



Potential loss of nutrients, carbon and metals in simulated runoff associated with dairy processing sludge application

S. M. Ashekuzzaman¹ · P. Forrestal¹ · K. Richards¹ · O. Fenton¹

Received: 27 January 2020 / Revised: 13 April 2020 / Accepted: 6 May 2020 / Published online: 17 May 2020
© Islamic Azad University (IAU) 2020

Abstract

Dairy processing wastewater treatment sludge (DPWTS) is applied to grassland to recycle valuable nutrients and organic matter. Until recently the chemical composition of the dominant DPWTS types (calcium, aluminium or iron rich) was unknown, but the latest study (Ashekuzzaman et al. in *J Clean Prod* 230:1266–1275, 2019) indicates that DPWTS has high nitrogen (N) and phosphorus (P) content and their inherent chemical properties may influence their availability and losses. Herein for the first time edge of field-losses and plant-available P changes are simulated in an agitator test 48 h after application. In terms of overall losses, metals are not of concern across treatment types, iron-rich DPWTS has the highest losses in terms of concentration of ammonium-N and carbon, whereas calcium-rich DPWTS has the highest dissolved reactive P (DRP) losses but lowest losses of ammonium-N and carbon. The loss of DRP was found to be strongly positively associated with the increase in water-extractable P, degree of P saturation, Morgan's and Mehlich-3 extractable phosphorus (P_m , M3-P), respectively. Runoff concentrations presented here represent edge of field-losses and may not represent final concentrations entering a surface waterbody. In terms of plant-available P, there were no negative effects with P_m exhibiting a positive trend across DPWTS types and soils. In particular, a significant increase was found for the calcium-rich DPWTS across all soils (average increase in P_m by fivefold). As applying DPWTS to grassland is an understudied area, further field trials should focus on emissions to air and water and examine the fertilizer value from N and P perspectives. Thus a better management of DPWTS as fertiliser can be maintained from an agronomic and environmental context.

Keywords Dairy processing sludge · Phosphorus · Nitrogen · Carbon · Metals · Runoff loss

Introduction

As a by-product of the dairy food industry's wastewater treatment process, significant volumes of "dairy processing wastewater treatment sludge (DPWTS)" are applied to agricultural land as an organic fertilizer for crop production (Britz et al. 2006). Although land application is seen within the dairy industry as only a short to medium term solution for disposal of DPWTS, it may continue in many

countries, e.g. Ireland for some time. For example in that country, 63% of the DPWTS is land spread, 13.6% is used for composting, and the remaining is removed by licensed contractors (Ryan and Walsh 2016). Recent study showed that the annual generation of DPWTS in Ireland has been significantly increased from ~91,000 to 126,718 tonnes (wet weight) between the year 2012 and 2017 (Ashekuzzaman et al. 2019). This increase is consistent with the processing of more raw milk by the dairy processors in Ireland after the abolition of European milk quotas in 2015. Land application of DPWTS benefits farmers by supplying cheap source of nitrogen (N), phosphorus (P) and potassium (K) for crop production, which may provide an excellent opportunity to reduce using chemical fertilisers and improving profit margins. However, this needs to be done without any possible adverse impacts associated with land spreading practice. For example, land spreading of organic fertilisers such as animal slurry/manure, biosolids (treated sewage sludge) and DPWTS may pose the risk of accumulation of nutrients or

Editorial responsibility: Josef Trögl.

Electronic supplementary material The online version of this article (<https://doi.org/10.1007/s13762-020-02768-z>) contains supplementary material, which is available to authorized users.

✉ S. M. Ashekuzzaman
sm.ashekuzzaman@teagasc.ie

¹ Teagasc, Environment Research Centre, Johnstown Castle, Co. Wexford, Ireland



metals in soils, and subsequent runoff and leaching losses to water bodies that can damage the aquatic ecosystem, e.g. causing eutrophication (Peyton et al. 2016). This can happen due to the long-term and over application of bio-based fertilisers to limited regional land areas, where the relevant production process is intense.

In the European Union (EU), agricultural land spreading is regulated to comply with the waste management and nutrient application regulations like “EU Good Agricultural Practice for the Protection of Waters” [the Nitrates Regulations (S.I. No. 31 of 2014)] and “Waste Management (Use of Sewage Sludge in Agriculture) Regulations” (S.I. No. 148 of 1998). The application rate is typically determined by pH, metal and nutrient content of the soil, and the nutrient and metal content of the biosolids as per limits recommended in the “Codes of Good Practice for the Use of Biosolids in Agriculture” (Fehily Timoney Company 1999). Commonly, P is considered to be the limiting factor to estimate a legal application rate when applying biosolids and cattle slurry as organic fertiliser (Lucid et al. 2013). For example, P application rate of 40 kg ha⁻¹ annually is recommended for grassland in Ireland at a low Morgan’s P Index soil such as P index 1 (up to 3 mg-P L⁻¹ in soil) considering silage production where Morgan’s P provides the P index system in Ireland to describe soils with plant-available P (Teagasc Greenbook 2016). It was estimated that DPWTS application rate of 1 tonne DM ha⁻¹ year⁻¹ (assuming median P concentration in DPWTS) would supply approximately 40 kg total P ha⁻¹ without any harmful increase in concentrations of metals (chromium, copper, nickel, lead and zinc) in grassland pasture (Ashkuzzaman et al. 2019). However, the plant-available P from DPWTS is yet uncertain and it was observed to vary widely for different organic fertilisers (e.g. 100% for cattle slurry and 40% for biosolids) (Teagasc Greenbook 2016; Sullivan et al. 2015).

The risk of P runoff from land spreading of DPWTS can be associated with chronic (long-term, due to the build-up of nutrients in soil) and/or incidental (short-term within 48 h of application) losses due to episodic rainfall events (Brennan et al. 2012). Incidental losses (the focus of this study) occur when a rainfall event interacts shortly with inorganic and organic fertilisers spread on the land surface. The study with dairy cattle slurry and four types of treated biosolids showed that incidental losses of P, N and metals were highest in the first 24 h runoff after application compared to the losses in 48 and 360 h rainfall runoff (Peyton et al. 2016). The current legislation in Ireland restricts farmers not to apply slurry if heavy rain is forecasted within 48 h of application which

means the 24 h runoff losses represent a worst-case scenario. In terms of P loss, dissolved reactive phosphorus (DRP) was one of the dominant losses, while N losses were dominant by ammonium-N (NH₄-N) (Peyton et al. 2016). Such losses may pose a significant risk to water quality by causing ecological imbalance (e.g. depletion of oxygen, production of toxin from some harmful algal blooms) in water bodies (Ashkuzzaman and Jiang 2018). For example, 38% of the EU water bodies are at high risk from agricultural pollution (FAO 2017), and agriculture was the plausible cause in 53% of river pollution in Ireland in the period 2010–2012 (EPA 2015). In Ireland, the fourth Nitrates Action Programme (NAP), effective from 2018 to 2021, prioritized new measures to lessen nutrients (N, P) transport pathways to water bodies arising from farmland for preventing and reducing water pollution (DAFM 2017).

However, there are no data yet available on the potential runoff losses of nutrients (N, P), metals and carbon associated with land spreading of DPWTS. Although metals in DPWTS were found to be significantly lower than the EU limit for agricultural land application (Ashkuzzaman et al. 2019), their migration potential and concentrations from the surface applied soil to runoff need to be quantified to compare with water quality metal limits. The runoff of carbon from agricultural sources may become a potential threat of forming disinfectant by-products such as trihalomethanes (THM) and other known carcinogens in the public drinking water treatment facility when a fraction of total organic carbon reacts with added chlorine (Lentz and Lehrs 2014). Data pertaining to DPWTS, water and soil interactions at the micro scale are important to investigate nutrient, carbon and metal dynamics during this interaction. Losses from soil and an organic fertilizer to the overland flow pathway can be examined under laboratory controlled conditions using an agitator test developed by Mulqueen et al. (2004). In an agitator test, an intact soil core is placed in a beaker (with or without receiving surface applied DPWTS) and is overlain with continuously stirred water simulating overland flow. This simple method has already been used to investigate the mitigation of dissolved reactive phosphorus in overland flow after amendment of dairy cattle slurry (Brennan et al. 2011b), dairy soiled water (Fenton et al. 2011) or pig slurry (O’Flynn et al. 2012) with different chemical amendments.

Therefore the objectives of the present study were to use a laboratory agitator test to quantify losses of nutrients (N, P), carbon and metals in runoff and to assess any changes to plant-available P across treatments. Grassland soils representing well, moderate and poor drainage classes were selected and combined with DPWTS treatments in a replicated agitator test.



This research was carried out during the year 2017 at the Teagasc Environment Research Centre laboratory at Johnstown Castle, Co. Wexford in Ireland.

