

Effect of Rainfall Time Interval on Runoff Losses of Biosolids and Meat and Bone Meal when Applied to a Grassland Soil

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Abstract This study assessed runoff losses following laboratory rainfall simulation on a grassland soil at two time intervals (48 and 216 h) after a single application of biosolids and meat and bone meal (MBM). The treatments were, a soil-only control, three types of biosolids (lime-stabilised (LS), thermally dried (TD) and anaerobically digested (AD)) and two types of MBM (low ash and high ash content) all applied at two rates (the maximum and double the maximum legal application rate currently permitted in Ireland). Results showed that treatment, time interval and their interactions all had significant effects on dissolved reactive P (DRP), total P (TP) and total dissolved P (TDP) concentrations. Time interval had the greatest effect for DRP and TP concentrations, while treatment was more significant for TDP. All treatments released DRP concentrations in excess of $30 \mu\text{g DRP L}^{-1}$. Anaerobically digested biosolids released the least amount of DRP into surface runoff for

both application rates at both time intervals. Low ash content MBM, applied at the maximum legal rate, released the most DRP at both time intervals, and the TD biosolids released the most DRP when applied at double the maximum rate. Lime-stabilised biosolids released the most TP in runoff at both application rates. Runoff comprised of $>50\%$ particulate P for all treatments. Besides TD biosolids, all treatments, applied at both rates, released lower concentrations of suspended solids (SS) during the second time interval than the first. Soil-specific effects were also evident, although the soil was non-calcareous and had a low pH and high amounts of available aluminium and iron, high organic matter ensured low levels of P adsorption.

Keywords Biosolids · Meat and bone meal · Phosphorus · Agriculture · Runoff

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1 Introduction

Biosolids, derived from the wastewater treatment process, and meat and bone meal (MBM), the by-products of the rendering industry, may be used as agricultural fertilisers, as they contain organic matter (OM) and inorganic elements (Ylivainio et al. 2008). When spread on tilled land or grassland, they supply nutrients and metals required for plant and crop growth, and may also be used as an aid in the development of a soil's physical and chemical characteristics (Meyer et al. 2001; Mondini et al. 2008). A large amount of sewage sludge and MBM are produced in Europe. Annual production

of MBM in Ireland peaked in 2003 (150,000 t p.a.; Inter Departmental/Agency Committee on Disposal Options for MBM 2003), but declined due to a decrease in herd sizes (SEI 2004). Quantities of sewage sludge produced at urban waste water treatment plants in 2009 were approximately 106,000 t, with 62 % (down from 70 % in 2007) of this being reused on agricultural land (Monaghan et al. 2012). The drive to reuse sewage sludge has been accelerated by the Landfill Directive, 1999/31/EC (European Commission (EC) 1999) and the Renewable Energy Directive (2009/28/EC; EC, 2009), which places an increased emphasis on the production of biomass-derived energy. One such means of reuse is landspreading, although potential hazards associated with their application to land, such as excessive nutrient release, need to be evaluated.

To dispose of treated sewage sludge (biosolids) in Ireland, farmers must abide by the European Union (EU) (Good Agricultural Practice for Protection of Waters) Regulations 2014 (Statutory Instrument (SI) 31 2014). Compliance with the regulations requires protection of waters against pollution from agricultural sources and management of manures and fertilisers. In addition, the application of biosolids to agricultural land is governed in Europe by EU Directive 86/278/EEC (EEC 1986), and within Ireland, is enacted by the 'Codes of Good Practice for the Use of Biosolids in Agriculture' (Fehily Timoney and Company 1999).

The handling (and disposal) of MBM is subject to strict enforcement in many European states. For example, in Ireland, S.I. 551 of 2002 requires possession of a licence for the manufacture, sale or supply of MBM. In addition, strict guidelines govern its reuse as a bioenergy resource (SEI 2004). Land application of various categories of MBM is permitted within the EU member states, provided certain criteria are adhered to (EC 2002; EC 2006). These criteria include the handling and processing procedures and the prevention of land application of fertilisers consisting of Category 1 MBM. However, EC Regulation No. 181 of 2006 (EC 2006) provides for stricter national rules to be implemented by each individual member state (EC 2000), and this has resulted in the prohibition of the land application of organic fertilisers composed of Category 2 and 3 MBM materials in certain member states (e.g. the Republic of Ireland has banned the land application of MBM under S.I. No. 253 of 2008).

