per unit DS (European Lime Association, 2014). A 16% liming based on dry weight, used in this study, would give an 84% CaO per unit DS.

4.5.2. Full–scale Bench test

For the full-scale test, the temperature reached the recommended temperature for gauge B, but not for gauge A. However, gauge B did not stay above the recommended ≥ 52 °C for the 12-hr period. However, as biosolids are stored in mounds in WWTPs, the area of the heap exposed to the elements will cool quickly and may not maintain the ≥ 52 °C for a 12-hr period. Further study is required to ensure that WWTPs are complying with these recommendations. The temperature results in this test are similar to a study undertaken by Smith et al. (1998), who reported maximum temperature reached within 1 hr before declining exponentially when mixing 100g (DW) dewatered biosolids (15% dry solids content) mixed with lime at different rates of 5, 15, 20, 25, 30, 40, 50% by weight based on wet weight. Although the heating requirement was not maintained for the required 12 hr as per code of good practice, the average total and faecal coliforms were of Class A standard. This result is similar to the experiences of lime stabilization of sludge conducted by Araque (2006) and Torres et al. (2009), and proves that the code of good practice concerning the heating requirement may need updating and if it commonly being obtained or followed at WWTP.

4.5.3. Importance of uniform lime incorporation and potential problems

Uniform lime incorporation is critical to the lime stabilisation process as it is important for the elimination of regions with low pH within the lime-sludge mix. Poor lime incorporation will result in inadequately stabilised regions, leading to microbial regrowth, driving further pH reduction and causing increased odour (Burns et al., 2007). A study by Krach et al. (2008) showed that longer mixing times and proper lime dosage could efficiently reduce odour

offensiveness and that lime biosolids with a better mixing time has a much slower pH decrease than biosolids with poor mixing. It has also been noted that a drop in pH levels creates a favourable environment for the reactivation of regrowth of pathogens (Wong at al., 2000). North et al. (2008) found that faecal coliform levels were reduced by longer mixing times as a result of a more uniform distribution of lime into the mixture.

The slurry pH method used to test lime biosolids pH can be prone to error (Burns et al., 2007). The disadvantage of using this method is that when biosolids are made into a slurry, the lime and biosolids are homogenized together, making all the lime reactive, thus masking regions with poor lime incorporation. In addition to this, the heating pasteurisation requirement stated in Irish good practices, especially the monitoring temperature for an extended period (i.e. >52◦C for 12 hr), has also failed to be replicated in studies by Lozada et al. (2009) and Smith et al. (1998). However, the experiences of lime stabilization of Araque (2006) and Torres et al. (2009) showed that the biosolids derived from WWTPs that do not fulfil this requirement may also achieve Class A biosolid status in terms of microbial kill. In addition, the overdosing of lime to obtain the heating requirement may result in higher than normal operation cost.

Other problems with the heating requirement is the overall monitoring as the way that lime biosolids are produced in Ireland means this measurement is not that feasible. In addition, under Section 51 of Waste Management Act, lime stabilisation plants in Ireland are exempt from a waste permit/licence if sludge goes onto agriculture land, resulting in no processing standard monitoring e.g. temperature and no testing for pathogens before release of material (Cré, 2013). As there is currently a knowledge gap surrounding the heating requirement, the effectiveness of lime stabilisation in Ireland and pH maintenance and pathogen survival in storage, there is a need for research into lime stabilising process and its effectiveness to

minimise food safety concerns. This study also hypothesised that a slower rotor or longer rotor mixing area before the conveyor belt will allow better incorporation of lime and therefore a better chance at fulfilling the regulations.

4.5. Conclusion

The objective of the study was to produce a lime stabilised biosolids under laboratory conditions following as closely as possible the guidelines set down in code of good practice. Maintaining temperatures above ≥52°C for 12 hr, as stated in the code of good practice, does not seem to be practical at small scale, but may be possible at full scale, provided a sufficient amount of lime is incorporated into large volumes of sludge. A decrease in temperature was measured within 12 hr in all tests. However, the recommended pH was achieved and maintained for the 72-hr period. The heating requirement and uniformity of lime incorporation requires further study to ensure that WWTPs are complying with temperature regulations.

Chapter 5 - PLOT-SCALE RAINFALL SIMULATOR STUDY

5.1. Overview

This plot scale experiment was designed and developed to understand the potential environmental impact of surface runoff resulting from the land spreading of three types of biosolids on agricultural land. Biosolids were surface applied to grassland with no incorporation into the soil, and simulated rainfall was used to produce surface runoff. For comparison to a commonly spread organic fertiliser in Ireland, DCS was also applied to plots. Surface runoff was collected and tested for nutrients, metals, and microbes.

5.2. Introduction

Incidental losses of nutrient, metal and microbes as a result of an episodic rainfall event soon after land application of biosolids are of particular concern, as they have the potential to cause eutrophication and pollution to water bodies. As land application is currently promoted through legislation in the European Union, any potential benefits arising from the reuse of biosolids must be considered against possible adverse impacts associated with their use. The objectives of this study was to simultaneously assess, surface runoff of nutrients (P and N), metals (Cd, Cr Cu, Pb, Ni, and Zn) and microbes (FC and TC), under controlled conditions in field conditions during and after rainfall simulation events, using three types of treated biosolids. For comparison, a commonly spread organic fertiliser in Ireland, DCS, was also applied to plots, and surface runoff was tested for the same parameters as the biosolids.

5.3 Materials and Methods

5.3.1. Field Site characterisation

The study site was a 0.6-ha plot located at Teagasc, Johnstown Castle Environment Research Centre, Co. Wexford, Ireland (latitude 52.293415, longitude -6.518497) in the southeast of Ireland. The area has a cool maritime climate, with an average temperature of 10^{o}C and mean annual precipitation of 1002 mm. The site has been used as a grassland sward for over twenty 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). Soil nutrient analysis for the field site was characterised by dividing the site into an upper, middle and lower section, and by taking three bulked soil samples (n=20) before characterising each section separately (Fig. 5.1). The soil nutrient status at these locations (Morgan's P (P_m), K, and magnesium (Mg)) was determined using Morgan's extractant (Morgan, 1941), and are presented in Table 5.1. Mehlich-3 P extractant was also used to determine P levels (Mehlich, 1984). Soil pH (n=3) was determined using a pH probe (Mettler-Toledo Inlab Routine) and a 2:1 ratio of deionised water to soil. The optimal location for the 25 individual micro-plots in the field site was then determined by topography and slope, but most significantly, the area chosen had the soil nutrient analysis, pH levels and soil texture, which permitted biosolids application.

