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# Nutrient, metal and microbial loss in surface runoff following treated sludge and dairy cattle slurry application to an Irish grassland soil

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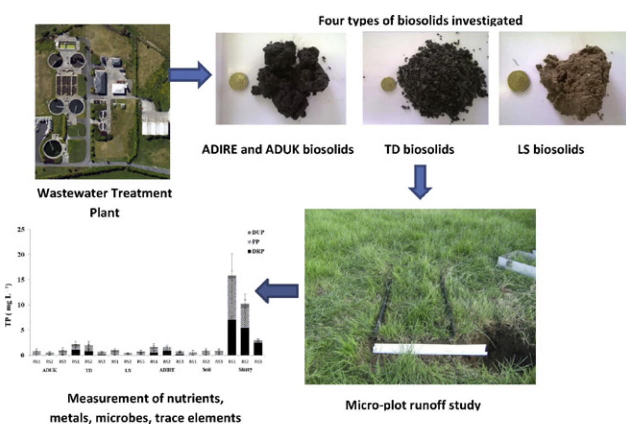
## HIGHLIGHTS

- This study investigated surface runoff of contaminants from biosolids in field plots.
- Contaminants investigated were nutrients, metals, microbes and trace elements.
- Compared to slurry, biosolids do not pose a greater risk of contaminant losses.
- Fears concerning contaminant losses from land applied biosolids may be unfounded.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Treated municipal sewage sludge ("biosolids") and dairy cattle slurry (DCS) may be applied to agricultural land as an organic fertiliser. This study investigates losses of nutrients in runoff water (nitrogen (N) and phosphorus (P)), metals (copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), cadmium (Cd), chromium (Cr)), and microbial indicators of pollution (total and faecal coliforms) arising from the land application of four types of treated biosolids and DCS to field micro-plots at three time intervals (24, 48, 360 h) after application. Losses from biosolids-amended plots or DCS-amended plots 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 and faecal coliforms and some metals (Ni, 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 from DCS-amended plots were 5 and 11.2 mg $L^{-1}$, respectively, which were in excess of the losses from the biosolids plots. When compared with slurry treatments, for the parameters monitored biosolids generally do not pose a greater risk in terms of losses along the runoff pathway. This finding has important policy implications, as it shows that concern related to the reuse of biosolids as a soil fertiliser, mainly related to contaminant losses upon land application, may be unfounded.



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## 1. Introduction

In the European Union (EU), implementation of directives and other legislative measures in recent decades concerning the collection, treatment and discharge of wastewater, as well as technological advances in the upgrading and development of wastewater treatment plants (WWTPs) (Robinson et al., 2012), has resulted in a rise in the number of households connected to sewers, increasing the loadings on WWTPs (EC., European Commission, 2014). Production of untreated sewage sludge across the EU has increased from 5.5 million tonnes of dry matter (DM) in 1992 to an estimated 10 million tonnes in 2010 (EUROSTAT. EC, European Commission, 2014), with production further expected to increase to 13 million tonnes in all EU member states by 2020 (EC., European Commission, 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 accelerated by legislation such as the Landfill Directive 1999/31/EC (EC., European Commission, 1999), the Urban Wastewater Treatment Directive 91/271/EEC (EEC., European Economic Community, 1991), the Waste Framework Directive 2008/98/EC (EC., European Commission, 2008), and the Renewable Energy Directive 2009/28/EC (EC., European Commission, 2009). This has prompted those involved in sewage sludge management to find alternative uses for it, 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 and Rowley, 2009; Healy et al., 2015). However, before this can occur, it must be treated to prevent harmful effects on soil, vegetation, animals and humans (EC., European Commission, 2014). Chemical, thermal or biological treatments, which may include composting (USEPA., US Environmental Protection Agency, 2002), aerobic and anaerobic digestion (USEPA., US Environmental Protection Agency, 2006a), thermal drying (USEPA., US Environmental Protection Agency, 2006b), or lime stabilisation (USEPA., US Environmental Protection Agency, 2000), produces a stabilised organic material frequently referred to as ‘biosolids’. The term biosolids was formally adopted in 1991 by the Name Change Task Force of the Water Environment Federation (WEF., Water Environment Federation, 2005) to differentiate raw, untreated sewage sludge from treated and tested sewage sludge that can legally be as a soil amendment and fertiliser.