Materials and methods

Soil selection and characterisation

For the agitator tests, intact topsoil cores ($n = 48$) representing three target drainage classes were identified using diagnostic features of the soil profile to 1 m and then collected using the soil map (1:25,000) for the Johnstown Castle Research Farms (latitude 52° 17'N, longitude 6° 29'W) in the southeast of Ireland. A random 1 m² area was selected within the designated fields and a composite topsoil sample (2 kg) was taken, oven-dried at 40 °C for 72 h and crushed to pass a 2-mm sieve.

The soils used in the present study were characterised in terms of their physical and chemical attributes (see Table 1). Particle size analysis using the pipette method was conducted on these samples and the corresponding sand–silt–clay% and textural classes were documented. The remaining composite soil sample was used for chemical characterisation. Plant-available P is measured with Morgan’s P (P_m) extracting solution (Morgan 1941), which provides the P index system in Ireland to describe a soils plant-available P status and to estimate a legal P application rate when applying biosolids and cattle slurry as organic fertiliser (Teagasc Greenbook 2016; Brennan et al. 2011a). The simple water-extractable P (WEP) test was conducted on samples using 100:1 deionized water-to-soil solution according to Kleinman et al. (2007). This test is a good indicator of dissolved reactive P to be lost during runoff. Another parameter often used to estimate risk of P losses from soils is the degree of P saturation (DPS). It is calculated as follows using Eq. 1 (O’Flynn et al. 2018; Maguire and Sims 2002):

$$DPS(\%) = \frac{M3P \times 100}{M3Al + M3Fe} \tag{1}$$

where Mehlich-3 (M3) extracting solution (Mehlich 1984) was used to analyse M3-P, M3-Al, M3-Fe and M3-Ca (mmol kg⁻¹).

Also, total concentrations of P, Al, Fe, Ca and trace metals (arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn)) in soil were analysed using the microwave-assisted acid digestion method (USEPA 1996). Soil pH was determined using a pH probe (Jenway 3510 pH meter) and a 2.5:1 ratio of deionized water-to-soil. Soil organic matter (OM) was determined by loss on ignition using B.S. 1377-3 method (BSI 1990).

Table 1 Generic soil physical and chemical properties from well, moderate and poor drained sites

Soil	pH	OM (%)	TOC (%)	WEP (mg/kg)	DPS ^d (%)	P index	P_m (mg/L)	M3P ^c (mg/kg)	M3Al ^e (mg/kg)	M3Fe ^e (mg/kg)	M3Ca ^e (mg/kg)	P ^s (mg/kg)	Al ^e (mg/kg)	Fe ^e (mg/kg)	Ca ^e (mg/kg)	Sand (%)	Silt (%)	Clay (%)	Texture
S01	6.6	9.3	3.6	24	11.5	4	11.8	116	724	315	1707	1169	12,028	14,230	3107	65.3	20.2	14.5	Sandy loam
S02	6.1	8.9	3.1	5	2	1	2.5	18.7	571	490	1153	525	14,102	13,333	1643	47 ^b	36 ^b	18 ^b	Sandy silt loam ^b
S03	6.3	6.7	2.2	10	6.4	2	3.5	53.8	732	313	1178	814	16,945	17,650	1823	55	28.9	16.1	Sandy loam

^aMorgan’s extractable phosphorus

^bPeyton et al. (2016)

^cMehlich-3 extractable phosphorus, aluminium and iron

^dDegree of phosphorus saturation

^eTotal content



In summary from Table 1, the generic physical and chemical properties of the three mineral soils chosen were as follows: Soil-01 (S01) was a well-drained sandy loam (65.3% sand, 20.2% silt, 14.5% clay) with a soil test P (STP) of 11.7 mg L⁻¹ (P Index 4), soil-02 (S02) was poorly drained sandy silt loam (47% sand, 36% silt, 18% clay) with STP of 2.5 mg L⁻¹ (P Index 1), and soil-03 (S03) was moderately drained sandy loam (55% sand, 28.9% silt, 16.1% clay) with STP of 3.5 mg L⁻¹ (P Index 2). All three soils were within optimal pH ranges (6.1 to 6.6) for grassland mineral soils i.e. ~6.2 (Teagasc Greenbook 2016).

Dairy processing wastewater treatment sludge (DPWTS)

Three types of DPWTS samples were targeted in this study representing the main DPWTS applied to land in Ireland. Each DPWTS type was collected from a different processing plant: (1) Ca–P-rich lime-treated sludge (Ca-DPWTS), (2) Al-rich activated sludge (Al-DPWTS), and (3) Fe-rich activated sludge (Fe-DPWTS), which are representative of the major DPWTS only recently characterised by Ashekuzzaman et al. (2019). The activated sludge is generated after dairy food processing wastewater being treated by aeration and a biological flocculation with alum or ferric salt (aluminium or iron flocculent) dosing. On the other hand, lime-treated Ca–P-rich sludge is generated after dissolved air floatation (DAF) technique of treating dairy wastewater rich in fats, oils and greases. The samples (~2 kg) were collected in plastic containers with screw top lids and transferred to the Teagasc, Environmental Research Centre, Johnstown Castle for analysis. The samples were stored in a cold room at 4 °C and subsequently prepared for dry matter (DM), OM, pH, nutrients and metal analysis following standard analytical methods. For example, the DM and OM were determined using the standard gravimetric method 2540 G (APHA 2005). Sludge pH was determined in a 1:2.5 (w/v) ratio of fresh sludge to deionised water solution (making up to 25 mL) by Jenway 3510 pH meter after 1 h of mixing at 20 rpm by an end-to-end shaker. The concentrations of nutrients [P: 20.0 to 74.8 g kg⁻¹ DM, K: 6.1 to 11.6 g kg⁻¹ DM, magnesium (Mg), sulphur (S), sodium (Na), and calcium (Ca)] and metals [arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), aluminium (Al), iron (Fe), cobalt (Co), molybdenum (Mo) and manganese (Mn)] were determined by an Agilent 5100 synchronous vertical dual-view inductively coupled plasma optical emission spectrometer (Agilent 5100 ICP-OES) following the microwave-assisted acid digestion of freeze dried powder samples (USEPA 1996). Also, freeze dried samples were analysed for total carbon [TC, ranged from 27.6 to 37.2% of DM)] and total nitrogen (N, ranged from

Table 2 Characterization of dairy processing sludge

Source	Nutrient and other elemental composition (g kg ⁻¹ DM)																
	DM (% wt.)	OM (% DM)	TC (% DM)	pH	N	P	K	NH ₄ -N	TON	WEP	Ca	Al	Fe	Mg	Mn	Na	S
Ca-DPWTS	22.3 (0.3)	55.7 (0.2)	27.6	6.2	44.9	74.8 (0.5)	6.1 (0.08)	0.7 (0)	<0.1	1.3 (0.02)	142.0 (0.24)	0.5 (0.01)	0.3 (0)	5.1 (0.06)	0.02 (0)	5.4 (0.01)	1.9 (0.04)
Al-DPWTS	13.1 (0.2)	79.7 (0.4)	37.2	6.8	62.6	20.0 (0.1)	6.9 (0.04)	1.5 (0)	<0.1	1.2 (0.04)	36.6 (0.03)	10.5 (0.3)	1.44 (0.04)	2.63 (0.04)	0.04 (0)	5.0 (0)	4.0 (0.05)
Fe-DPWTS	18.4 (0.03)	60.6 (0.1)	29.4	7.1	59.7	35.7 (0.13)	11.6 (0.3)	4.9 (0.03)	<0.1	0.1 (0)	60.3 (0.25)	0.27 (0)	94.4 (0.07)	2.6 (0.02)	0.17 (0)	3.7 (0.02)	3.1 (0.03)

Values presented as average values (± standard deviation) for different parameters
 TON total oxidized nitrogen, DPWTS dairy processing wastewater treatment sludge, Ca-DPWTS calcium-phosphorus-rich lime-treated sludge, Al-DPWTS aluminium-rich activated sludge, Fe-DPWTS iron-rich activated sludge

44.9 to 62.6 g kg⁻¹ DM) by the high-temperature combustion method using a LECO TruSpec CN analyser. A full summary DPWTS characterisation can be seen from Table 2 whereby the DPWTS composition reflects the processing stream it originates from. For example, Ca-DPWTS is high in Ca due to lime treatment, Al-DPWTS is high in Al due to the use of alum-based coagulant within the wastewater treatment process, and the same is true for Fe-DPWTS as this is high in Fe. An important consideration to note is that these DPWTS waste streams change over time as technologies within the processing plant could negate or minimise the use of coagulants for achievement of discharge licence targets e.g. biological removal of P. In terms of P content, the Ca-DPWTS is the highest, but the availability of this P to the plant versus the environment is unknown. In terms of heavy metals, all DPWTS are below EU regulatory limits (supplementary Table S2).