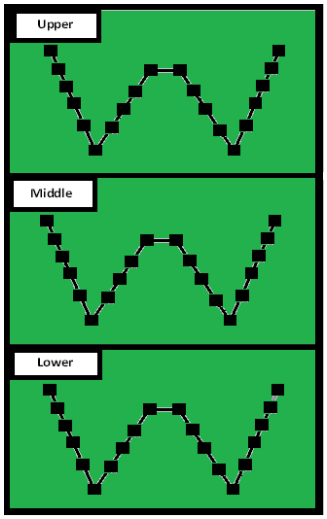


Figure 5.1. The "W" soil sample procedure outlined in the S.I. No 610 2010. This soil sample procedure was carried out for the Upper, Middle and Lower sections of the field

Table 5.1. Soil characteristics from the upper, middle and lower section of the 0.6 ha field site.

Position	pH	Morgan P	Mehlich 3-P	WEP	P index	K[a]	Mg[a]	LR[a]	Sand[b]	Silt[b]	Clay[b]	Textural[c]Class
		mg L^{-1}	mg L^{-1}	mg kg^{-1}		mg L^{-1}	mg L^{-1}	t/ha	%	%	%	
Upper	5.6	2.3	36.1	6.8	1.0	128.9	133.0	4.0	44%	36%	21%	Clay Loam
Middle	5.4	2.3	35.3	5.6	1.0	70.5	108.8	5.5	47%	36%	18%	Sandy Silt Loam
Lower	5.5	2.6	25.9	9.0	1.0	121.6	137.0	5.0	52%	30%	18%	Sandy Loam
Average	5.5	2.4	32.6	7.1	1.0	107.0	126.3	4.8	47.7	34	19	
Std. dev	0.1	0.2	4.6	1.4	0.0	26.0	12.5	0.6	4	3.5	1.7	

[a]Morgan's extractable potassium (K) and magnesium (Mg), lime requirement (LR)
[b]Brennan et al. (2012) [c]USDA classification system

5.3.2. Micro-plot installation and characterisation

Thirty grassland micro-plots, each 0.9 m in length and 0.4 m in width (0.36 m^2), 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 (Fig. 5.2). Micro-plots were orientated 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 (Fig. 5.2).

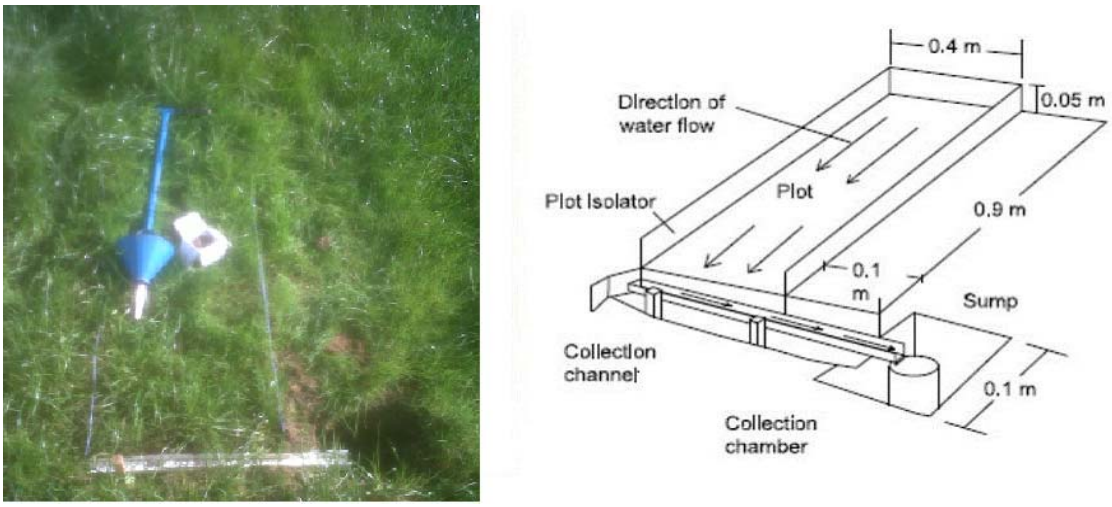


Figure 5.2. Picture of micro-plot fitted with runoff off collection channel and micro-plot set up.

For textural analysis, each micro-plot was tested at before start of experiment (t_0) for particle size distribution (% sand/silt/clay) using the hydrometer method (ASTM D422, 2002). Results of analyses are presented in Table 5.2. Soil nutrient status of each micro-plot was taken at t_0 and analysed for soil pH, Mehlich 3-P, Pm, K, Mg, water extractable P (WEP), organic matter (OM) and lime requirement (LR) (Table 5.2). In addition, composited soil samples were oven dried and grinded to 2 mm before being sent to ALS Environmental Global, Co. Dublin, Ireland at t_0 for metal content (Cu, Ni, Pb, Zn, Cd, Cr) by Inductively Couples Plasma Optical Emission Spectrometry (ICP-OES) (MEWAM, 1992), following aqua-regia digestion (MEWAM, 1986) (Tables 5.3). Soil nutrient and metal status analysis was also repeated immediately at the end of the experiment (t_{360}) (Tables 5.2 and 5.3). Background checks were performed on the soil microbial status (TC and FC) (Table 5.4) at t_0 and t_{360} by taking composite soil samples from the four corners outside the micro-plots (top left, top right, bottom left, bottom right). Total coliforms were tested in accordance with ISO 4832 (ISO, 2006) at both t_0 and FC were tested in accordance with ISO 16649-2 (ISO, 2001) at t_0 and ISO 4831 (ISO, 2006) at t_{360}.

Table 5.2.Average topographical and soil characteristics for the 30 individual micro-plots pooled together as per treatment applied, on the day before experiment (t_0) and immediately after the experiment ended (t_{360})

Treatment	Slope	pH_0/pH_{360}	WEP_0/WEP_{360}	Morgans P_0/P_{360}	Mehlich 3-P_0/P_{360}	K_0/K_{360}[a]	Mg_0/Mg_{360}[a]	LR_0/LR_{360}[a]	Sand[b]	Silt[b]	Clay[b]	Textural class[C]	BD
	%		mg kg^{-1}	mg L^{-1}	mg L^{-1}	mg L^{-1}	mg L^{-1}	t/ha	%	%	%		g/cm3
ADUK	2.89	5.94/5.90	7.10/5.9	3.60/5.57	38.0/37.1	94.94/60.78	147.13/147.80	2.70/3.00	45.70	39.49	14.82	Loam	1.3
TD	3.69	5.90/5.90	9.25/7.5	4.80/6.79	47.4/41.9	66.08/55.66	156.75/164.00	2.30/2.70	47.41	37.63	14.97	Loam	1.3
LS	2.84	5.90/6.25	6.60/5.4	3.82/6.24	38.3/32.7	58.20/52.12	136.47/146.40	2.60/1.00	48.74	36.58	14.69	Loam	1.3
ADIRE	2.87	5.96/5.93	7.7/6.1	4.32/6.11	41.4/35.7	78.39/55.74	152.68/147.40	2.40/2.70	48.17	36.55	15.28	Loam	1.4
SOIL	3.53	5.99/5.96	8.6/6.9	4.71/5.59	46.8/39.2	65.95/54.30	149.49/149.60	2.80/2.90	45.52	39.43	15.05	Loam	1.4
DCS	2.73	5.81/6.10	2.86/1.63	5.00/9.13	31.93/-	62.40/208.42	84.20/167.17	3.30/1.60	50.00	29.20	20.80	Loam	1.4

[a] Morgan's extractable potassium (K) and magnesium (Mg), lime requirement (LR) and Organic Matter (OM)

[b]ASTM D422. (2002).

