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PRESENCE OF EMERGING CONTAMINANTS IN TREATED SLUDGES AND THEIR POTENTIAL IMPACT ON THE ENVIRONMENT

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Abstract

Landspreading of sewage sludge remains the most economical and practical means for its disposal, with some countries, such as Ireland, disposing up to 80% to land. Its nitrogen, phosphorus and metal content make it an effective fertiliser replacement. However, there are potentially serious issues associated with its use in agriculture. These range from impacts on the environment through surface losses of nutrients, metals, pharmaceuticals and personal care products (PPCPs), and emerging contaminants, as well as build-up of contaminants in the soil and transfer to the human food chain. This paper details the main findings of an Irish Environmental Protection Agency (EPA)-funded study which investigated these issues. This study characterised treated sewage sludge ('biosolids') from wastewater treatment plants employing different means of sludge treatment, examined surface losses of various physico-chemical parameters following land application, measured metal uptake by ryegrass, and modelled the potential impacts on human health.

Keywords

Biosolids; sewage sludge; land application; human health; emerging contaminants.

Introduction

In 2015, more than 4 million tonnes (dry solids) of sewage sludge was produced in the European Union, of which approximately 27% of used in agriculture (Eurostat, 2018) – by far the single largest pathway of reuse of sewage sludge. Full datasets from many EU countries are unavailable on Eurostat, but in those countries for which data are available, the highest percentage of reuse of sewage sludge in agriculture (over 80 %) occurs in Ireland. Traditionally, sewage sludge was disposed of to landfill, but legislation such as the Landfill Directive (1999/31/EC; EC, 1999), which imposes strict rules on the amount of biodegradable municipal waste sent to landfill, has meant that other disposal pathways are now used. Recycling to land is currently the most economical and beneficial method of sludge management, but concerns have been raised by scientists, policy makers, and the public over this disposal pathway.

The main issue is the presence of known and unknown contaminants in sewage sludge, which may potentially cause a risk to human health if applied to land (Smith, 2009). Indeed, some scientists have stated that this practice is equivalent to "playing Russian roulette with sewage sludge", and that over time "this game will be lost at the public's expense" (Rayne and Honour, 2016). These concerns have led some countries, such as Belgium and Switzerland, to ban the land application of sewage sludge.

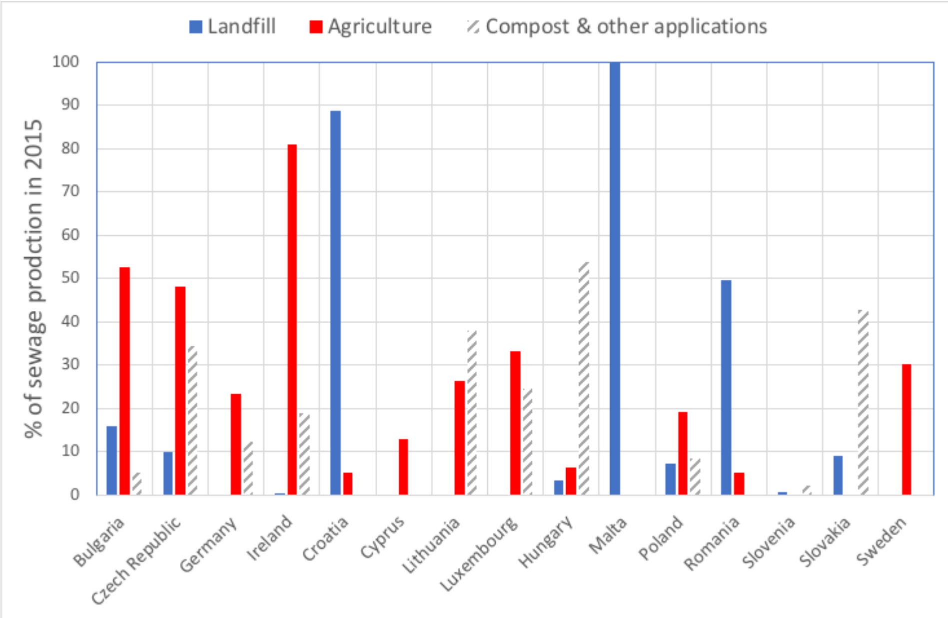

Figure 1. Percentage of sewage sludge production (settled solids from treated wastewater) sent to landfill, reused in agriculture, and compost and other applications. Data presented from countries from which data are available (Eurostat, 2018).

Undoubtfully, there are potentially huge benefits associated with the application of treated sewage sludge ("biosolids") to land, including the reuse of a valuable resource as a fertiliser replacement and the attainment of EU objectives of a "circular economy". However, set against these, are concerns regarding potential impact on water and soil quality, and the migration of potentially very harmful contaminants into the human food chain. It was these concerns that led the Irish Environmental Protection Agency (EPA) to fund a study to investigate these issues, so as a holistic view of the environmental and human health risk arising from the land application of sewage sludge could be elucidated. This paper aims to summarise the overall findings of this study. It focuses on four areas: (1) the characterisation of treated sewage sludge (biosolids) for metals and microplastics (MPs) (synthetic polymers measuring less than 5 mm in diameter, which have recently emerged as contaminants of concern; Mahon et al., 2016) (2) the impact of the land application of biosolids on surface water quality (3) the uptake of metals by vegetation in the weeks following the land application of biosolids, and (4) the risk to human health, following the treatment in water treatment plants of biosolid-contaminated waters.