Agitator test

Such a test was introduced by Mulqueen et al. (2004) and has been used by others to examine runoff losses after a single application of organic fertilizer e.g. dairy cattle slurry and municipal wastewater treatment plant-derived biosolids (i.e. treated sewage sludge) (e.g. Lucid et al. 2013; O'Flynn et al. 2012; Fenton et al. 2011). In an agitator test, organic fertiliser like DPWTS can be applied on the surface of the soil with the provision to dry, interact with soil, and then subjected to overland flow (Fig. 1). This test simulates the interaction between applied DPWTS and soil, and the effect of infiltration and skin formation on the release of nutrients, carbon and metals to surface runoff. So, runoff concentrations presented here represent edge of field-losses and may not represent final concentrations entering a surface waterbody.

Within the validated 1 m² areas representing well, moderate and poorly drained soils 16 intact soil cores were extracted using aluminium rings (50-mm high and 80-mm diameter). The soil cores were inserted into 1-L capacity glass beakers. Wax was then heated and poured in the gap

to form a seal. The soil core depth of 40–50-mm in the beakers was considered sufficient to include the full depth of influence on release of P to runoff (Mulqueen et al. 2004). The beakers were kept at room temperature for 1 day before starting the agitator test experiment. The application rate of DPWTS samples to soil was 1 ton DM ha⁻¹, which was considered to be within the safe limit of sludge application to agricultural land by the EU regulation (EC 2001). This application rate provided a P application rate of between 20 and 74.8 kg ha⁻¹ (Table 3).

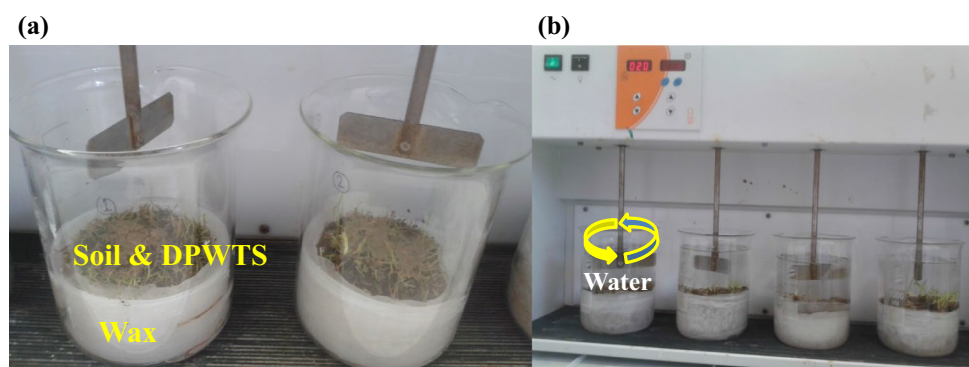
In the present study, the agitator test comprised 12 treatments in triplicate, where a single test incorporated four treatments—a grassed soil only (study control, e.g. S01) and grassed soil receiving three DPWTS samples, e.g. S01 + Ca-DPWTS, S01 + Al-DPWTS, and S01 + Fe-DPWTS. Thus the treatments included the interactions among 3-soil × 3-DPWTS samples to investigate potential losses of nutrients, C and metals in runoff but also enabled the accumulation in soil to be examined. At *t* = 0 h the DPWTS treatments were applied to the soil surface with a spatula and were left to stand and dry for 24 h. The treatments were then saturated by gently adding deionized water to the soil sample intermittently until ponding occurred implying saturated conditions (over 24 h). The present experiment examined incidental losses 48 h after application which mimics the current regulations pertaining to rainfall forecasts. After saturation

Table 3 Application rates of three dairy processing sludge samples

	Ca-DPWTS	Al-DPWTS	Fe-DPWTS
Application rate (t DM/ha)	1	1	1
Fresh mass (g)	2.25	3.84	2.73
DM (g)	0.5	0.5	0.5
Treatment surface area (m ²)	0.005	0.005	0.005
P application rate (kg/ha)	74.8	20	35.6

Ca-DPWTS calcium–phosphorus-rich lime-treated sludge, *Al-DPWTS* aluminium-rich activated sludge; *Fe-DPWTS* iron-rich activated sludge. *DPWTS* dairy processing wastewater treatment sludge

Fig. 1 The agitator test set-up **a** 1 L capacity glass beakers with DPWTS added soil cores and gaps sealed by wax and **b** set-up of single test run including four treatments in the flocculaor



($t = 48$ h), 500 mL of deionized water was added to the beaker to create a contact between soil, DPR and runoff. To simulate movement, the agitator paddle was lowered to mid-depth in the overlying water and rotated at 20 rpm for 24 h. Thus, nine agitator test runs were conducted to assess the interactions among 3-soil \times 3-DPWTS samples including 3 replications for each treatment, where each test run included four treatments (Fig. 1).

Water samples (4 mL) were taken from mid-depth of the runoff water overlying the soil at 0.25, 0.5, 1, 2, 4, 8 and 24 h after 48 h of each test (i.e. after the 500 mL was added). All samples were filtered immediately after sample collection using 0.45- μ m filters and kept at 4 °C for 1–2 days prior to being analysed colorimetrically (APHA 2005) for dissolved reactive phosphorus (DRP) and mineral nitrogen [total oxidized nitrogen (TON), nitrite nitrogen ($\text{NO}_2\text{-N}$), ammonium nitrogen ($\text{NH}_4\text{-N}$)] using a Aquakem 600 Discrete Analyser. The concentration of nitrate-nitrogen ($\text{NO}_3\text{-N}$) was calculated by subtracting the concentration of $\text{NO}_2\text{-N}$ from TON. After 24 h of simulated runoff, water samples were also analysed for total phosphorus (TP), total nitrogen (TN), total carbon (TC), total organic carbon (TOC), and heavy metals such

as Cd, Cr, Cu, Ni, Pb and Zn. These parameters were determined according to the standard methods for the examination of waters and wastewater (APHA 2005). The DRP, $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ concentrations were used to calculate the mass of these parameters in the water overlying the soil samples in the beaker, taking into account the water volume reduction as the test progressed. Soil samples after 24 h of runoff study were also analysed for desired parameters as described in Sect. 2.1.

Statistical analysis

Significant differences (statistically significant when $p < 0.05$) in the treatment groups of three soils by three DPS interactions for desired parameters were assessed by one-way analysis of variance (ANOVA) and Kruskal–Wallis test, respectively, for normal and non-normal data. The Shapiro–Wilk test was used to assess the distribution of dataset for the measured soil parameters. Correlation analysis (Pearson's product–moment correlation for normal distributions and Spearman's rank order correlation for non-normal data) was performed to identify the association between different soil parameters.

Table 4 Soil parameters without (soil only)/with (soil+DPWTS) the addition of three DPWTS samples—A calcium–phosphorus-rich lime-treated sludge, B aluminium-rich activated sludge, C iron-rich activated sludge, respectively

	Effect on S01 soil	Effect on S02 soil	Effect on S03 soil
Parameters	S01/S01 + A/S01 + B/S01 + C	S02/S02 + A/S02 + B/S02 + C	S03/S03 + A/S03 + B/S03 + C
P_m (mg/L)	11.0 ^a /33.4 ^b /18.7 ^{ab} /16.4 ^{ab}	3.2 ^a /33.9 ^b /7.4 ^{ab} /6.1 ^{ab}	4.3 ^a /21.4 ^b /6.0 ^{ab} /6.0 ^{ab}
M3-P (mg/kg)	124 ^a /169 ^b /156 ^b /144 ^{ab}	33.3 ^a /84.8 ^b /48.0 ^{ab} /39.1 ^{ab}	54.3 ^a /86.6 ^b /64.8 ^{ab} /63.1 ^{ab}
M3-Al (mg/kg)	745/759/732/746	532/567/553/561	688/696/707/715
M3-Fe (mg/kg)	334/319/345/336	458/453/447/472	307/317/318/316
M3-Ca (mg/kg)	1788/1801/1896/1856	1275/1309/1326/1147	1221/1182/1259/1422
P (mg/kg)	1144 ^a /1253 ^{ab} /1221 ^{ab} /1425 ^b	707 ^{ab} /892 ^a /718 ^{ab} /656 ^b	850/988/903/881
Al (g/kg)	10.6/11.2/9.4/9.9	11.7 ^a /14.3 ^{ab} /12.4 ^{ab} /9.4 ^b	16.7 ^{ab} /16.9 ^{ab} /17.5 ^a /14.5 ^b
Fe (g/kg)	12.8/13.2/11.9/13.1	12.1/12.1/12.1/11.4	17.4/17.9/18.0/16.6
Ca (g/kg)	2.7/2.9/2.8/2.9	1.8/2.1/1.9/1.6	1.8/1.9/1.8/1.9
DPS (%)	11.9 ^a /16.1 ^b /15.1 ^{ab} /13.9 ^{ab}	3.9 ^a /9.4 ^b /5.4 ^{abc} /4.3 ^{ac}	5.7 ^a /8.9 ^b /6.6 ^{ab} /6.4 ^{ab}
WEP (mg/L)	0.29/0.39/0.35/0.36	0.19/0.28/0.22/0.16	0.12/0.16/0.14/0.14
TC (% DM)	5.5 ^a /5.0 ^a /6.4 ^b /5.0 ^a	5.9/5.1/6.2/6.0	3.3/3.1/3.3/3.2
TOC (% DM)	4.0/4.0/4.2/4.6	4.8/4.9/5.0/4.3	2.9/2.3/2.4/2.6
pH	6.7/6.6/6.7/6.6	6.2/6.2/6.2/6.2	6.3/6.2/6.3/6.3