[c]USDA classification system

Table 5.3. Average soil metals concentration of copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), cadmium (Cd), chromium (Cr) before start of experiment (t_0) and after the experiment (t_{360})

Treatment	Cd_0/Cd_{360}	Cr_0/Cr_{360}	Cu_0/Cu_{360}	Pb_0/Pb_{360}	$Ni_0/_{360}$	Zn_0/Zn_{360}
	---mg kg^{-1}---					
ADUK	<0.20/0.54	11.8/13.8	8.12/6.74	15.5/27.2	7.14/9	35.2/29.8
TD	<0.2/0.56	11.5/14.4	9.54/7.8	16.12/25	6.86/9.42	33.2/31.2
LS	<0.2/0.54	11.6/13.8	7.8/7.4	15/22	7.2/8.96	34.6/27.6
ADIRE	<0.2/0.54	12/14.4	8.42/7.34	16/21.8	7.66/9.4	36/30
SOIL	<0.2/0.56	11.8/14.4	8.62/7.16	17.22/24.4	7.28/9.34	35.2/31.2

5.3.3. Biosolids characterisation

Three types of biosolids were examined in this study: two types of AD sludge, one sourced from a WWTP in Ireland (ADIRE) and another used in an EU-funded FP7 project (END-O-SLUDG, 2014) (ADUK); TD and LS biosolids (Fig. 2.2, 2.3, 2.4). With the exception of ADUK (Fig. 5.3), all biosolids were sourced from the same WWTP in Ireland. As the Irish WWTP only employed two methods to treat sludge (anaerobic digestion and thermal drying), an untreated, dewatered sewage sludge cake was also collected from the same WWTP, so that it could be manually lime treated as described in Chapter 4. The treated sludge and the dewatered sludge cake were collected in sealed, 50 L-capacity plastic storage boxes and transported to Teagasc, Environment Research Centre, Johnstown Castle, Co Wexford, South East Ireland, where they were labelled and stored at 4^{o}C. The treated sludge samples (each at n=3) were tested for (Brookside Laboratories Inc, Ohio, USA): DM, total Kjeldahl nitrogen (TKN), nitrite (NO_2-N), NH_4-N, organic-N, total P (TP), P as phosphorus pentoxide (P_2O_5), K, K as potassium oxide (K_2O), pH, and metal content (Cu, Ni, Pb, Zn, Cd, Cr, Hg) (Table 5.5). Water extractable P was also tested after Kleinman et al. (2007) (Table 5.5). In addition, the biosolid samples (each at n=3) were also tested for TC and FC using the same methods as for soil (Table 5.4).

Table 5.4. The average total and faecal coliforms (±std. dev.) for soil, biosolids and DCS on the day before experiment (t_0) and after the experiment (t_{360}). Standard deviation in brackets.

Microbe	ADUK	TD	LS	ADIRE	SLURRY	Soil
Presumptive Coliforms (cfu g^{-1}) (t0)	$<1.0 \times 10^7$	$<1.0 \times 10^7$	$<1.0 \times 10^7$	$<1.0 \times 10^7$	5.43×10^4 (6.34×10^3)	$<1.0 \times 10^7$
ß-Glucuronidase + E. coli (cfu g^{-1}) <100 (t0)	6.5×10^3 (3.6×10^3)	$<1.0 \times 10^2$	$<1.0 \times 10^2$	$<1.0 \times 10^2$	1.10×10^3	$<1.0 \times 10^2$
Total coliform (Product) (t360)	7.4×10^2 (4.5×10^2)	6.3×10^1 ($4.5 \times 10^{1)}$	1.3×10^1 (4.7×10^0)	5.0×10^1 ($5.0 \times 10^{0)}$	-	1.3×10^3 (6.9×10^2)
Faecal Coliforms (MPN) (t360)	1.7×10^1 (2.1×10^1)	1.9×10^0 (1.7×10^0)	$<3.0 \times 10^{-1}$ (0)	2.3×10^0 (0)	-	7.7×10^0 (4.9×10^0)

(t_0) - Test performed by Tellab, Co. Carlow, before experiment
(t_{360}) - Test performed CLS Labs, Co. Galway, end of experiment

Figure 5.3. Anaerobically digested biosolid source from END-O-SLUDGE, 2014 (ADUK)

5.3.4. Slurry Characterisation

Dairy cattle slurry was collected from the dairy farm unit at the Teagasc, Environmental Research Centre, Johnstown Castle. 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 (Table 5.5). The DCS (each at n=3) were tested for (Southern Scientific Ireland, Co. Kerry, Ireland): DM, N (Kjeldahl, 1883), P and K and metal content (Cu, Ni, Pb, Zn, Cd and Cr) (Table 5.5). 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 (Table 5.4).

Table 5.5. Nutrient and metal characteristics of the biosolids and slurry

Treatment	DM	Total N	Total P	Total K	pH	WEP (dry)	OM	Cu	Ni	Pb	Zn	Cd	Cr	Hg	NO_3-N	NH_4-N	Organic - N	P_2O_5[a]	K_2O[b]
	%	----------mg kg $^{-1}$----------				g kg^{-1}	%	----------mg kg $^{-1}$----------											
ADUK	25.1	43216.3	23512.1	2145.8	7.8	15.5	-	287.0	140.2	115.3	682.8	1.84	31.46	0.0	3979.4	3846.6	39369.7	53875.6	2584.9
	(0.1)	(1670.8)	(273.9)	(39.8)	(0.0)	(7.6)	-	(4.0)	(1.5)	(0.8)	(3.0)	(0.0)	(0.5)	(0.0)	(14.0)	(293.7)	(1961.8)	(627.5)	(47.9)
LS	34.2	17620.5	3938.7	2229.5	12.6	8.9	28.4	111.7	12.2	10.7	218.5	0.4	8.1	0.0	2922.3	449.2	17171.3	9137.5	2686.0
	(0.2)	(395.5)	(396.1)	(43.8)	(0.0)	(0.3)	(0.5)	(11.4)	(0.3)	(1.0)	(20.1)	(0.0)	(0.3)	(0.0)	(13.1)	(28.6)	(395.1)	(790.2)	(52.3)
TD	87.10	51446.0	17114.4	2055.1	6.9	492.7	79.5	504.8	19.6	62.2	876.9	1.0	22.1	0.4	1148.2	573.3	50872.7	39215.9	2475.6
	(0.07)	(2897.3)	(186.9)	(50.7)	(0.0)	(25.6)	(2.0)	(18.8)	(2.0)	(0.6)	(5.5)	(0.0)	(0.06)	(0.5)	(1.0)	(32.1)	(2875.8)	(428.3)	(61.2)
ADIRE	23.6	54577.8	25185.7	2198.7	8.1	302.2	72	756.4	26.3	91.6	1109.6	1.5	31.7	0	4234.6	3428	51149.8	57710.5	2648.6
	(0.2)	(1530.3)	(609.0)	(78.4)	(0.0)	(1.0)	(1.0)	(20.7)	(1.3)	(3.1)	(21.6)	0.0	(1.9)	0.0	(38.1)	(239.7)	(1775.5)	(1395.4)	(94.5)
DCS	8.35	2.2	0.5	4.38	8.2	93.3	-	3.9	0.44	<0.25	14.3	<0.25	0.71	-	-	-	-	-	-
	(0.2)	(0.2)	(0)	(0.4)	(0)	(3.34)	-	0	(0.3)	0	(0.2)	0	(0.62)	-	-	-	-	-	-

