Science of the Total Environment 566–567 (2016) 102–112

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

A quantitative risk assessment for metals in surface water following the application of biosolids to grassland

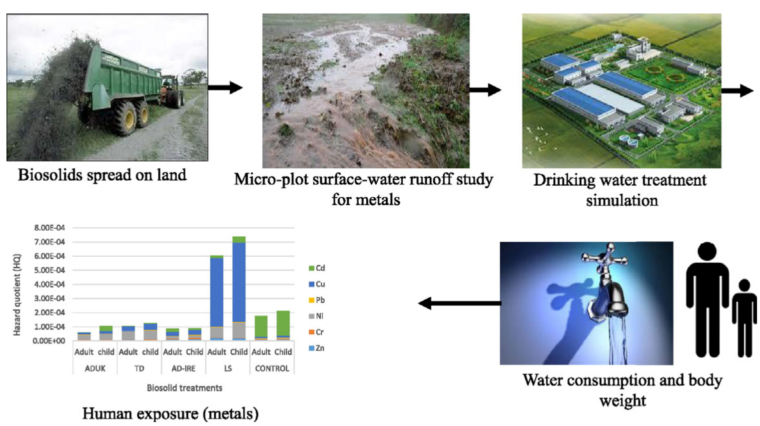
Rachel Clarke [a,*], Dara Peyton [b], Mark G. Healy [b], Owen Fenton [c], Enda Cummins [a]

[a] School of Biosystems and Food Engineering, Agriculture and Food Science Centre, University College Dublin, Belfield, Dublin 4, Ireland
[b] Civil Engineering, National University of Ireland, Galway, Co. Galway, Ireland
[c] Teagasc Environment Research Centre, Johnstown Castle, Co. Wexford, Ireland

HIGHLIGHTS

- The application of biosolids on agricultural land may lead to accumulation of metals in soil.
- Results show that child exposure was highest for copper and lime stabilised biosolids.
- Sensitivity analysis reveal tap water intake and filtration reduction as parameters of importance.
- Metal concentrations in the biosolids were not considered a risk to human health.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 January 2016
Received in revised form 10 May 2016
Accepted 13 May 2016
Available online xxxx

Editor: F.M. Tack

Keywords:
Biosolids
Drinking water
Metals
Risk
Water treatment

ABSTRACT

During episodic rainfall events, land application of treated municipal sludge ('biosolids') may give rise to surface runoff of metals, which may be potentially harmful to human health if not fully treated in a water treatment plant (WTP). This study used surface runoff water quality data generated from a field-scale study in which three types of biosolids (anaerobically digested (AD), lime stabilised (LS), and thermally dried (TD)) were spread on micro-plots of land and subjected to three rainfall events at time intervals of 24, 48 and 360 h following application. Making the assumption that this water directly entered abstraction waters for a WTP without any grassed buffer zone being present, accounting for stream dilution, and modelling various performance scenarios within the WTP, the aim of this research was to conduct a human health risk assessment of metals (Cu, Ni, Pb, Zn, Cd and Cr), which may still be present in drinking water after the WTP. Different dose-response relationships were characterised for the different metals with reference to the lifetime average daily dose (LADD) and the Hazard Quotient (HQ). The results for the LADD show that child exposure concentrations were highest for Cu when the measured surface runoff concentrations from the LS biosolids treatment were used as input into the model. The results for the HQ showed that of all the scenarios considered, Cu had the highest HQ for children. However, values were below the threshold value of risk (HQ < 0.01 - no existing risk). Under the conditions monitored, metal concentrations in the biosolids applied to grassland were not considered to result in a risk to human health in surface water systems.