Statistical significance is compared within same soil type and is indicated by different superscript letter, for non-significance: same or common or no superscript letter is present across the row (Kruskal–Wallis test with Dunn's post hoc was used to assess if addition of DPWTS samples had significant changes in the measured soil parameters across each soil category)

DPWTS dairy processing wastewater treatment sludge

Results and discussion

Results

Soil

The changes in the measured soil parameters across soil category, i.e. different drainage class and soil P index due to interaction with 3 DPWTS samples, are presented in Table 4. In terms of extraction methods, P_m showed a positive trend across DPWTS treatments and soil drainage classes, but a significant increase ($p < 0.01$) was only found for the Ca-DPWTS (A) amendment across all soil drainage classes (Table 4). The change in P_m went from ~ 11 to 33 (S01), ~ 3 to 34 (S02) and ~ 4 to 21 (S03) mg L^{-1} after Ca-DPWTS (A) application. For the Mehlich-3 extractable P (M3-P), the same trend was observed with significant increases observed for all soil types (S01–S02: $p < 0.01$, S03: $p < 0.05$) for the Ca-DPWTS (A) amendment. Another significant increase ($p < 0.05$) was for S01 in the Al-DPWTS (B) application increasing from 124 to 156 mg kg^{-1} (Table 4). There was a trend of total extractable P increases, but no general significant trend was found except significant increase in S01 from 1144 to 1425 mg kg^{-1} by Fe-DPWTS (C) application. For S02 (low P index soil), total P increase was positive for Ca-DPWTS (A) and Al-DPWTS (B) amendment but negative for Fe-DPWTS (C). This could point to no overall change in total P but instead a change in the P distribution during the test.

In addition, the amendment with three DPWTS samples generally changed the status of total and Mehlich-3 extractable Al, Fe and Ca concentrations across soil types with an increasing trend (not significant). The degree of P saturation (DPS in %) was increased overall across soils from sludge applications, but this was significant for Ca-DPWTS (A) amendment only (e.g. 11.9 to 16.1 in S01 ($p < 0.05$), 3.9 to 9.4 in S02 ($p < 0.01$) and 5.7 to 8.8 in S03 ($p < 0.05$)). This can be explained by the similar trend of significant increase in P_m and M3-P across the studied soils for Ca-DPWTS (A) application. The WEP was found to be strongly correlated with the increase in both P_m ($r_s = 0.732$, $p < 0.001$) and M3-P ($r_s = 0.754$, $p < 0.001$) across soils due to sludge application. An overall increasing trend (not significant) for WEP across soil types was observed due to the amendment with sludge (Table 4). This trend was highest for the Ca-DPWTS (A) application with an average 38% increase in WEP across soils followed by 18% and 8% increase for Al-DPWTS (B)

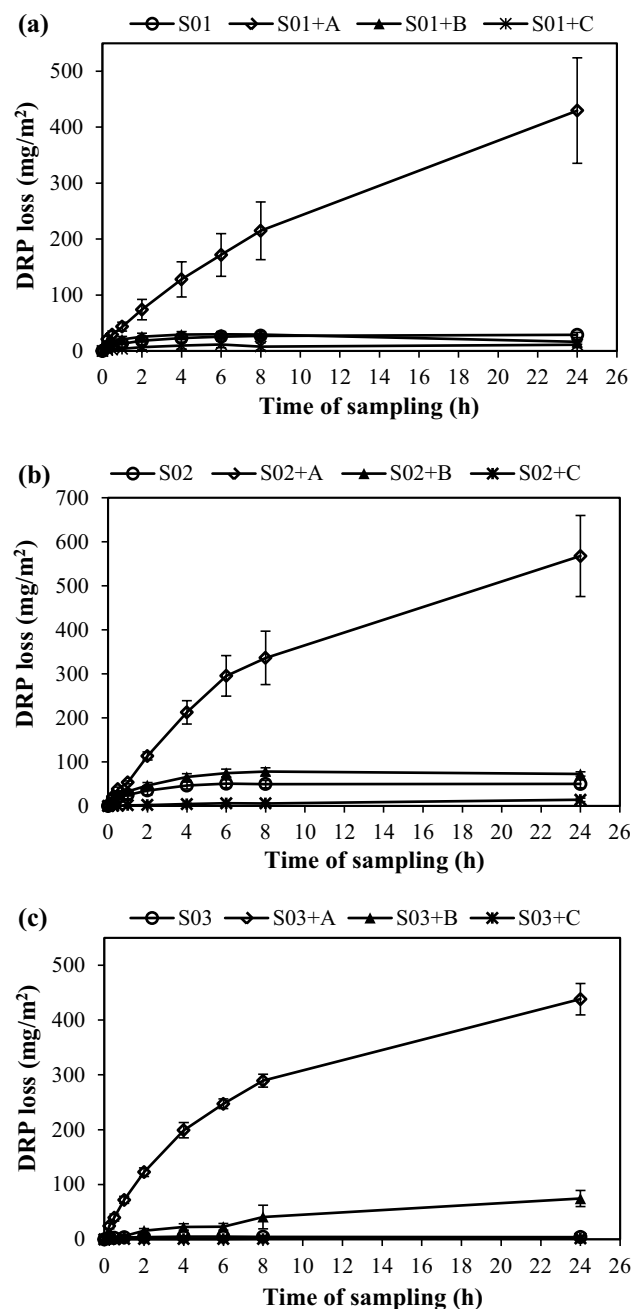


Fig. 2 Simulated runoff of DRP from the DPWTS and soil interaction **a** soil S01 only versus S01 receiving A, B, and C, respectively; **b** soil S02 only versus S02 receiving A, B, and C, respectively; and **c** soil S03 only versus S03 receiving A, B, and C, respectively. DRP: dissolved reactive phosphorus; DPWTS: dairy processing wastewater treatment sludge; A: calcium–phosphorus-rich lime-treated sludge; B: aluminium-rich activated sludge; C: iron-rich activated sludge



and Fe-DPWTS (C), respectively. For the TC, TOC and pH, the changes across soils from sludge amendments were not significant overall and remained quite similar to background condition.

In terms of potential runoff WEP loss risks, the increasing trends of P_m , M3-P and DPS across studied soils due to amendment by DPWTS may have an effect on the likely differences in DRP runoff concentrations that are likely to be controlled by the soils buffering capacity and ability to bind and store P. In terms of metal (As, Cd, Cr, Cu, Ni, Pb and Zn) concentrations in DPWTS amended soils, no significant increase found compared to soil-only concentrations across soil and sludge types and also, these concentrations are significantly lower in comparison with EU limit for metals in soil (supplementary Table S3).

Runoff losses

In terms of DRP losses in runoff, the Ca-DPWTS (A) sludge dominated runoff losses and the loss was significantly higher for Ca-DPWTS (A) across soil types in comparison with other sludge samples (Al-DPWTS and Fe-DPWTS) (Fig. 2). The losses ranged from 430 to 567 mg m^{-2} in the simulated overland flow after 24 h from Ca-DPWTS (A) amended soils, which is equivalent to 4.6 to 5.7 mg L^{-1} of DRP runoff loss. The DRP release from Ca-DPWTS (A) sludge across all soils showed a consistent increase from 0.21 mg L^{-1} at 15 min to about 5.7 mg L^{-1} over a 24 h of simulated overflow. On the contrary, the DRP release in the simulated overflow from 15 min to 24 h was between ~ 0.5 and 74 mg m^{-2} (equivalent to 0.005 to 0.8 mg L^{-1}) for the Al-DPWTS (B) and Fe-DPWTS (C) application across soils and these DRP concentrations were not statistically different (except Al-DPWTS (B) amendment for S03) than those measured from soil-only control (i.e. S01–S03). For Al-DPWTS (B) application in S03, the DRP concentration in the 24 h simulated runoff was 0.8 mg L^{-1} , which is significantly higher than that measured from S03 (0.05 mg L^{-1}). Similarly in terms of total P (TP) loss after 24 h, Ca-DPWTS (A) exhibited significantly higher release between 4 and 6 mg L^{-1} compared to the 0.2 to 1.3 mg L^{-1} across Al-DPWTS (B) and Fe-DPWTS (C) sludge-amended soil and soil-only control (e.g. 0.1 to 0.7 mg L^{-1} across S01–S03) in runoff, respectively (Fig. 4). The results pointed out that DAF (A) sludge was the most concerning in terms of P runoff potential during the rainfall event.