[a] P_2O_5 - Phosphorus pentoxide

[b] K_2O - Potassium oxide

(Standard deviation in brackets)

5.3.5. Rainfall event simulation and application

One Amsterdam drip-type rainfall simulator, as described in Chapter 3, was used to provide rainfall in this study (Fig. 3.2). The simulator was calibrated to deliver a rainfall intensity of 11 mm hr^{-1}. Water samples, used in the rainfall simulations, were collected over the duration of the three rainfall events, and had average concentrations of: 0.07±0.0 mg NH_4-N L^{-1}, 3.81 ±0.02 mg NO_3-N L^{-1}, 3.80±0.02 mg total oxidised nitrogen (TON) L^{-1}, 0.01±0.00 mg dissolved reactive phosphorus (DRP) L^{-1}, 0.02±0.0 mg TP L^{-1}, 0.30±0.09 µg Cd L^{-1}, 0.38±0.07 µg Cr L^{-1}, 10.10±0.75 µg Cu L^{-1}, 0.65±0.46 µg Ni L^{-1}, 0.93±1.25 µg Pb L^{-1}, 78.91±6.67 µg Zn L^{-1}, 11.04±1.05 µg aluminium (Al) L^{-1}, 0.00±0.00 µg iron (Fe) $L^{-1}$and 9.95±0.05 µg manganese (Mn) L^{-1}.

The six treatments (four biosolids, 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). As metal content was not limiting in soil, DCS or biosolids application to the micro-plots was governed by the P content of the biosolids, and DCS and the P index of the soil. For comparable results, all micro-plots were classified into Index 2 P soil, which meant that all biosolids and DCS treatments were applied to all plots at a rate of 40 kg P ha^{-1} (Coulter et al., 2008). As a result of the P content and the DM of each individual biosolid, application rates per individual plot was of 96.6 g of TD, 242.2 g of ADIRE, 1063.3 g of LS, 243.9 g of ADUK biosolids were applied to each designated plot. The DCS was spread at 2880 g per individual plot.

Prior to application, grass on all plots was cut to 50 mm, 48 hr 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 (Fig. 5.4D) (Hoekstra et al., 2014). Biosolids were hand surface applied to each micro-plot. 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 (Fig. 5.4C). The DCS was applied in rows using a watering can to replicate normal trailing shoe application. The biosolids and DCS were then left 24 hr with the soil before RS1. The RS1 event occurred 24 hr after biosolids and DCS application, so as to demonstrate losses representative of a worst-case scenario. The second rainfall event (RS2) was two days (48 hr) after initial biosolids/DCS application, which was representative of current legislation, and the third (RS3) 15 days (360 hr) 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 sterilise them.

Figure 5.4. A) and B) show site set up, C) Quadrant used to apply biosolids evenly, D) Rainout shelters to excluded natural rainfall

5.3.6. 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 mL (t=0) was used to indicate time to runoff (TR), and was used for part of the microbial analysis. Samples for nutrient and metal analysis were collected every 10 min (t=10, T=20, T=30) from TR to allow for the flow weighted mean concentration (FWMC) to be calculated (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. The rainfall simulator was then switched off and a final sample (T=F) was collected to determine the final runoff ratio. This sample was also analysed for nutrient and metal content. Immediately after collection, all samples were stored in cool boxes with ice until they were returned to the laboratory for analysis. Fig. 5.5 shows the collection of water samples for microbes, nutrients and metal.

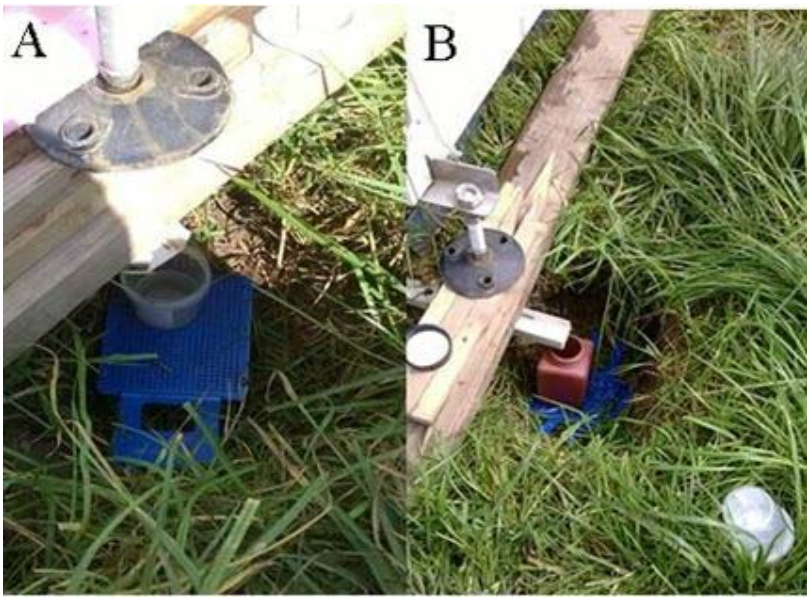


Figure 5.5. A) Sterile collection cups for Microbes, B) Collection cups for nutrients and metals

5.3.7. 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 DRP, NO_3-N, NO_2-N and NH_4-N using a nutrient analyser (Aquachem Labmedics Analytics, Thermo Clinical Labsystems, Finland). A second filtered sub-sample was analysed for total dissolved phosphorus (TDP) using acid persulphate. Unfiltered runoff water samples were analysed for TP with an acid persulphate digestion and total reactive phosphorus (TRP) using the Aquachem Analyser. Metal analysis

was tested on the filtered samples using inductively coupled plasma optical emission spectroscopy (ICP-OES). Particulate phosphorus was calculated by subtracting TDP from TP. The DRP was subtracted from the TDP to give the dissolved un-reactive phosphorus (DUP). All samples were tested in accordance with the Standard Methods (APHA, 2005).