In the EU, land application of biosolids and MBM is typically based on their nutrient and metal contents. This

approach results in phosphorus (P) becoming the limiting factor in determining the application rates. This is in contrast to the USA, where biosolids are applied based on their nitrogen (N) content (US EPA 1993). Such a discrepancy means that land application rates of biosolids in the USA tend to be far greater than the EU, resulting in less land required for the application of biosolids—but potentially greater environmental losses.

Phosphorus losses to a surface water body originate from either the soil (chronic) or in runoff where a storm event follows land application of fertilizer (incidental sources) (Brennan et al. 2012). Such losses to a surface water body occur via direct discharges, surface and near surface pathways, and/or groundwater discharge. Where there is a hydrological continuum between a nutrient source (chronic or incidental) and surface water receptor, dissolved reactive phosphorus (DRP) and particulate P (PP) can be delivered to a receptor and, where loads are significant, may adversely affect water quality (Wall et al. 2011). Biosolids and MBM application to soils may also contribute to soil test phosphorus (STP) build-up in soils, thereby contributing to chronic losses of P, metal and pathogen losses in runoff (Gerba and Smith 2005). Dissolved reactive P losses may also be leached from an agricultural system to shallow groundwater (Galbally et al. 2013) and, where a connectivity exists, may affect surface water quality for long periods of time (Domagalski and Johnson 2011; Fenton et al. 2011).

Issues surrounding the potential 'edge-of-field' nutrient concentration and load losses arising from land application of biosolids and MBM may be effectively investigated using low-cost laboratory rainfall simulation studies, as they provide for more control over the variability of the soils' physical and chemical characteristics and surface slope, and allow comparison to be made between treatments (Regan et al. 2010).

The aims of this study were to determine chronic (study control) and incidental flow weighted mean concentrations (FWMCs) and loads (mass release) of DRP, PP, total dissolved phosphorus (TDP), total phosphorus (TP) and suspended solids (SS) in runoff from a grassland soil following the application of three types of biosolids (lime-stabilised (LS), thermally dried (TD) and anaerobically digested (AD)) and two types of MBM (low ash and high ash content) applied at two rates (the maximum and double the maximum legal application rate currently permitted in Ireland) over two successive rainfall events (at the 48-h period

outlined in S.I. 31 of 2014 and after 216 h) to see the lasting effects of potential losses of 11 mm h⁻¹-intensity rainfall events.

2 Materials and Methods

2.1 Biosolids and MBM Characterisation

Three types of biosolids—AD, TD and LS—were collected from three wastewater treatment plants in Ireland. Two types of MBM, one with low ash (MBM LA) and one with high ash (MBM HA) were collected from a slaughterhouse in County Mayo, Ireland. All results with respect to the characterisation of biosolids and MBM were presented previously in Lucid et al. (2013). Briefly, AD, TD and LS biosolids and MBM (high ash) and MBM (low ash) had total P (mg kg⁻¹) contents of 6,916, 7,600, 6,332, 27.9 and 31.1, respectively, and AD, TD and LS biosolids and MBM (high ash) and MBM (low ash) had total N (mg kg⁻¹) contents of 6.8, 30.8, 3.1, 39.7 and 59.1, respectively. Metal content and release were presented in Lucid et al. (2013).

2.2 Soil Collection and Analysis

The soil used in this study was collected from a grassland field in County Galway, Ireland (ITM reference 528060, 727322) and was from the same site as in Lucid et al. (2013). Intact soil sods measuring 0.7 m in length, 0.4 m in width and 0.1 m in depth were collected from site for use in the rainfall simulation study. A separate set of soil samples ($n=3$), 0.1 m in depth and 0.1 m in diameter, was collected from the same site for classification studies. A 2:1 ratio of deionised water-to-soil was used to determine the soil pH. The soil samples were then air dried at 40 °C for 72 h, crushed to pass a 2-mm sieve and analysed for Morgan's P (P_m; the national test used for the determination of plant available P in Ireland) using Morgan's extracting solution after Morgan (1941). To determine the particle size distribution (PSD) of the soil, a sieving and pipette technique was used (BSI 1990b). The OM of the soil was determined by LOI after BSI (1990a). Soil characteristics are presented in Table 1.