* Corresponding author.
E-mail addresses: rachel.clarke.1@ucdconnect.ie (R. Clarke), enda.cummins@ucd.ie.ph (E. Cummins).

http://dx.doi.org/10.1016/j.scitotenv.2016.05.092
0048-9697/

1. Introduction

Long-term application of treated municipal sewage sludge ('biosolids') to agricultural land has led to concerns regarding the potential accumulation of metals in soil, their subsequent runoff into surface waters, and the potential risk to human health through drinking water consumption. While the environmental occurrence of these contaminants is usually low (μg kg^{-1} down to sub ng kg^{-1}), toxicologists, epidemiologists and risk assessment experts advise that there may still be significant and widespread adverse environmental and human health consequences (i.e. cancer risk and adverse reproductive development) at the detected levels (Clarke and Cummins, 2014). The metals of concern and those primarily linked to human poisoning are lead (Pb), iron (Fe), copper (Cu), cadmium (Cd), zinc (Zn), chromium (Cr), mercury (Hg) and arsenic (As) (Singh et al., 2011; Tchounwou et al., 2012). Essential metals such as Cu, Zn and Cr are required by the body in trace amounts, but can be toxic in large doses (Mohod and Dhote, 2013). A distinguishable feature of metals is that, unlike any other toxic substance, they are not biodegradable and can accumulate in the sludge to potentially toxic concentrations (Chen et al., 2008). The main cause of this toxic effect is due to the chemical binding of metals to enzymes and subsequent disruption to enzyme structure and function (Appels et al., 2008). Metal toxicity can result in brain damage or a reduction in mental processes (Fernández-Luqueño et al., 2013). Salem et al. (2000) reported that in some cities in Egypt, there was a strong correlation between consumption of water heavily contaminated with metals and chronic diseases such as renal failure, liver cirrhosis, chronic anaemia and hair loss. Excessive consumption of Cu can lead to gastrointestinal problems, kidney damage, anaemia and lung cancer (Mahiya et al., 2014). Children are more vulnerable to metal exposure, which can lead to several paediatric effects including neurodevelopment disorders (Oyoo-Okoth et al., 2013). Davis et al. (2014) reported that infants and children are more vulnerable to neurotoxic effects of metals due to more rapid bone growth and differences in physiology, even at low levels of exposure. Due to the adverse effects on the central nervous system, the US Centre for Disease Control and Prevention (CDC) introduced guidelines that identifies a blood level > 0.48 μmol Pb L^{-1} (100 μg L^{-1}) to be of concern in children, and it was recommended to lower the Pb level to 0.24 μmol Pb L^{-1} (50 μg L^{-1}), the amount that sometimes may occur as background levels in some countries (Nordberg et al., 2014).

Increasingly, there is evidence to show negative health effects from cumulative, lower level exposures to some metals (Tchounwou et al., 2012). The biological half-lives of metals vary and the amounts excreted can reflect a combination of recent and past exposures (Quandt et al., 2010). For instance, the half-life of Cd is one-to-four decades, and urinary excretion of Cd reveals long-term exposure to the metal (ATSDR, 2008). Liu et al. (2013) reported an increased life-time risk of death due to lung cancer resulting from occupational exposure to dusts and mists containing hexavalent Cr.

Soils represent a major sink for metal ions that can then enter the food chain (i.e. drinking water) *via* surface (e.g. in runoff after episodic rainfall events) and subsurface pathways (i.e. ground water) (Fernández-Luqueño et al., 2013; Clarke et al., 2015). In fact, groundwater and surface waters can be linked and thereby affect each other (Vero et al., 2014). Previous studies have shown that overland transport of metals from fields (with eventual runoff to the transfer continuum at delivery points) amended with biosolids can impact the quality of surface waters (Topp et al., 2008). These metals may be present in mobile forms in biosolids, which may migrate to the fertilised soil, or in immobile forms, which do not produce any toxicological effect (Gawdzik and Gawdzik, 2012). Chang et al. (1984) found that >90% of the Cd, Cr, Zn, Cu, Ni and Pb present in biosolids, which were land applied over a 6-year period in a field-scale experiment, remained in the cultivated layer (0–15 cm) in both sandy and loam soils. Similarly, Hinesly et al. (1972) reported the movement of Cd, Cr, Ni, Zn and Cu to a depth of 30–45 cm in arable agricultural soil (permeable silt loam texture) following biosolids application (applied at 13.6 t $acre^{-1}$) over a 4-year period. Therefore, greater concentrations of metals in biosolids, combined with long-term use on some soil types, may potentially be a hazard to the environment. Joshua et al. (1998) monitored the surface and subsurface movement of nutrients and metals in runoff and the soil profile following land application of biosolids over a 3-year period, and found that biosolids reduced runoff and increased surface retention of rainfall. The study concluded that there was a low potential for pollution of surface or groundwaters by metals.