5.3.8. Total and faecal coliform analysis

The two 50 ml runoff water samples from the start of rainfall simulation experiment and near the end were bulked together in one sterile collection pot in the laboratory. The water volume in the collection pot was then prepared by serial dilution using sterile water form a Millipore automatic sanitization module. For detection and enumeration of total and faecal coliforms, IDEXX Coilisure Quanti Tray/2000 method (IDEXX Laboratories, Westbrook, ME) was used to determine the most probable number (MPN) in each sample. Samples were incubated at 37±0.5°C degrees for 24 hr. All analyses were carried out in accordance with the standard methods (APHA, 2005).

5.3.9 Data analysis

The structure of the data set was a blocked one-way classification (treatments) with repeated measures over time (rainfall events (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. A large number of covariates were recorded, including measurements on the simulators and for each analysis; this set of covariates was screened for any effects that should be included in an analysis of covariance. The interpretation was conducted as a treatment by time factorial. Comparisons between means were made with compensation for multiple testing effects using the Tukey adjustment to p-values. Significant interactions were interpreted using simple effects before making mean comparisons. For

comparison of soil characteristics before and after the experiment, the relationship between the paired measurements, adjusted for treatment, was tested and, given a significant relationship, the difference between each pair of results was analysed by treatment. In some cases an intercept only model was fitted to determine if there had been an overall change across all treatments. Residual checks were made in all cases to ensure that the assumptions of the analyses were met.

5.4. Results

5.4.1. Nutrient losses in runoff

The average FWMC of TP, comprising DUP, PP and DRP, for all treatments and rainfall events is shown in Fig. 5.6. 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 losses for DCS during RS1 was 7.0 mg L^{-1} and 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.

Particulate P losses comprised only 14% and 32 % of TD biosolids and DCS, respectively, due to the high proportion of DRP losses. However, when only considering the PP losses, DCS plots for RS1 and RS2 had significantly higher PP losses ($p < 0.05$) than all other measurements, which were statistically indistinguishable.

The average FWMC of TN across all treatments is shown in Fig. 5.6. There was a significant interaction between treatment and the rainfall simulation for NH_4-N. The application of all biosolids treatments and DCS increased the average FWMC of NH_4-N for RS1 compared to the study control, and while there was a downward 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 NH_4-N for all biosolids treatments in RS1 (15.3 mg L^{-1}). Thermally dried and ADIRE treatments had the next highest FWMCs of NH_4-N, but these were not significantly different from each other or from the LS runoff during RS1. While total losses from DCS were greatest, they were significantly different only from LS ($p=.005$) and the control ($p<0.001$). The median FWMC of NH_4-N in RS1 for DCS was 17.4 mg L^{-1}. The addition of biosolids and DCS had no effect on FWMCs of NO_3-N in runoff, except for LS biosolids, which significantly reduced, relative to the control, the incidental losses of NO_3-N during RS1 and RS2 ($p<0.001$), before it increased during RS3. Nitrite losses were negligible in all treatments, with only exception being the DCS.

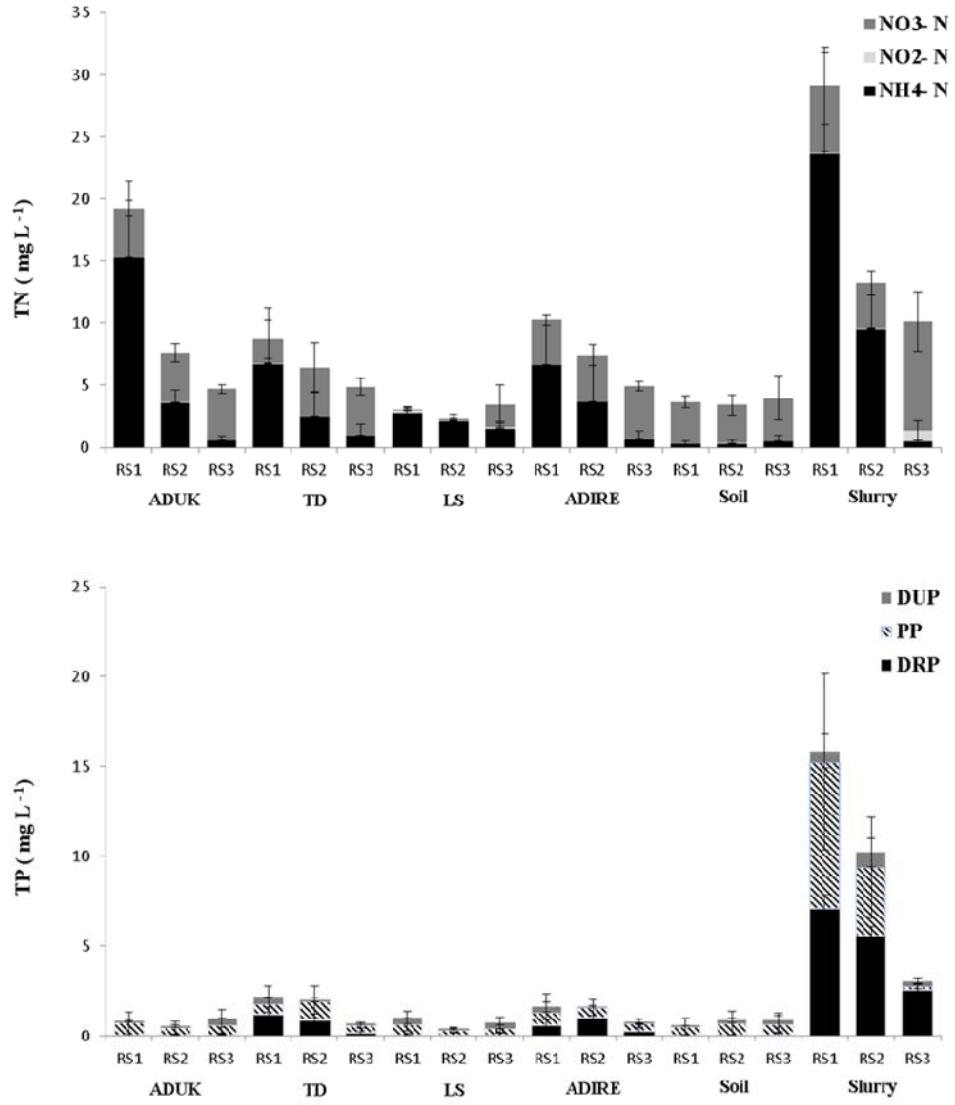


Figure 5.6. Flow weighted mean concentrations of phosphorus (top) and nitrogen (bottom) in the runoff over three successive rainfall events at 24 hr (RS1), 48 hr (RS2) and 360 hr (RS3) after application to grassland.

5.4.2. Metal losses in runoff

The average FWMC of metals (Cu, Ni, Pb, Zn, Cd, Cr) in runoff are shown in Fig. 5.7. All runoff samples were below their respective drinking water standards intended for human

consumption (S.I. No. 122 of 2014). There was no difference in the FWMCs in surface runoff of Cd and Cr of any treatment compared to the study control, except for DCS. Cadmium losses for DCS during RS1 were significantly lower than other treatments, but were significantly higher during RS3. 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 compared to control. The highest median FWMC for Pb (1.5 µg L^{-1}) was measured during RS3 for the DCS and the second highest was 0.82 µg 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 µg L^{-1}) was during RS1 for DCS-amended plots, but there were no significant differences across treatments or events.