Table 1 Classification of soil used in this experiment. Standard deviations, where multiple sampling ($n=3$) was conducted, are in brackets

Water extractable phosphorus (g kg ⁻¹)	0.00249 (0.00054)
pH	5.63
Lime requirement pH	5.90
Morgan's P (mg L ⁻¹)	1.12 (0.09) (P Index 1)
K (mg L ⁻¹)	203.24
Mg (mg L ⁻¹)	239.5
Organic matter (%)	18.18 (1.19)

2.3 Phosphorus Desorption Study and Rainfall Simulator Test

A P desorption study was conducted on each of the five media used in this study to determine the time over which DRP may be released to surface water. This involved placing 6 g of either biosolids or MBM into 120-ml-capacity plastic cups, overlaying them with 100 ml of distilled water, sealing the containers and placing them into an end-over-end shaker for a period of 24 h. All tests were carried out in triplicate ($n=3$). At time intervals of 1, 4, 8 and 24 h, 2.5 ml of water was removed, filtered through 0.45- μ m filters and stored at 4 °C until testing (normally conducted within 1 day of collection). The water samples were tested for DRP in accordance with the standard methods (APHA 1995) by a nutrient analyser (Konelab 20, Thermo Clinical Labsystems, Finland).

The following treatments were examined in a laboratory rainfall simulator ($n=3$): grassed soil-only treatment (the study control) and grassland onto which either TD, LS and AD biosolids or MBM (HA) and MBM (LA) were spread. Each type of biosolids and MBM was applied to the soil at two different rates based on a soil with a P Index of 1 after Lucid et al. (2013; Table 2): the maximum legal application rate and double the maximum legal application rate.

Laboratory runoff boxes, 0.1 m long, 0.225 m wide and 0.075 m deep and inclined at a 5° slope and with the side-walls 0.025 m higher than the grassed sods, were used in this experiment. Each runoff box had 0.005-m-diameter drainage holes, located at 0.3-m-centres in the base, after Regan et al. (2010) and Brennan et al. (2011), to allow for drainage of water at the base. Muslin cloth was placed at the base of each runoff box, covering these drainage holes, before packing the soil in order to prevent soil loss. Immediately prior to the start of each

Table 2 Application rates of biosolids and meat and bone meal (MBM) to the soil in this study using a P Index 1 soil

Nutrient type	Maximum legal application rate ^a		Double the maximum legal application rate	
	Wet weight t ha ⁻¹	Dry solids	Wet weight	Dry solids
AD biosolids	14.8	3.3	29.6	6.6
TD biosolids	3.3	3.0	6.5	6.0
LS biosolids	18.0	5.2	36.0	10.4
MBM (HA)	0.9	0.8	1.7	1.6
MBM (LA)	0.8	0.7	1.5	1.4

^a Legal limits, estimated after Lucid et al. (2013)

experiment, the soil sods were trimmed and packed into the runoff boxes. The runoff boxes were then positioned under the rainfall simulator. The rainfall simulator consisted of a single 1/4HH-SS14SQW nozzle (Spraying Systems Co., Wheaton, IL) attached to a 4.5-m-high metal frame. The rainfall simulator was calibrated to achieve an intensity of $10.85 \pm 0.14 \text{ mm h}^{-1}$ with droplet impact energy of $260 \text{ kJ mm}^{-1} \text{ ha}^{-1}$ at 86 % uniformity after Regan et al. (2010) and Brennan et al. (2011). The source of the water that was used for the rainfall simulations was potable tap water, which had a DRP concentration of less than 0.005 mg L^{-1} , a pH of 7.73 ± 0.2 and an electrical conductivity (EC) of 0.494 dS m^{-1} .

The packed sods were then saturated using a rotating disc, variable-intensity rainfall simulator (after Williams et al. 1997) and left to drain for 24 h by opening the 5-mm-diameter drainage holes at the base of the runoff box before continuing with the experiment. After this time elapsed, the grassed sods were assumed to be at field capacity after Regan et al. (2010). The biosolids and MBM were spread over the surface of the sods, and the drainage holes were sealed for the remainder of the experiment. In accordance with S.I. No. 31 of 2014, the laboratory runoff boxes were then left in this state for a period of 48 h. The first rainfall event (RE 1) was applied at $t=48 \text{ h}$ (after the application of the treatments). To investigate the breakdown, if any, of the treatments with time and the lasting effects that this may have on surface runoff, the second rainfall event (RE 2) occurred at $t=216 \text{ h}$ (on the same sod).

Rainfall was applied to each runoff box until consistent, continuous droplets of water flowed from the

runoff box; once this state had been achieved, each rainfall event lasted for 30-min duration. Surface runoff samples for each event were collected in 5-min intervals over 30 min, with a final sample collected in the period after rainfall had completed and runoff had ceased.