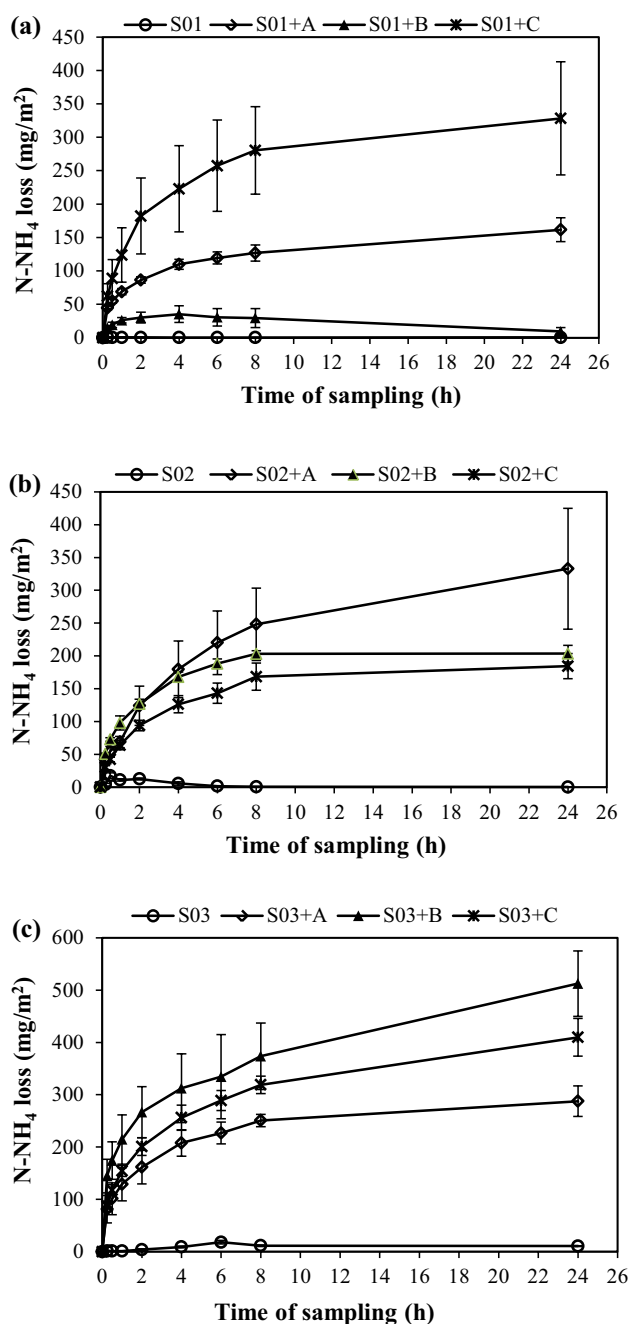


Fig. 3 $\text{NH}_4\text{-N}$ losses in the simulated runoff over a 24-h duration from DPWTS and soil interaction **a** soil S01 only versus S01 receiving A, B, and C, respectively; **b** soil S02 only versus S02 receiving A, B, and C, respectively; and **c** soil S03 only versus S03 receiving A, B, and C, respectively. DPWTS: dairy processing wastewater treatment sludge; A: calcium-phosphorus-rich lime-treated sludge; B: aluminum-rich activated sludge; C: iron-rich activated sludge

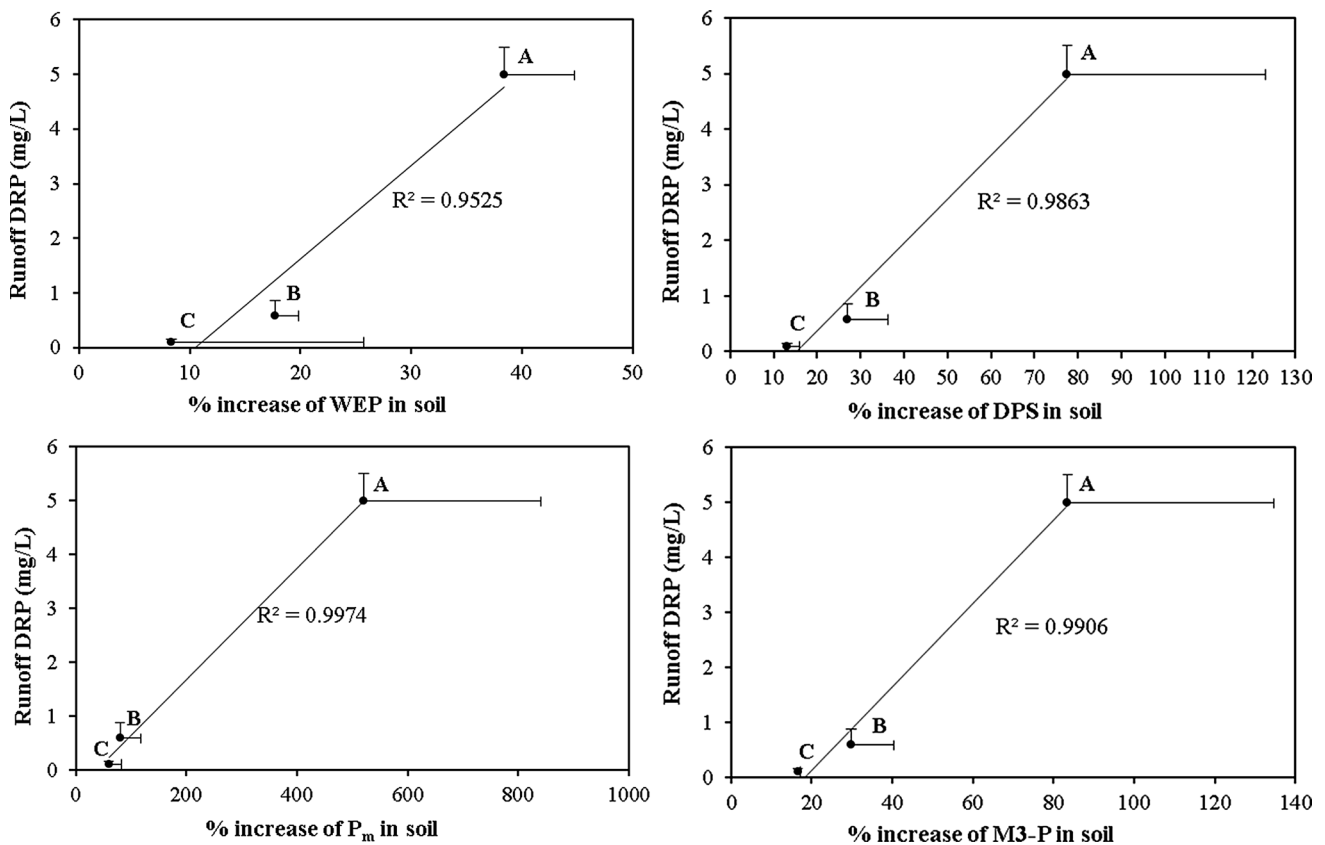


Fig. 4 Dissolved reactive P (DRP) losses in relation to dairy sludge (A: calcium–phosphorus-rich lime-treated sludge; B: aluminium-rich activated sludge; C: iron-rich activated sludge) application associated

increase in WEP, DPS, Morgan P (P_m) and Mehlich 3 extractable P (M3-P) in soils. The plotted points are average of studied soils with X, Y error bars indicating the standard deviation at each point ($n=9$)

In terms of NH_4-N losses in the simulated runoff from 15 min to 24 h, a consistent increase in NH_4-N concentration from ~ 30 to 512 mg m^{-2} (equivalent to 0.3 to 5.5 mg L^{-1}) was observed across all soils due to sludge amendments (except for Al-DPWTS (B) application in S01), and these concentrations were significantly higher than those of the study control (e.g. 0.001 to 0.2 mg L^{-1} across S01–S03) (Fig. 3). Al-DPWTS (B) sludge showed opposite NH_4-N loss pattern in S01 (well-drained) and S02 or S03 (poorly to moderately drained). In the 24 h simulated runoff, NH_4-N loss was overall significantly higher due to sludge amendment compared to soil-only control and this loss was highest for the Fe-DPWTS (C) (3.3 mg L^{-1}) followed by Ca-DPWTS (A) (2.7 mg L^{-1}) and Al-rich (B) (2.6 mg L^{-1}), respectively, considering the average loss across all soils. These concentrations exceed the EU drinking water standards for NH_4-N of 0.3 mg L^{-1} and indicate the contamination of water (S.I. No. 122 2014). In terms of protecting freshwater ecosystems, the trigger value of NH_4-N to cause aquatic toxicity starts from 0.2 mg L^{-1} at $\text{pH}=9.0$ to 2.8 mg

L^{-1} at $\text{pH}=6.0$ as per guidance provided in the “Australian and New Zealand Guidelines for Fresh and Marine Water Quality” (ANZECC and ARMCANZ 2000).