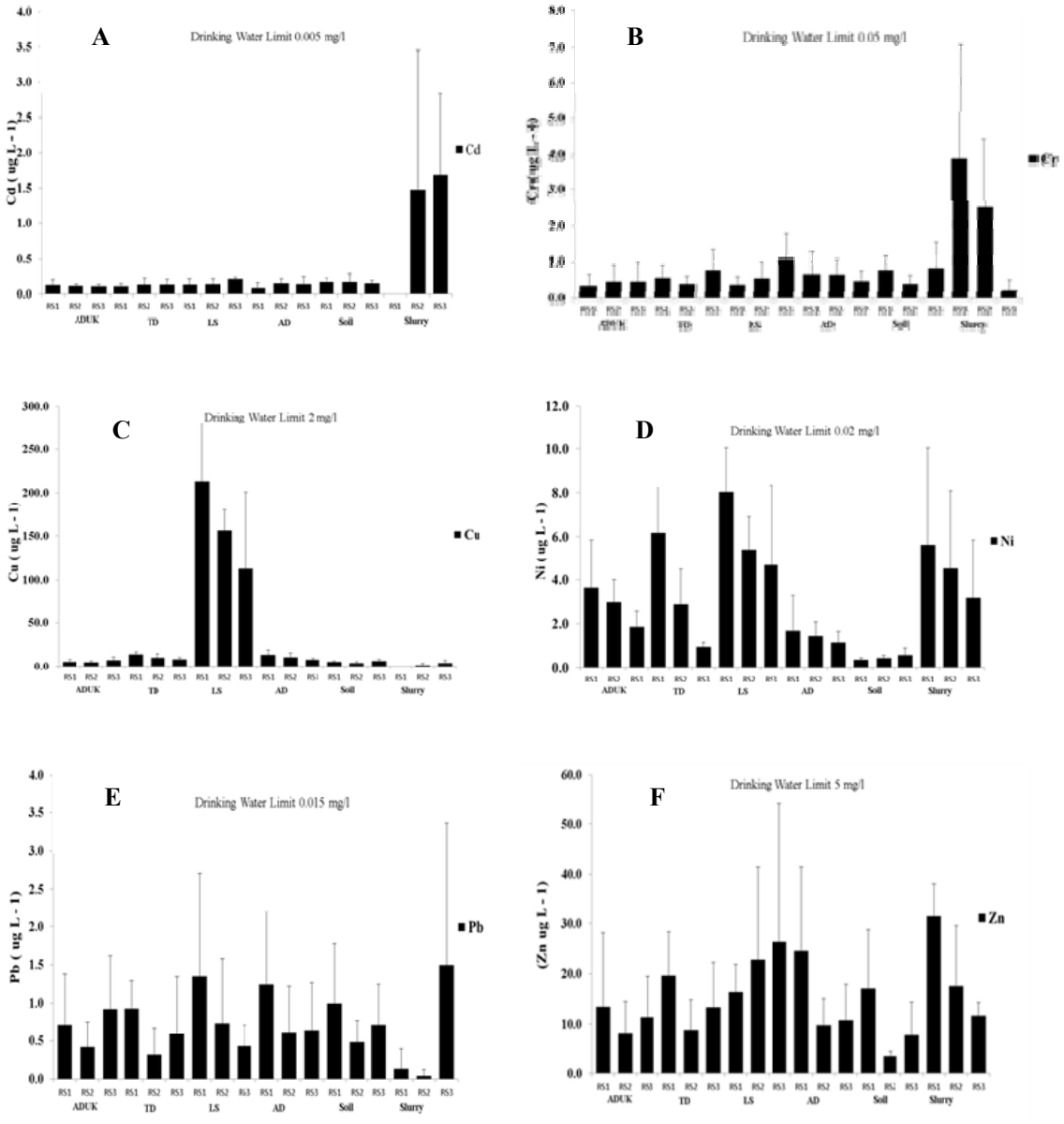


Figure 5.7. Flow weighted mean concentrations of cadmium (A), chromium (B), copper (C), nickel (D), lead (E) and zinc (F) in the runoff over three successive rainfall events at 24 hr. (RS1), 48 hr. (RS2) and 360 hr. (RS3) after application to grassland

5.4.3. Microbial losses in runoff (Total and faecal coliform)

The average losses of TC and FC are shown in Fig. 5.8. 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: 7.1×10^3 MPN per 100 ml during RS1 and RS2. For TC losses there was an interaction between treatment and event ($p=0.01$), but only the highest and lowest event outcomes were significantly different. While median losses from the TD-amended plots increased with successive rainfall events from 1.9×10^5 MPN per 100 ml during RS1 to 1.0×10^6 MPN per 100 ml during RS3, there were no significant differences within treatments. There was no evidence of interaction between treatment and event for TC, so it is impossible make inference about the factors separately. There was no change from RS1 to RS2, but there was a decrease from RS2 to RS3 ($p<0.0001$) from a median of 7.6×10^1 MPN per 100 ml during RS1 to 5.4×10^1 MPN per 100 ml during RS3. Overall losses from DCS (3.1×10^2 MPN) were greatest and significantly greater than LS, ADIRE and the control. ADUK losses (1.7×10^2 MPN) were not statistically different from DCS, but were significantly greater than the control ($p=0.009$). The highest median count of TC and FC measured in LS biosolids-amended plots was 5.6×10^5 and 1.5×10^1 MPN per 100 ml, respectively. The highest median loss of TC for DCS-amended plots was 1.5×10^5 MPN per 100 ml.