2.4 Water Sample Collection and Analysis

Runoff samples were collected in 1-L containers (covered to prevent rain water entering the container) at the bottom of the runoff box. Immediately after collection, a sub-sample of the runoff water was passed through a $0.45\text{-}\mu\text{m}$ filter and analysed colorimetrically for DRP using a nutrient analyser (Konelab 20, Thermo Clinical Labsystems, Finland). A second filtered sub-sample was removed, and TDP was measured using potassium persulphate and sulphuric acid digestion (Hach Lange, Germany). An unfiltered sub-sample was removed and analysed for the TP in the same manner as for TDP analysis. The DRP of every sample was measured, while the TDP and TP were measured for the 10, 20 and 30-min interval samples, as well as for the last sample removed after the rainfall had stopped. The DRP was subtracted from the TDP to give the dissolved unreactive phosphorus (DUP). Particulate phosphorus was calculated by subtracting TDP from TP. In order to determine the SS concentration, a well-mixed, unfiltered sample of runoff water was passed through Whatman GF/C (pore size; $1.2 \mu\text{m}$) filter paper by vacuum filtration. All water samples were tested in accordance with standard methods for the examination of water and wastewater (APHA 1995). Flow-weighted mean concentrations for nutrients and SS in runoff were determined by dividing the total mass load for the runoff event by the total flow volume for the same period.

Measurements of pH and dissolved oxygen (DO) were also conducted using a pH probe (WTW SenTix 41 probe with a pH 330 metre, WTW, Germany) and a DO probe (WTW Oxi 315i metre with a CelloX 325 oxygen sensor, WTW, Germany), respectively. The time to runoff was also recorded, and the runoff ratio for each flume was determined. The runoff ratio is defined as the ratio of the volume of surface runoff to the amount of rainfall applied over the duration of the rainfall simulation.

2.5 Statistical Analysis

The analysis was carried out as a rainfall interval (time 48 or 216 h after application) by treatment (biosolids or

MBM type and rate) factorial experiment with the non-randomised times modelled with a repeated measures structure. As there were only two time intervals, all correlation models gave the same result. The rainfall simulator data were analysed using the GLIMMIX Procedure of the Statistical Analyses System (SAS Institute 2004) with each flume as the experimental unit. For all analyses, significance was given as $p < 0.05$. Dissolved reactive phosphorus, DUP, PP, TDP, TP, SS, DO, pH and runoff ratio were analysed as repeated measures using the MIXED procedure of SAS with Tukey–Kramer adjustment for multiple comparisons with a covariance structure to account for correlations between the repeated measures. The dependent variables were DRP, DUP, PP, TDP, TP, SS, DO, pH and runoff ratio for this experimental analysis. For all the above analyses, the fixed effects were treatment, time, treatment \times time and flume. Time and treatment were the repeated measures. Where interactions were significant, the comparisons within the means for the time by treatment combinations were examined. Where interactions were not significant, the main effects were interpreted and comparisons within each main effect were examined.

3 Results and Discussion

3.1 Soil Analysis and Rainfall Simulator Test

The soil used in this study was a non-calcareous soil with a low pH. This could lead to high amounts of available Al and Fe, which can bind with P, rendering it unavailable. However, the soil had high OM levels (18 %; Table 1), and given that ~12–14 % is the threshold between a mineral and organic soil type, the Al and Fe in biosolids may be complexed in OM and would be unavailable. Therefore, this soil had limited capacity to bind the added P, and the high OM may have worked against the amendments by complexing the Al or Fe in them. The OM content of the soil is an important parameter, which determines soil suitability of receiving MBM and biosolids.

Treatment, time and their interactions all had significant effects on DRP, TP and TDP concentrations. Time had the greatest effect for DRP and TP concentrations, while treatment was more significant for TDP. Treatment and a treatment \times time interaction were significant for DUP and PP, but not time on its own. The

average FWMCs and load (mass release) of DRP, DUP and PP in the surface runoff for the two rainfall events, at both application rates, are shown in Figs. 1 and 2. Anaerobically digested biosolids released the least amount of DRP into surface runoff for both application rates and both rainfall events. There was no significant difference in DRP losses between the control and all treatments when compared for the maximum legal application rate ($p < 0.05$). All treatments, at both application rates, and the study control, released DRP in excess of the $30 \mu\text{g L}^{-1}$, the concentration over which significant deterioration of rivers and other surface water bodies may occur (Clabby et al. 2008). However, owing to the buffering capacity of receiving waters, it is likely that these concentrations would be considerably reduced in receiving watercourses.