For NO_3-N loss, there is no trend over the duration of the simulated runoff study and this is indicative of biological transformation processes during the test (supplementary Figure S1). The concentrations of NO_3-N were in the range between 4 and 28 mg m^{-2} (equivalent to 0.04 to 0.3 mg L^{-1}) in the 24 h overlaying water across studied soil to sludge interactions and these concentrations are below the indicative guideline value of high ($<0.9 \text{ mg-N L}^{-1}$) and good ($<1.8 \text{ mg-N L}^{-1}$) quality river waters in Ireland, respectively (EPA 2018). The concentrations of total N (TN) (between 1.8 and 7.9 mg L^{-1}) in the runoff after 24 h supports that N loss was mainly dominated in the form of NH_4-N across sludge-amended soils (supplementary Figure S2).

In terms of TC and TOC loss, the concentrations in the 24 h overlaying water showed an overall increasing trend across soils due to sludge amendment (supplementary Figure S2). The range of losses for TC and TOC was between 22

and 63 mg L⁻¹ and 7 to 28 mg L⁻¹ across soil and sludge interactions, while the range for soil-only control was between 8 and 36 mg L⁻¹ and 3 to 20 mg L⁻¹, respectively. The average highest TC and TOC loss were observed from Fe-DPWTS (53.9, 21.5 mg L⁻¹) followed by Al-DPWTS (46.3, 17.9 mg L⁻¹) and Ca-DPWTS (41.2, 17.4 mg L⁻¹) applied soils. In particular, these concentrations were significantly higher for all sludge amendments in S03 than soil-only control, whereas for S01 and S02 the significance varied across sludge interactions. This might be related to the lowest OM and TOC content of S03 among the soil samples (Table 1) and consequently, less loss of these (i.e. TC, TOC) when associated with soil-only runoff (observed in the case of S03 compared to S01 and S02 associated TC and TOC loss, supplementary Figure S2). So, the loss of TC and TOC from S03 was mainly associated with sludge amendment while such significant loss was not observed from sludge-amended S01 and S02 soils.

For metal (As, Cd, Cr, Cu, Ni, Pb and Zn) concentrations in the simulated runoff after 24 h associated with sludge amendment to soils, no overall significant increase was found compared to soil-only concentrations and also, these concentrations were significantly lower in comparison with the WHO drinking water limit (supplementary Table S4).

Discussion

The results in this study are an indicative of potential incidental losses of phosphorus, nitrogen, carbon and metals

from DPWTS applied land surface. The quantification of such losses is important to assess perceived environmental damage like water quality deterioration. However, a number of factors like—quantity and type of bio-based fertiliser (e.g. biosolids, manure) applied, rainfall occurrence timing after fertiliser application, volume of runoff generated, antecedent hydrologic conditions and field position, flow path length, vegetation cover and surface slope potentially have influence on the magnitude of these losses (Brennan et al. 2012). Currently, mainly four types of DPWTS exist depending on the dairy wastewater treatment process, such as: (1) biochemically treated activated sludge, (2) lime [calcium oxide (CaO)]-treated dissolved air floatation (DAF) processing sludge, (3) combined treated (using both biological–chemical and DAF process) sludge, and (4) anaerobically digested sludge, which are compositionally rich in calcium–aluminium or iron (Ca–Al or Fe), Ca–phosphorus (Ca–P), Ca–Al, and Ca, respectively (Ashekuzzaman et al. 2019). In terms of major nutrient content in DPWTS, N ranged from 20 to 70, P ranged from 15 to 66, and K ranged from 3 to 7 g kg⁻¹ dry weight, respectively. The N and P concentrations in DPWTS are particularly higher than those generally observed in most of the other commonly used organic fertilisers. For example, median of N and P in DPWTS is 48.9 and 35.9 compared to 36.1 and 14.6 in biosolids and 38.1 and 7.9 g kg⁻¹ dry weight in cattle slurry, respectively (Ashekuzzaman et al. 2019). This suggests DPWTS could be an interesting feeding material to develop novel bio-based fertiliser through innovative scientific research due to its high N and P content.

Table 5 Comparison of potential DRP loss from different bio-based fertilisers

Bio-based fertilisers	TP (mg kg ⁻¹)	Application rate(kg P ha ⁻¹)	WEP (mg kg ⁻¹)	DRP loss (mg m ⁻²)	P loss potential (%)	References
Cattle slurry	11,264	40	2.6	650	16.3	Brennan et al. (2011b)
Al-WTR-1 (11.1% Al)	234	40	2.5	480	12.0	Brennan et al. (2011b)
Al-WTR-2 (5.3% Al)	18.7	40	1.1	680	17.0	Brennan et al. (2011b)
Dairy soiled water	6454	0.7	–	69	98.6	Fenton et al. (2011)
AD biosolids	6916	22.8	74	22	1.0	Lucid et al. (2013)
TD biosolids	7600	22.8	413	148	6.5	Lucid et al. (2013)
LS biosolids	6332	32.9	301	28	0.9	Lucid et al. (2013)
Ca-DPWTS (14% Ca)	74,800	74.8	1300	567	7.6	This study
Al-DPWTS (1% Al, 3.7% Ca)	20,000	20	1200	74	3.7	This study
Fe-DPWTS (9.5% Fe, 6% Ca)	35,700	35.6	100	11	0.3	This study

DRP dissolved reactive phosphorus, TP total phosphorus, WEP water-extractable phosphorus, Al-WTR alum-based water treatment residuals, AD anaerobically digested, TD thermally treated, LS lime stabilised, Ca-DPWTS calcium–phosphorus-rich lime-treated sludge, Al-DPWTS aluminium-rich activated sludge, Fe-DPWTS iron-rich activated sludge, DPWTS dairy processing wastewater treatment sludge



Yet land spreading of DPWTS is the most common practice across the EU for disposal management. The simple fact is that although technological solutions are available to avoid land application or at least minimise volumes, these methods may still result in a by-product that needs to be land spread or the technologies have not been successfully up-scaled to operational scales (Arvanitoyannis and Giakoundis 2007; Zhong et al. 2015; Slavoc 2017).

The DRP loss potential was found to be significantly different for different dairy processing sludge samples where the highest DRP runoff was observed for Ca-DPWTS (A) among the other two in this study. This was related to the significant increase in P_m , M3-P and degree of P saturation in Ca-DPWTS (A) applied soils. Figure 4 shows a strong positive association of DRP loss with the increase in WEP, DPS (degree of P saturation), P_m and M3-P, respectively. Also past studies showed that the soil test P (STP) determined by WEP, P_m , and M3-P have significant relationship with the runoff DRP (Regan et al. 2010). In the present study, P_m , M3-P and DPS were significantly increased in all 3 soil types for Ca-DPWTS (A) sludge application which resulted in significant DRP runoff ($\sim 5 \text{ mg L}^{-1}$). In comparison with Ca-DPWTS (A), DRP runoff from soil S01 (P index 4, $P_m = 11.8 \text{ mg L}^{-1}$ which is highly likely to release P and onto which P fertiliser application is prohibited) was only 0.2 and 0.1 mg L^{-1} , respectively, for Al-DPWTS (B) and Fe-DPWTS (C) sludge. Instead of DRP loss trend, a decrease of 43% and 61% of DRP from Al-DPWTS (B) and Fe-DPWTS (C) applied S01 soils, respectively, was observed when compared to S01 only DRP loss (0.3 mg L^{-1}). Soils with $P_m > 10 \text{ mg L}^{-1}$ are likely to lose DRP in excess of 0.03 mg L^{-1} which may lead to eutrophication (O'Flynn et al. 2018). The EU considers that lakes with total P concentration $< 0.01 \text{ mg L}^{-1}$ are not at risk of eutrophication, and rivers with total P concentration less than 0.01–0.07 mg L^{-1} are considered excellent waters (EC 2009). In Ireland, the water quality guideline of the EPA states that average phosphate concentrations of ≤ 0.025 and $\leq 0.035 \text{ mg L}^{-1}$ are required in rivers to meet the legal environmental quality standards of high and good ecological health of rivers (EPA 2018).