Materials and Methods

Characterisation of biosolids

There are over 500 wastewater treatment plants (WWTPs) in Ireland, with population equivalents (PEs) ranging from less than 100 to over 2.3 million. Sludge samples, which were subject to either anaerobic digestion (AD), thermal drying (TD), or lime stabilisation (LS), were collected from 16 WWTPs representing a range of PEs. Metals for which legislation currently exists (Ni, Cu, Zn, Cd, Hg, and Pb), as well as metals for which there is no requirement for testing prior to land application ("unlegislated metals"), were measured using a handheld X-ray fluorescence (XRF) analyser, which was calibrated with sewage sludge certified reference materials (Healy et al., 2016a). The abundance of MPs was quantified using stereomicroscopy equipped with a polarizer attachment and a digital camera, attenuated total reflectance (ATR) and Fourier transform infrared spectroscopy (FTIR) (Mahon et al., 2017).

Surface runoff experiments

Small plot-scale experiments were used to measure the nutrient, metal, microbial, and antimicrobial concentration of surface water lost in surface runoff, following simulated rainfall events (Peyton et al., 2016). Anaerobically digested, LS, and TD biosolids were applied at the maximum permissible rate, at n = 5 replications per treatment, to plots each measuring 0.4 m in width and 0.9 m in length. Each plot was subject to a one-hour rainfall event with an intensity of 11 mm h^{-1} at 24, 48 and 360 h after application of the biosolids. To give the study context, dairy cattle slurry was applied at the same rate to separate plots of the same dimensions.

Measurement of metal uptake by vegetation

Current Irish legislation states that no animal fodder may be harvested until “at least three weeks” after application of sewage sludge to land, and that animals may not be grazed on the land until “three to six weeks” after application (Fehily Timoney and Co., 1999). To test this, cuts of grass sward were harvested from the plots at incremental periods up to 18 weeks after application and tested for legislated and unlegislated metals (Healy et al., 2016b).

Human health risk assessment

Using surface runoff data for total coliforms, *E. coli* (Clarke et al., 2017), antimicrobials (Clarke et al., 2018), and metals (Clarke et al., 2016), measured in the plot experiments, quantitative risk assessment models were developed which examined the risk to human health following ingestion of water following dilution, residence time in a stream, die-off rate, treatment in water treatment plants, and human exposure.

Results

Characterisation of biosolids

With the exception of Pb (which was in exceedance in biosolids samples from one WWTP), all of the legislated metals were below their limit values for sludge for use in agriculture. The PE of the WWTPs was not found to be significant in influencing the metal concentrations. The concentrations of the unlegislated metals were below international guideline values (where such values exist) and baseline values in soils. However, antimony, which ranged in concentration from 17 to 20 mg kg^{-1}, was substantially higher than in non-polluted soils (<1 mg kg^{-1}) (Healy et al., 2016a). Microplastics ranged in abundance from 4196 to 15,385 particles kg^{-1} (dry matter, DM) in all sites (Mahon et al., 2017).

Surface runoff experiments

The surface runoff water from all treatments contained elevated levels of nutrients, metals and coliforms, relative to control plots, which received no biosolids/slurry applications. However, with the exception of elevated concentrations of copper, these concentrations were much lower than plots which received applications of dairy cattle slurry (Peyton et al., 2016).

Measurement of metal uptake by vegetation

The metal concentrations of the ryegrass sward reduced over time, which was attributed to the dilution effects of grass growth. There was no significant difference between the metal content of the control plots (no biosolids/slurry application) and the biosolids-amended plots (Healy et al., 2016b).

Human health risk assessment

Dose-response relationships, characterised for the different contaminants, indicated that children may be at most risk for exposure to copper, arising from the land application of LS biosolids. However, despite this, the risk of illness was negligible for healthy individuals.

Discussion

This study found that for the parameters examined, the land application of biosolids pose no greater threat to water quality than dairy cattle slurry. Further, it found that cattle exclusion times on land on which biosolids have been applied, is perhaps overly strict, and that risk to human health arising from ingestion of biosolids-contaminated water, is negligible. However, the results are more nuanced than what these main findings suggest. While this study examined a comparatively wide range of contaminants from water and soil quality and human health perspectives, these represent only a small fraction of the potential contaminants that may be present in sewage sludge – the so-called “unknown unknowns” (Rayne and Honour, 2016). This is particularly pertinent in the case of pharmaceuticals and personal care products (PPCPs), which are expansive (Verlicchi and Zambello, 2015), cannot be fully treated in WWTPs (Narumiya et al., 2013), prohibitively expensive to routinely test, and for which no safety guidelines currently exist.

While these and other emerging contaminants, such as MPs, may be degraded by photo- and thero-oxidative degradation, or may naturally die-off, the gradual build-up in the soil, particularly when land is subjected to multiple applications of sludge over many years, may pose a serious risk to human health and the environment. Recently, much has been written about the presence of MPs (Hale, 2018), particularly in the marine environment (Setala et al., 2018). The large surface area of MPs per unit volume mean that they are potentially good adsorbers of metals (Turner and Holmes, 2015). As evidence from this and other studies suggest that MP abundance is relatively unaffected by the treatment process employed on sewage sludge, the soil could act as a pathway for these highly adsorbent MPs between the source of pollution and the receptor (surface and groundwater).

In general, legislation regarding the land application of treated and untreated sewage sludge has been very cognisant of these issues, with some countries banning the application of sewage sludge to land. This is in part due to concerns over the “unknown unknowns” in sewage sludge, but may be also driven by issues of perception. For example, despite Irish legislation allowing for the land disposal of untreated (for PEs < 5000; SI 148, 1998) and treated sewage sludge, the body responsible for the export of Irish produce worldwide, Bord Bia, prohibits its application to their certified farms (Bord Bia, 2013). This contradiction between government legislation and policy is very much a case of “perception, not science”, as one operator told this author.

While most studies, including the current one, indicate that the environmental and human health impacts are relatively minimal, they all acknowledge that a certain about of uncertainty still exists. How policy makers respond to current and future studies remains to be seen.

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