Low ash-content MBM, applied at the maximum legal rate, released the most DRP for both rainfall events, and the TD biosolids released the most DRP when applied at double the maximum legal rate ($p < 0.05$). These results followed the same general trend as the desorption study. Of the five materials examined in desorption test, at the 24-h period, the AD (0.013 g kg^{-1}) and LS (0.015 g kg^{-1}) biosolids released the least amount of DRP (Fig. 3); this was followed by MBM (HA) (0.446 g kg^{-1}), TD biosolids (0.569 g kg^{-1}) and MBM (LA) (0.713 g kg^{-1}), the latter of which produced DRP concentrations 70 times larger than the AD and LS biosolids. With the exception of the TD biosolids, all the treatments released 90 % of their DRP after 1 h of continuous mixing with water.

Lime-stabilised biosolids released the most TP into the surface runoff at both application rates ($p < 0.05$). The main purpose of the lime addition to biosolids is to remove pathogens (Epstein 2002). However, liming is sometimes associated with an initial flush of soluble OM and dissolved organic P, which can increase P losses in runoff (Murphy 2007). With the exception of the LS biosolids, at both application rates, there was no statistical difference between the TP concentrations during both rainfall events. Although there was no significant difference in pH between treatments, at an elevated pH similar to the range measured in the current study (pH 6–8), organic P can be more soluble due to desorption or dispersion of OM (Hannapel et al. 1964). With the exception of the three cases (RE 2 of the control and RE 1 of the TD biosolids applied at both rates), the surface runoff comprised mainly of PP throughout all of the treatments, with less than 50 % present as DRP

Fig. 1 Average flow-weighted dissolved reactive phosphorus (DRP), dissolved unreactive phosphorus (DUP) and particulate phosphorus (PP), which collectively make-up the total phosphorus (TP), in runoff after each rainfall simulation event for the treatments applied at both the maximum legal (a) and double the maximum legal (b) rate. The concentrations measured for first rainfall event and the second rainfall event are denoted by '1' and '2', respectively. Dashed line represents 30 $\mu\text{g P L}^{-1}$ maximum admissible concentration for surface waters (Clabby et al. 2008)

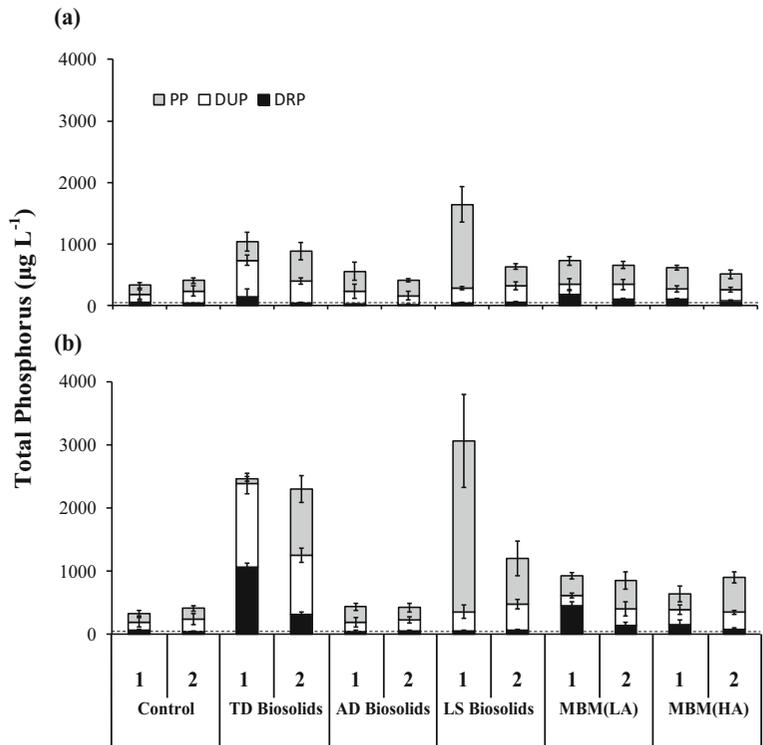


Fig. 2 Average flow-weighted dissolved reactive phosphorus (DRP), dissolved unreactive phosphorus (DUP) and particulate phosphorus (PP), which collectively make-up the total phosphorus (TP), in runoff after each rainfall simulation event for the treatments applied at both the maximum legal (a) and double the maximum legal (b) rate. The concentrations measured for first rainfall event and the second rainfall event are denoted by '1' and '2', respectively