Clearly, dairy processing sludge samples of Al- and Fe-rich type were found to be much less prone to DRP loss in comparison with Ca-P-rich sludge. In particular, Fe-DPWTS with a P application rate of 35.6 kg ha^{-1} did not increase the runoff DRP across low to high STP (e.g. $P_m = 3$ to 11 mg L^{-1}) although DRP loss is very likely from high STP soils, instead 55–72% of potential DRP loss was

avoided due to Fe-rich DPWTS application. When comparing the potential DRP losses from dairy sludge samples including Ca-P-rich DPWTS to cattle slurry and dairy soiled water or some alum-based water treatment residuals (Al-WTR), the indicative losses in the present study are much lower (Table 5). The content of Ca, Al and Fe in dairy sludge is likely to contribute in retaining P, meaning no further chemical amendments are required to control P loss when DPWTS is applied to land, whereas such amendments have been explored to control P loss from cattle slurry and found to be expensive (e.g. Brennan et al. 2011a;b). Biosolids of different types like thermally treated (TD), lime stabilised (LS) and anaerobically digested (AD) were also observed to pose much lower risk of DRP runoff compared to cattle slurry (Table 5). For example, median highest DRP runoff was quantified between 0.78 and 0.86 mg L^{-1} for biosolids and 7.0 mg L^{-1} for dairy cattle slurry, respectively (Peyton et al. 2016). For Al- and Fe-rich DPWTS in the present study, DRP losses were determined to be 0.18–0.79 mg L^{-1} and 0.02–0.15 mg L^{-1} , respectively, and for Ca-P-rich DPWTS 4.6–5.7 mg L^{-1} . Although the highest DRP loss of Ca-P-rich DPWTS could be related to the highest P application rate (74.8 kg ha^{-1}) in this study, this trend was not consistent for Al- and Fe-rich sludge. The findings reported in Elliott and O'Connor (2007) showed that the total P loading rate (kg P ha^{-1}) of different organic amendments such as manures and biosolids cannot reliably indicate the P concentrations in runoff. The P-source loading rate when adjusted with a factor that correlates better with dissolved P showed increase in DRP runoff (Elliott et al. 2005). The WEP has been reported to be an effective quantitative indicator of dissolved P losses (Elliott et al. 2005; Leytem et al. 2004). Likewise, the DRP runoff potential of DPWTS samples in the current investigation followed the order of WEP (higher to lower) which was Ca-P-rich > Al-rich > Fe-rich sludge.

The dairy sludge samples showed significant elevated level of $\text{NH}_4\text{-N}$ concentrations in the runoff compared to the study control, whereas $\text{NO}_3\text{-N}$ concentrations in the runoff were negligible considering good river water quality guideline value ($< 1.8 \text{ mg-N L}^{-1}$). The observed average values of $\text{NH}_4\text{-N}$ concentrations (2.6–3.3 mg L^{-1}) in this study are significantly lower than what would typically expect from dairy cattle slurry (17.4 mg L^{-1}). For different biosolids, Peyton et al. (2016) also observed N losses dominated by $\text{NH}_4\text{-N}$ runoff (e.g. 2.5–15.3 mg L^{-1}), while $\text{NO}_3\text{-N}$ loss was not significant relative to the control.

The implications of TC and TOC loss to surface water bodies are particularly important for drinking water

treatment facilities due to the risk of forming disinfection by-products like trihalomethanes (THMs). In Ireland, public water supplies (about 82%) are mainly sourced from surface waters which contain higher levels of organic matter compared to ground waters (EPA 2012). THMs are likely to form when chlorine (the most widely used disinfectant in Ireland) reacts with organic matter in water, and the maximum total concentration of THMs for drinking water has been limited to $100 \mu\text{g L}^{-1}$ (S.I. 278 of 2007). As a key indicator of forming THMs, the TOC concentration is routinely monitored at the drinking water treatment process and it was observed that $\text{TOC} > 4.0 \text{ mg L}^{-1}$ poses risk of exceeding THM concentration $100 \mu\text{g L}^{-1}$ (EPA 2012). The average TOC runoff potential from dairy sludge applied soils varied from 17.4 to 21.5 mg L^{-1} , which may pose significant organic carbon transfer potential to surface waters.

In terms of overall losses, metals are not of concern across treatment types, Fe-rich DPWTS has highest losses of $\text{NH}_4\text{-N}$ and C, whereas Ca-P-rich DPWTS has highest DRP losses but lowest losses of $\text{NH}_4\text{-N}$ and C. The Al-rich DPWTS seems to be somewhere in between for all parameters measured. In terms of changes in plant-available P, there were no negative effects in terms of DPWTS application across treatments, and in the case of Ca-rich DPWTS there was a significant increase in P_m during the experiment. Runoff from field trials which have received all three DPWTS types should now be investigated under low P_m soil conditions using a rainfall simulator or natural rainfall approaches. No single DPWTS provided a balanced agronomic-environmental footprint. Therefore at field scale all aspects of DPWTS should be investigated to form a holistic (soil, water, air and fertilizer equivalent value in terms of P and N and air) impression of DPWTS usage when compared with inorganic or indeed other organic alternatives. Only then can a farmer be given sound advice pertaining to the sustainable use of these industrial by-products converting them from a waste into an alternative organic fertilizer.

Conclusion

An agitator test is a quick method to elucidate nutrient, carbon and metal losses and changes to plant-available phosphorus in soils after a dairy processing sludge application. Results showed that each DPWTS had its own profile in terms of nutrient, carbon and metal losses to runoff. Furthermore, all potential losses to runoff were small when compared with dairy cattle slurry equivalents. In terms of overall losses, metals are not of concern across DPWTS types;

Fe-rich DPWTS has highest losses in terms of concentration of $\text{NH}_4\text{-N}$ and C, whereas Ca-P-rich DPWTS has highest DRP losses but lowest losses of $\text{NH}_4\text{-N}$ and C. In terms of plant-available P, there were no negative effects. To inform management decisions the next phase of testing should be brought to a field trial scale where additional parameter testing over longer periods of time could be incorporated e.g. fertilizer replacement value.

Acknowledgements This work has received funding from Enterprise Ireland and the Dairy Industry Partners in Ireland under Dairy Processing Technology Centre (DPTC) programme. Grant Agreement Number TC2014 0016. We would like to thank DPTC associated dairy industry partners - Kerry Group, Arrabawn Co-op, Carbery Foods, Tipperary Co-op, Lakeland Dairies, Dairygold, Glanbia and Aurivo for supplying samples. Thanks to Denis Brennan, Linda Moloney Finn, Carmel O'Connor and Brendan Healy from Teagasc Johnstown Castle Research Centre for help and advice during sample analysis. Also, we appreciate help from John Murphy and Matthias Bacher to identify different soil types and getting soil cores from the field.

Compliance with ethical standards

Conflict of interest The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

References

- ANZECC and ARMCANZ (2000) Australian and New Zealand guidelines for fresh and marine water quality, volume 2, chapter 8, page 8.3–161. Australian and New Zealand Environment and Conservation Council and Agriculture and Resource Management Council of Australia and New Zealand, Canberra. <https://www.waterquality.gov.au/sites/default/files/documents/anzecc-armcanz-2000-guidelines-vol2.pdf>. Accessed on 22 Aug 2019
- APHA (2005) Standard methods for the examination of water and wastewater, 21st edn. American Public Health Association, Washington
- Arvanitoyannis IS, Giakoundis A (2007) Current strategies for dairy waste management: a review. *Crit Rev Food Sci Nutr* 46:379–390
- Ashekuzzaman SM, Jiang J-Q (2018) Use of Ca- and Mg-type layered double hydroxide adsorbent to reduce phosphate concentration in secondary effluent of domestic wastewater treatment plant. *Desalin Water Treat* 127:64–70
- Ashekuzzaman SM, Forrestal P, Richards K, Fenton O (2019) Dairy industry derived wastewater treatment sludge: generation, type and characterization of nutrients and metals for agricultural reuse. *J Clean Prod* 230:1266–1275
- Brennan RB, Fenton O, Grant J, Healy MG (2011a) Impact of chemical amendment of dairy cattle slurry on phosphorus, suspended sediment and metal loss to runoff from a grassland soil. *Sci Total Environ* 409:5111–5118