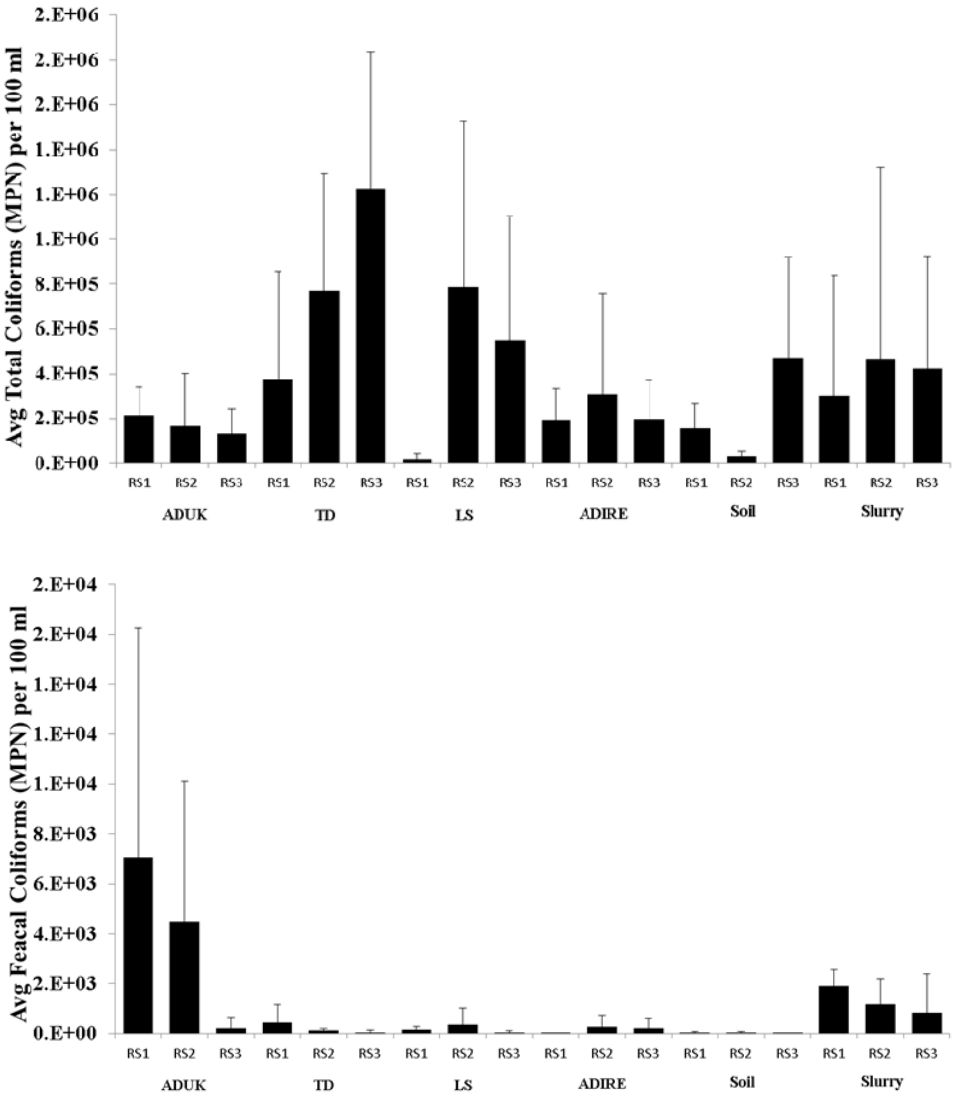


Figure 5.8. Total coliforms (top) and faecal coliforms (bottom) in the runoff per 100ml over three successive rainfall events at 24 hr (RS1), 48 hrs (RS2) and 360 hr (RS3) after application to grassland

5.4.4. Soil test P, Mehlich-3 P, K, LR, pH and metal

Morgan's P, Mehlich-3 P, WEP, Mg, K, pH, LR and metals results from analysis of plots before (t_0) and at the end of the experiment (t_{360}) are presented in Tables 5.2 and 5.3. Average P_m (3.6 to 4.8 mg L^{-1}), Mehlich-3 P (38.0 to 47.4 mg L^{-1}), K (58.2 to 94.94 mg L^{-1}), LR (2.3 to 2.6 t ha^{-1}) and pH (5.90 to 5.99) across all plots before application of treatments were similar. At the end of the experiment, P_m increased across all treatments ($p<0.0001$), with no significant differences between treatments. The Pm of the control plots also increased by 18%. Mehlich-3 P decreased across all treatments ($p=0.0001$), with no significant differences between treatments. Potassium concentrations showed no significant decrease for LS and TD treatments, while the greatest reduction was in the ADUK plots (35%) and the lowest in the lime-amended plots (10%). Magnesium showed no significant changes over the duration of the experiment. Lime requirement increased in the ADUK, TD, control plots and ADIRE by 11%, 10% 8% and 3.8%, respectively, but reduced by 56% in the lime-amended plots.

Average metal results across all treatments before the start of the experiment were similar. At the end of the experiment, Cd and Cr ($p<0.0001$) increased across all treatments, while Cu showed a significant decrease only for TD. Lead ($p=<0.0001$) and Ni ($p<0.0001$) increased across all treatments, but there were no significant differences between treatments. The average increase for Pb was 50.8% and was 27.6% for Ni. Zinc decreased ($p<0.0001$) across all treatments, but there was no difference between treatments.

5.5 Discussion

5.5.1. Incidental nutrient losses for all rainfall events

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 (MAC) 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 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 is similar to several studies that have examined runoff of nutrients resulting from the land application of different types of biosolids and DCS (Rostagno et al., 2001; Penn et al., 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 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; Elliot et al., 2005; Kleinman et al., 2007). Thermally dried and ADIRE biosolids, which also had high WEPs (Table 5.5), had the highest losses of dissolved P from their respective plots.

All biosolids treatments had elevated FWMCs of NH_4-N in runoff compared to the study control across all rainfall simulations, whereas the study control and biosolids-amended plots had the same NO_3-N concentrations. Ammonium can be volatilised (or rapidly mobilised by runoff and leaching) after organic matter spreading (Quilbé et al., 2005). ADUK biosolids, which had the highest initial NH_4-N concentration in the biosolids at the time of application (3846 mg kg^{-1} DM), also had the highest FWMC of NH_4-N in runoff compared to biosolids treatments during RS1. Similar trends were noted for the ADIRE and LS biosolids. However, the initial concentration of NH_4-N in TD biosolids before application (573 mg kg^{-1}; Table 5.5) was lower than the ADIRE biosolids (3428 mg kg^{-1}; Table 5.5), but had similar losses of

NH_4-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 easier 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. Dairy cattle slurry had the highest FWMC of NH_4-N and DRP. A possible reason for this is that DCS had a DM of 8%, and was highly mobile following an episodic rainfall event. This study shows that biosolids, although having a higher DM than DCS, are not as easily mobilised.

5.5.2. Incidental metal losses for all rainfall events

The concentrations of metals in runoff were below drinking water standards intended for human consumption (S.I. No. 122 of 2014). Similar results have been reported for several runoff studies using different types of biosolids at higher application rates than the current study (Joshua et al., 1998; Dowdy et al., 1991; 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 waterbodies. 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 code of good practices (Fehily Timoney and Company, 1999).

While there was generally low FWMC of metals over all rainfall simulations, the LS biosolids-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 consists of larger sized particles of a more compact consistency, time to runoff increased (results not shown), giving these metals more contact time to dissolve and 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 FWMC of Cd and Cr in DCS-amended plots were higher than any of the biosolids plots, with peak concentrations of 1.68 ug L^{-1} during RS3 for Cd and 3.89 ug L^{-1} during RS1 for Cr, respectively. However, even at these concentrations, they were still well below drinking water standards.