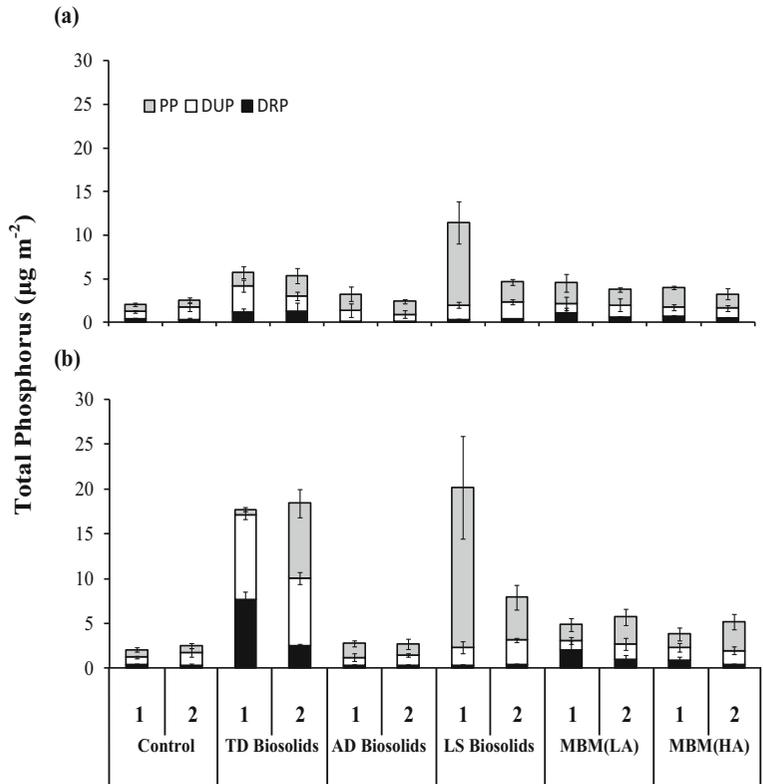
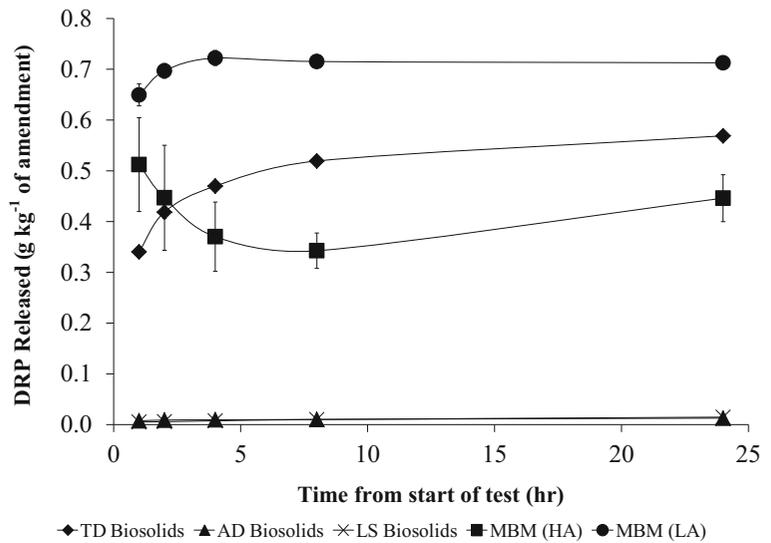


Fig. 3 Phosphorus desorption from the treatments examined



(Fig. 4). The relatively small amount of highly mobile P means that measures such as buffer zones would trap most of the P in surface runoff.

The runoff ratio for each rainfall event is displayed in Fig. 5. The addition of biosolids to grass has been found

in some cases to affect the volume of surface runoff from soil: Joshua et al. (1998) applied a one-time application of AD biosolids at the rates of 0, 30, 60 and 120 t DS ha⁻¹ and, over a 3-year period, found that soil-only control plots produced more runoff than those

Fig. 4 The average % of dissolved reactive phosphorus (DRP), dissolved unreactive phosphorus (DUP) and particulate phosphorus (PP), which comprise of total phosphorus (TP), in runoff for the first ‘1’ and second ‘2’ rainfall events