- Brennan RB, Fenton O, Rodgers M, Healy MG (2011b) Evaluation of chemical amendments to control phosphorus losses from dairy slurry. *Soil Use Manag* 27:238–246
- Brennan RB, Healy MG, Grant J, Ibrahim TG, Fenton O (2012) Incidental phosphorus and nitrogen loss from grassland plots receiving chemically amended dairy cattle slurry. *Sci Total Environ* 441:132–140
- Britz TJ, Van Schalkwyk C, Hung YT (2006) Treatment of dairy processing waste waters. In: Wang LK, Hung YT, LoH H, Yapijakis C (eds) *Waste treatment in the food processing industry*. CRC Press, Taylor & Francis Group, Boca Raton, USA, pp 1–28
- BSI (British Standards Institution) (1990) British standard methods of test for soils for civil engineering purposes. Determination of particle size distribution. BS 1377. BSI, London
- DAFM (2017) Nitrates explanatory handbook for good agricultural practice for the protection of Waters Regulations 2018. Department of Agriculture, Food and the Marine, Wexford, Ireland. <https://www.agriculture.gov.ie/media/migration/ruralenvironment/environment/nitrates/2018Nitratesexplanatoryhandbook03042018.pdf>. Accessed 10 Dec 2019
- EC (European Commission) (2001) Disposal and recycling routes for sewage sludge. Part 2- Regulatory Report, European Commission, October 2001. http://ec.europa.eu/environment/archives/waste/sludge/pdf/sludge_disposal2.pdf. Accessed 18 Dec 2019
- EC (European Commission) (2009) Guidance Document on Eutrophication Assessment, Common Implementation Strategy for the Water Framework Directive (2000/60/EC), Guidance Document No 23, Technical Report
- Elliott HA, O'Connor GA (2007) Phosphorus management for sustainable biosolids recycling in the United States. *Soil Biol Biochem* 39:1318–1327
- Elliott HA, Brandt RC, O'Connor GA (2005) Runoff phosphorus losses from surface-applied biosolids. *J Environ Qual* 34:1632–1639
- EPA (Environmental Protection Agency) (2012) EPA drinking water guidance on disinfection by-products Advice Note No. 4. Version 2, Disinfection By-Products in Drinking Water. EPA, Wexford, Ireland. http://www.epa.ie/pubs/advice/drinkingwater/DrinkingWaterGuide4_v8.pdf. Accessed 17 Dec 2019
- EPA (Environmental Protection Agency) (2015) Water quality in Ireland Report 2010 to 2012. EPA, Wexford, Ireland. www.epa.ie/pubs/reports/water/waterqua/wqr20102012/. Accessed 24 Aug 2016
- EPA (Environmental Protection Agency) (2018) Water quality in 2016: an indicators report. Environmental Protection Agency, Ireland. <http://www.epa.ie/pubs/reports/water/waterqua/Water%20Quality%20in%202016%20An%20Indicators%20Report.pdf>. Accessed 22 Aug 2019
- FAO (2017) Water pollution from agriculture: a global review. The Food and Agriculture Organization of the United Nations Rome and the International Water Management Institute on behalf of the Water Land and Ecosystems research program Colombo. <http://www.fao.org/3/a-i7754e.pdf>. Accessed 10 Dec 2019
- Fehily Timoney and Company (1999) Codes of good practice for the use of biosolids in agriculture-guidelines for farmer. <http://www.housing.gov.ie/sites/default/files/migrated-files/en/Publications/Environment/Water/FileDownload%2C17228%2Cen.pdf>. Accessed Mar 14 2018
- Fenton O, Serrenho A, Healy MG (2011) Evaluation of amendments to control phosphorus losses in runoff from dairy-soiled water. *Water Air Soil Pollut* 222:185–194
- Kleinman P, Sullivan D, Wolf A, Brandt R, Dou Z, Elliott H, Kovar J, Leytem A, Maguire R, Moore P, Saporito L, Sharpley A, Shober A, Sims T, Toth J, Toor G, Zhang H, Zhang T (2007) Selection of a water-extractable phosphorus test for manures and biosolids as an indicator of runoff loss potential. *J Environ Qual* 36:1357–1367
- Lentz RD, Lehrsch GA (2014) Manure and fertilizer effects on carbon balance and organic and inorganic carbon losses for an irrigated corn field. *Soil Sci Soc Am J* 78:987–1002
- Leytem AB, Sims JT, Coale FJ (2004) Determination of phosphorus source coefficients for organic phosphorus sources: laboratory studies. *J Environ Qual* 33:380–388
- Lucid JD, Fenton O, Healy MG (2013) Estimation of maximum biosolids and meat and bone meal application to a low P index soil and a method to test for nutrient and metal losses. *Water Air Soil Pollut* 224:1464–1475
- Maguire RO, Sims JT (2002) Measuring agronomic and environmental soil phosphorus saturation and predicting phosphorus leaching with Mehlich 3. *Soil Sci Soc Am J* 66:2033–2039
- Mehlich A (1984) Mehlich 3 soil test extractant: a modification of Mehlich 2 extractant. *Commun Soil Sci Plant Anal* 15:1409–1416
- Morgan MF (1941) Chemical soil diagnosis by the universal soil testing system. Connecticut Agricultural Experiment Station 450. <https://portal.ct.gov/-/media/CAES/DOCUMENTS/Publications/Bulletins/B450pdf.pdf?la=en>. Accessed 18 Dec 2019
- Mulqueen J, Rodgers M, Scally P (2004) Phosphorus transfer from soil to surface waters. *Agric Water Manag* 68:91–105
- O'Flynn CJ, Fenton O, Healy MG (2012) Evaluation of amendments to control phosphorus losses in runoff from pig slurry applications to land. *Clean Soil Air Water* 40:164–170
- O'Flynn CJ, Fenton O, Wall D, Brennan RB, McLaughlin MJ, Healy MG (2018) Influence of soil phosphorus status, texture, pH and metal content on the efficacy of amendments to pig slurry in reducing phosphorus losses. *Soil Use Manag* 34:1–8
- Peyton DP, Healy MG, Fleming GTA, Grant J, Wall DP, Morrison L, Cormican M, Fenton O (2016) Nutrient, metal and microbial loss in surface runoff following treated sludge and dairy cattle slurry application to an Irish grassland soil. *Sci Total Environ* 541:218–229
- Regan JT, Rodgers M, Healy MG, Kirwan L, Fenton O (2010) Determining phosphorus and sediment release rates from five Irish tillage soils. *J Environ Qual* 39:1–8
- Ryan MP, Walsh G (2016) The characterisation of dairy waste and the potential of Whey for industrial fermentation. EPA Research Report, Environmental Protection Agency, Ireland
- S.I. No 148/1998 Statutory Instrument S.I. No 148 of 1998 (1998) Waste management (use of sewage sludge in agriculture) regulations. Available online (Accessed 03/02/2019): www.irishstatutebook.ie/eli/1998/si/148/made/en/print?q¼WasteþManagementþUseþofþSewageþSludgeþinþAgriculture%252529þRegulationþs¼Cp1998
- S.I. No. 278 (2007) European Communities (Drinking Water) (No. 2) Regulations 2007, page 22. <http://www.irishstatutebook.ie/eli/2007/si/278/made/en/pdf>. Accessed 18 Dec 2019
- S.I. No. 122 (2014) European Union (Drinking Water) Regulations 2014, page 24. <http://www.irishstatutebook.ie/eli/2014/si/122/made/en/pdf>. Accessed 22 Aug 2019
- S.I. No 31/2014 Statutory Instrument S.I. No 31 of 2014 (2014) European Union (Good agricultural practice for protection of waters) Regulations. www.agriculture.gov.ie/media/migration/ruralenvironment/environment/nitrates/SI31of2014290114.pdf. Accessed 03 Feb 2019



- Slavoc AK (2017) General characteristics and treatment possibilities of dairy wastewater—a review. *Food Technol Biotechnol* 55:14–28
- Sullivan DM, Cogger CG, Bary AI (2015) Fertilising with biosolids. A Pacific North west Extension Publication, Oregon State University, Washington State University, University of Idaho. www.catalog.extension.oregonstate.edu/. Accessed 03 July 2017
- Teagasc Greenbook (2016) Major and micro nutrient advice for productive agricultural crops. In: Johnstown Castle Wall DP, Plunkett M (eds) (Wexford: Teagasc; Environment Research Centre), p 81
- USEPA (1996) SW-846 Test Method 3052: Microwave Assisted Acid Digestion of Siliceous and Organically Based Matrices. United States Environmental Protection Agency
- Zhong J, Stevens DK, Hansen CL (2015) Optimization of anaerobic hydrogen and methane production from dairy processing waste using a two-stage digestion in induced bed reactors (IBR). *Int J Hydrog Energy* 40:15470–15476