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

There are many potential problems associated with the land application of biosolids, and these have been reviewed by Lu et al. (2012) and Singh and Agrawal (2008), amongst others. Nutrient losses in runoff are affected not only by biosolid type, but also application rate. In the EU, land application of biosolids is based on the pH, metal and nutrient content of the soil and the nutrient and metal content of the biosolids (Fehily Timoney and Company, 1999). Frequently, the phosphorus (P) content of the biosolids becomes the limiting factor in determining the land application rate (Lucid et al., 2013). In the USA, the application of biosolids to land is governed by the standard for the use or disposal of sewage sludge (USEPA., US Environmental Protection Agency, 1993) and as a result, the rate of application of biosolids to land are applied based on an estimate of crop nitrogen (N) need and biosolids N availability (Lu et al., 2012), and is not based on a soil test (McDonald and Wall, 2011). However, due to concerns about the effects of repeated manure or biosolids applications on the soil and the risk of P loss to surface water, some states (e.g. Maryland) have introduced regulations based on the P content of the biosolids (Lu et al., 2012).

Losses of nutrients to surface or subsurface waters bodies originates in two ways: as chronic (long-term, due to the build-up of nutrients in soil), or as incidental (short-term losses within 48 h of application) losses following episodic rainfall events soon after land application of a fertiliser or amendment (Brennan et al., 2012). Such losses to a surface waterbody occur via direct discharges, surface and near surface pathways, and/or groundwater discharge, where there is a hydrological transfer continuum between a nutrient source (chronic or incidental) and surface water receptor (Wall et al., 2011). Losses of P have been reported by Lucid et al. (2013, 2014) following the application of thermally dried (TD), lime stabilised (LS) and anaerobically digested (AD) biosolids. Increased N losses have also been reported following biosolids application to land. For example, Ojeda et al. (2006) reported elevated concentrations of ammonium ($NH_4$–N) and nitrate ($NO_3$–N) in surface waters following the application of TD and composted biosolids at rates of 10 t DM $ha^{-1}$. Quilbe et al. (2005) measured elevated runoff $NH_4$–N concentrations following the spreading of AD biosolids applied at 7.5 DM $ha^{-1}$, whereas LS biosolids had no significant effect on such concentrations in runoff when applied at the same rate.

Although many studies have not reported elevated metal concentrations in runoff following the application of various types of biosolids (Joshua et al., 1998; Dowdy et al., 1991; Eldridge et al., 2009; Lucid et al., 2013), there is a dearth of data comparing the impact of several types of biosolids, applied during the same application, on surface runoff of metals. In addition, concerns have been raised about the accumulation of heavy metals in both soil and crops after repeated applications of biosolids (McBride, 2003; Bai et al., 2010) and the migration of metals from the soil profile to surface and subsurface waters (Lu et al., 2012).

Other concerns associated with the land spreading of biosolids have focused around human enteric pathogens found in biosolids, as inactivation of pathogens is difficult to achieve (Sidhu and Toze, 2009). Typically, the densities of pathogens are reduced by two to three orders of magnitude by the wastewater treatment and biosolids processing (Apedaile, 2001). Whilst these reductions are significant, appreciable numbers of pathogens survive, which may subsequently re-grow to hazardous levels when exposed to favourable environmental conditions (Zaleski et al., 2005), especially during storage (Iranpour and Cox, 2006). Pathogen survival is evidenced by the survival of faecal coliforms (FC) as indicators for the possible presence of microbial pathogens. The use of indicator organisms allows for the limitation of potential contaminating effects (Sidhu and Toze, 2009).

Studies have shown that elements of pathogen population may exhibit enhanced survival due to advantageous physiological properties or colonisation of more favourable sites (Brennan et al., 2012). However, as the soil environment is very hostile to the survival of pathogens, their survival time, following the land application, is 2 to 4 months (Brennan et al., 2012). Consequently, pathogens are more likely to be transported to watercourses in incidental rainfall events soon after land application. Studies examining the transport of pathogens in runoff following the application of biosolids have generally shown increased runoff of FC compared to control plots (Dunigan and Dick, 1980; Nelson and Choi, 2005; Wallace et al., 2014).

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 dairy cattle slurry (DCS) are influenced by climatic and agronomic variables (Lang et al., 2003).