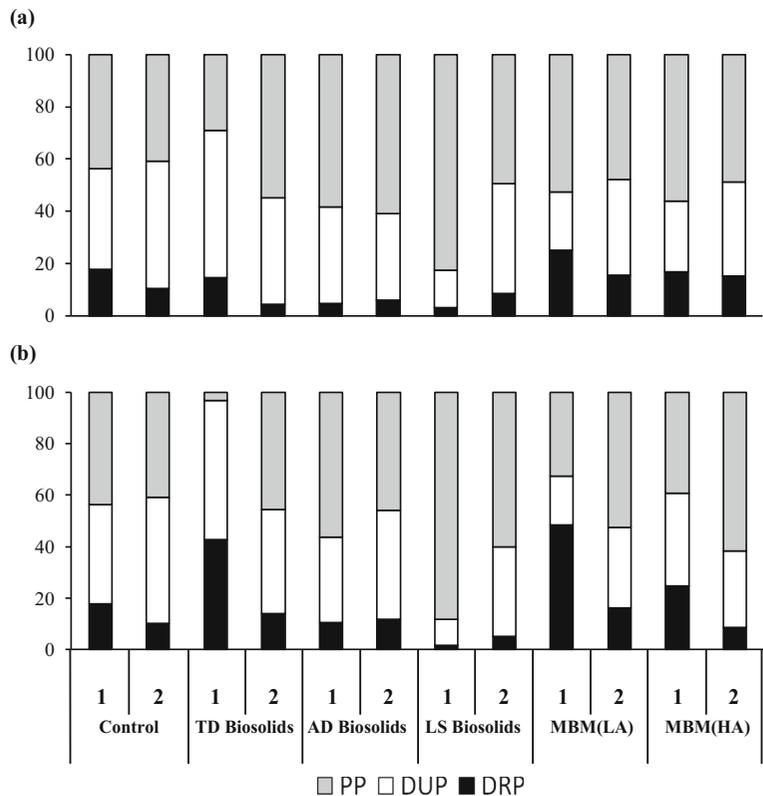
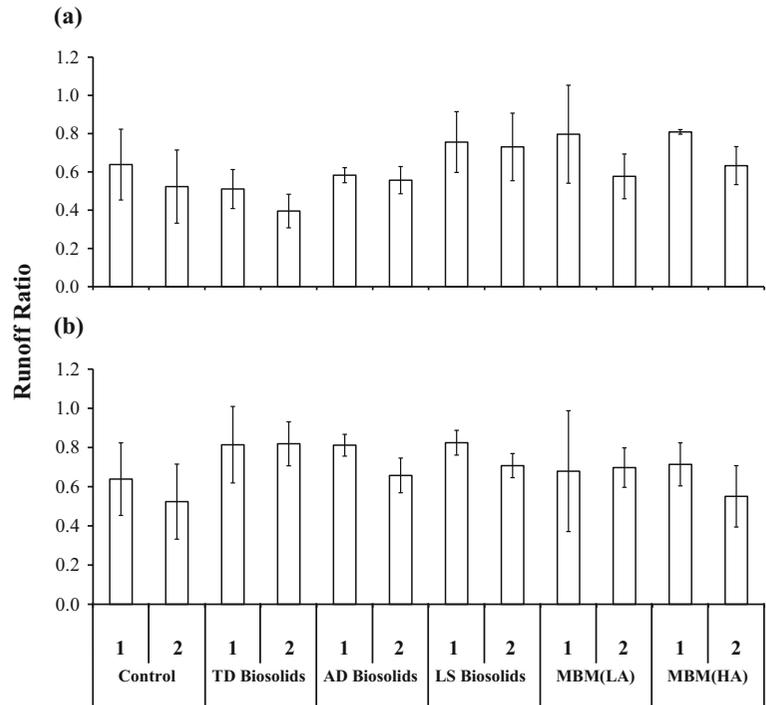