With regards to the behaviour and fate of metals in soils and transfer along the food chain, the "plateau" and "time bomb" theories are opposite philosophies used to explain the behaviour of metals in soil and uptake by plants in response to biosolid application on agricultural land. The "plateau" hypothesis considers that metals are so tightly bound by the organic matter in biosolids and hydrous oxides of Fe and Mn and clays in the soil, that their bioavailability or toxicity is greatly reduced and that they are retained in the soil's surface horizon or in the plough layer instead of the being taken up by plants or leaching down the soil profile (Lu et al., 2012). The "time bomb" hypothesis considers that the slow mineralisation of the organic matter present in the biosolids could release metals in readily soluble form, which then may become available for plant up-take (Silveira et al., 2003). Chang et al. (1997) obtained experimental data from a 10-year field biosolids study on agricultural land to evaluate the hypothesis of the plateau and time bomb theories. They concluded that neither a plateau nor time bomb was evident despite an increasing rate of biosolid application (2880 mg ha^{-1}), which represented a "worst case scenario" in terms of contaminant loading.

1.1. Drinking water treatment process

Drinking water treatment may involve several stages such as pretreatment or primary treatment (coarse screening, storage and neutralisation), secondary treatment (coagulation/flocculation/sedimentation, rapid and slow sand filtration) and tertiary treatments (disinfection, activated carbon and membrane processes). The pretreatment process is defined depending on the closeness of the water source to the treatment plant and whether it is an upland or lowland water source. Storage is used primarily for water abstracted from lowland rivers to improve water quality before treatment and to ensure adequate supplies at periods of peak demand (Gray, 2010).

Secondary treatment involves the coagulation, flocculation, sedimentation and filtration of the influent. The commonest types of coagulants used are aluminium-based (e.g., aluminium sulphate (alum) or polyaluminium chloride (PAC)). Both aluminium (Al) and ferric salts, either in monomer or polymeric forms, have been reported to be effective coagulants in treating metals in wastewater (Kang et al., 2003; Pang et al., 2009). In Ireland, the most commonly used coagulant is alum, followed by a very small number of plants using Fe-based coagulants (ferric chloride or ferric sulphate) (Cummins et al., 2010). Fatoki and Ogunfowokan (2004) reported removal efficiencies of 90% for Cr, 68% for Zn, and 100% for Ni using ferric sulphate, compared to alum, which had removal efficiencies of 81%, 47% and 55%, for Cr, Zn and Ni, respectively. Jiménez (2005) reported 78, 39 and 36% removals of Cd, Ni and Cr, respectively, following 100 mg L^{-1} dose of alum on wastewater in Mexico. With the use of recycled alum sludge in the coagulation process, Chu (1999) reported that Pb removals increased from 79 to 98% with 100–180 mg L^{-1} of recycled alum sludge. Hannah et al. (1977) reported metal removals of between 25 and 100% using alum and incorporating chemical clarification and carbon adsorption.

The filtration process in a conventional WTP consists of slow or rapid sand filtration. The purpose of filtration is to remove suspended particles in the water by moving the water through a medium such as sand. Aulenbach and Chan (1988) reported the effect of rapid sand filtration on metal removal from mixed industrial and domestic