5.5.3. Incidental pathogen losses for all rainfall events

Understanding the environmental persistence and fate of enteric pathogens introduction 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 biosolids-derived and other organic amendment pathogens 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 to survival in the soil environment, and pathogens that are land applied from biosolids and DCS are influenced by climatic and agronomic variables (Lang et al., 2003). When biosolids and DCS are incorporated into the soil, pathogen 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 have been reviewed by Erickson et al. (2014). However, when

biosolids and DCS are surface applied, as in the current study, desiccation and ultraviolet light are the key factors in the decay of pathogens (Lu et al., 2012). Desiccation of pathogens is influenced by the soil, biosolids and DCS moisture content. In the current study, soil moisture remained consent 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 decayed by RS3, which was most likely due to desiccation of pathogens 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 $>1 \times 10^3$ MPN g^{-1} (Fehily Timoney and Company., 1999), and, as a result, were equivalent to Class B microbial matter under the US EPA Part 503 regulations (USEPA, 1993), which allows detectible levels of FC up to 2×10^6 MPN g^{-1} DS. All the Irish biosolids were some 10-fold below the Class A Irish standard. Dairy cattle slurry had high FC losses compared to the Irish biosolids, suggesting that pathogen losses to surface water bodies following land application of untreated organic fertiliser may be a concern in Ireland.

It is important to evaluate the risks arising from the application of biosolids to land relative to other common agricultural practices such as the land application of animal waste (Vinten et al., 2010), 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 1.2×10^5

CFU per 100 ml, two days after application, and 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 (4.0×10^3 CFU per 100 ml). This was also observed in the current study, as the DCS had the second highest FC during RS1 and RS2, but was the highest by RS3, showing that FC survive for a longer period in DCS compared to biosolids, and may result in losses of pathogen to waterbodies for a longer period following application. Moreover, Payment et al. (2001) found that the pathogen concentration was lower in untreated sludge (3×10^2 to 6×10^2 cfu g^{-1}) compared to fresh and stored cattle slurries (2.6×10^8 to 7.5×10^4 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 provided no buffering capacity to the runoff samples, and overland flow was not sampled at delivery end of the transfer continuum, so the bacterial results represent a worst case scenario.

While this study and many others focus on the TC group as an indicator of the presence of pathogens, the drawback of relying on them is that it they are a poor indicator for the presence of viruses and parasitic protozoa, which may survive for much longer periods (NHMRC, 2003). However, due to the lack of well-developed methods for the detection and enumeration of these pathogens (Sidhu et al., 2009), the use of indicator organisms allows for the limitation of potential contaminating effects.

5.5.4. Soil characteristics before and after experiment

In the current study, differences in soil nutrient concentration following amendments were observed. The application of all biosolids increased the P_m in all amended plots from an Index 2 soil to an Index 3. Whilst the P_m of the control plots also increased from an Index 2 soil to an Index 3 soil, the increase was less than half the increase of the nearest biosolids amendment (ADIRE). Lime stabilised biosolids had the greatest increase in P_m, and this may have been a result of the evaluated pH in the soil as liming improves the availability of soil P. This result also shows that although LS biosolids are low in nutrient content, they can be applied for their pH adjusting characteristics and, as a result, may enhance nutrient availability to soil and plants.

This study also investigated the accumulation of metals before and after the experiment. Results showed that while there was an increase for some metals, none exceed the recommended guideline limits for soil set out in code of good practices (Fehily Timoney and Company, 1999). It should be noted, however, that the current study encompassed a single application of biosolids, and that concerns have been raised about the accumulation of metals in both soil and crops after repeated applications of biosolids (McBride, 2003; Bai et al., 2010). However, in Ireland, the application rate of biosolids to land is governed by legislation and whilst best practice is followed, problems in terms of metal or nutrient build-up will be avoided.

5.6. Conclusion

The results of this plot-scale study showed that there were elevated losses of nutrients and faecal coliforms from biosolids-amended plots compared to unamended plots. However, nutrient and pathogen losses were higher from DCS-amended plots. Metal concentrations in runoff were below their respective drinking water limits for human consumption for both biosolids and DCS. The runoff concentrations measured in this study represented a 'worst case' scenario for potential losses, as further buffering may be possible further down the transfer continuum. This study was conducted at micro-plot scale, but the results should be verified at field-scale. In addition, future work should also be carried out to assess 'emerging' organic pharmaceutical contaminants that may be present in biosolids. Notwithstanding these caveats, these results are significant as they show that issues surrounding the reuse of a resource, mainly concerning fears over elevated losses of nutrients, metals and pathogens, may be unfounded.

5.7. Summary

The results of this study, while indicative only, allow comparison to be made between amendments when applied at the same rate. The findings of this study need to be verified at full field scale. In addition, further research is required to determine their effect on the physical and chemical properties of soil.

Chapter 6 - CONCLUSIONS AND RECOMMENDATIONS

6.1. Overview

The objective of this study was to determine the impact of land application of three types of biosolids and compare them to another commonly spread organic fertiliser, dairy cattle slurry. 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. The main conclusions and recommendations arising from this study are now presented.

6.2. Conclusions

1. Losses from biosolids-amended plots were higher than the study control (soil only) plots, and followed a general trend of highest losses occurring during the first rainfall event and reduced losses in the subsequent events.

2. With the exception of total coliforms and some metal parameters, the greatest losses were from the dairy cattle slurry-amended plots. This means that biosolids do not pose a greater risk in terms of runoff losses following land application.

3. Preliminary tests examining ways to incorporate lime into sewage sludge suggested that it may be difficult to satisfy the Code of Good Practice standards, which state that the quantity of lime added to sewage sludge must increase the pH of the lime-sludge mix to ≥12 and the temperature to 70˚C for 30 min, or increase the pH above 12 for 72 hr and maintain a temperature of ≥ 52˚C for 12 hr. While the pH criterion was

achieved in our preliminary studies, there was difficulty in achieving the temperature criterion. This finding may have implications for the quality of biosolids produced at field-scale from lime stabilisation.

6.3 Recommendations for future work

1. There is currently a knowledge gap concerning the effectiveness of lime stabilisation in adhering to the pH and temperature requirements of the Codes of Good Practice. There is a need for research into the lime stabilising process and its effectiveness to minimise food safety concerns.

2. Biosolids spread at the maximum application rate on grassland had no adverse impact on surface water quality compared to dairy cattle slurry in terms of nutrients and metal losses in surface runoff. However, further testing in a larger field-scale experiment will be needed verify the findings of this study.

3. This study did not examine the surface runoff for the presence of emerging contaminants, such as pharmaceuticals or personal care products. While the findings of this study suggest that there are no issues in runoff of nutrients, metals and microbial matter (in comparison to dairy cattle slurry), the surface runoff water from the biosolids-amended micro-plots of the current study must be tested for these, and other, emerging contaminants. At the time of writing, surface runoff water samples from our micro-plots are awaiting testing for a selection of emerging contaminants.

4. Gaseous emission studies following the land application of organic and chemical fertilisers are commonly conducted. However, little work has been conducted examining gaseous emissions following biosolids application to land. Work is needed to address this knowledge gap.

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