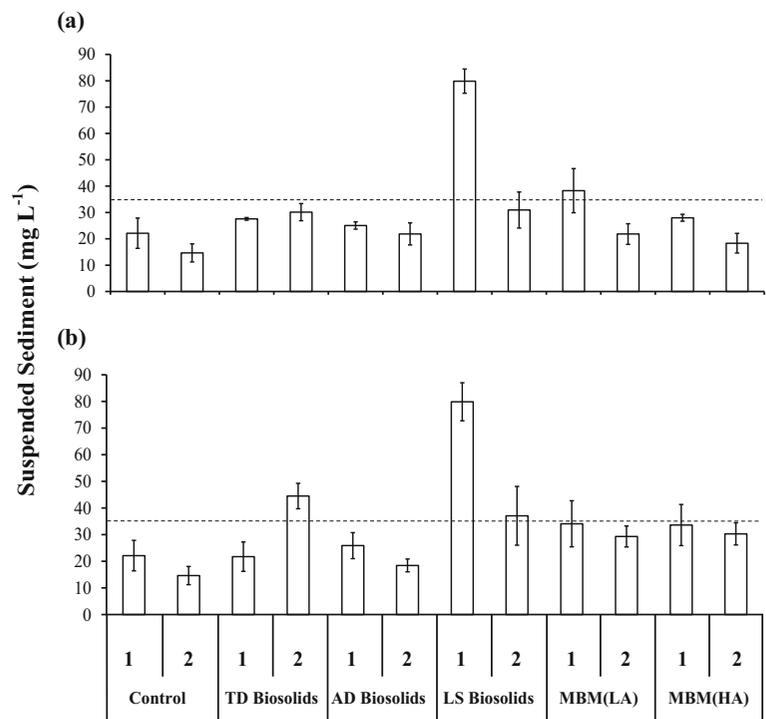
Fig. 5 Runoff ratios from each rainfall simulation event for the treatments applied at both the maximum legal (a) and double the maximum legal (b) rate



applied with biosolids, and that increasing biosolids application produced decreasing runoff volumes for the biosolids-treated plots. However, Meyer et al.

(2001) found that the application of composted biosolids, at rates of 0, 40 and 80 t ha⁻¹, did not significantly affect mean runoff, even though runoff values

Fig. 6 Average flow weighted mean concentrations of suspended solids in runoff after each rainfall simulation event for the treatments applied at both the maximum legal (a) and double the maximum legal (b) rate. The concentrations measured for first rainfall event and the second rainfall event are denoted by '1' and '2', respectively. The dashed line represents the 35 mg L⁻¹ standard (S.I. No 419 of 1994)



were smaller on biosolids-treated plots. In the current study, when applied at the maximum legal application rate, LS biosolids and MBM (HA) increased the volume of surface runoff. When the treatments were applied at double the maximum legal application rate, all treatments increased the volume of surface runoff, but in most cases, these increases were not significant ($p > 0.05$). Although the rate of application was low in comparison to other studies that examined surface runoff (Joshua et al. 1998; Meyer et al. 2001), these results would suggest that higher applications of biosolids and MBM could produce larger volumes of surface runoff, which would impede drainage through the soil structure. There was no correlation between runoff ratio and total mass or concentration of P released. The period between the rainfall events allowed both the soil and biosolids to dry out, therefore when rainfall was applied during RE 2, there was water taken in by both the biosolids and soil.

With the exception of TD biosolids, all treatments, applied at the maximum and twice the maximum legal application rate, released lower concentrations of SS during the second rainfall event than during the first rainfall event (Fig. 6). The increase in SS for the TD biosolids from RE 1 to RE 2 could be due to the breakdown of the biosolids over time. Lime-stabilised biosolids exceeded the allowable discharge limit for the release of SS to surface waters (35 mg L^{-1} ; S.I. 419 of 1994) at both application rates. Low ash MBM and TD biosolids also exceeded the discharge limits on the legal and double the maximum legal application rates, respectively. The incorporation of biosolids into the top layer of soil, as opposed to a surface application, may assist in the reduction of SS. Meyer et al. (2001) reported the reductions in the SS lost to surface runoff compared to control plots (plots receiving no biosolids) when biosolids were incorporated into the top 0.1–0.2 m soil layer.

3.2 Time to Runoff

The time to runoff for each amendment and application rate are presented in the [on-line supplementary data](#). For each treatment and application rate, the time to runoff increased from RE 1 to RE 2. The period between RE 1 and RE 2 allowed the grassed sods to dry out which, in turn, increased the amount of time required during RE 2 for the sods to saturate and for runoff to commence. With the exception of the AD biosolids applied at the

maximum legal rate and TD biosolids applied at both application rates, all other amendments decreased the time to runoff compared to the corresponding control results.

4 Conclusions

This study showed that treatment, time interval and their interactions all had significant effects on DRP, TP and TDP concentrations in surface runoff after the application of biosolids (lime-stabilised, thermally dried and anaerobically digested) and MBM (low ash and high ash content) to a grassland soil. Time interval had the greatest effect for DRP and TP concentrations, while treatment was more significant for TDP. All treatments released DRP concentrations in excess of $30 \text{ } \mu\text{g DRP L}^{-1}$ and runoff comprised of $>50 \%$ particulate P for all treatments. The study found that soil type influences the quality of runoff generated. Further research is needed to further investigate this effect.

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